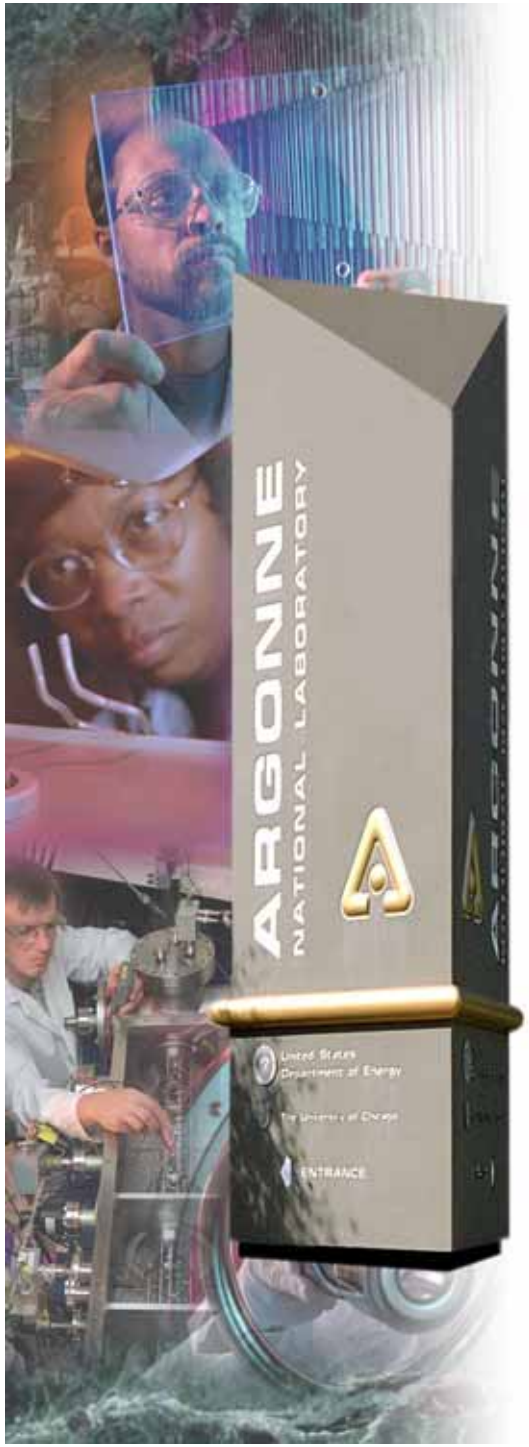

Tutorial 1: Advanced Fuels

M.K. Meyer
July 19, 2004

Big 10 Nuclear Engineering Workshop: Enabling our Energy Security through the Nuclear Fuel Cycle

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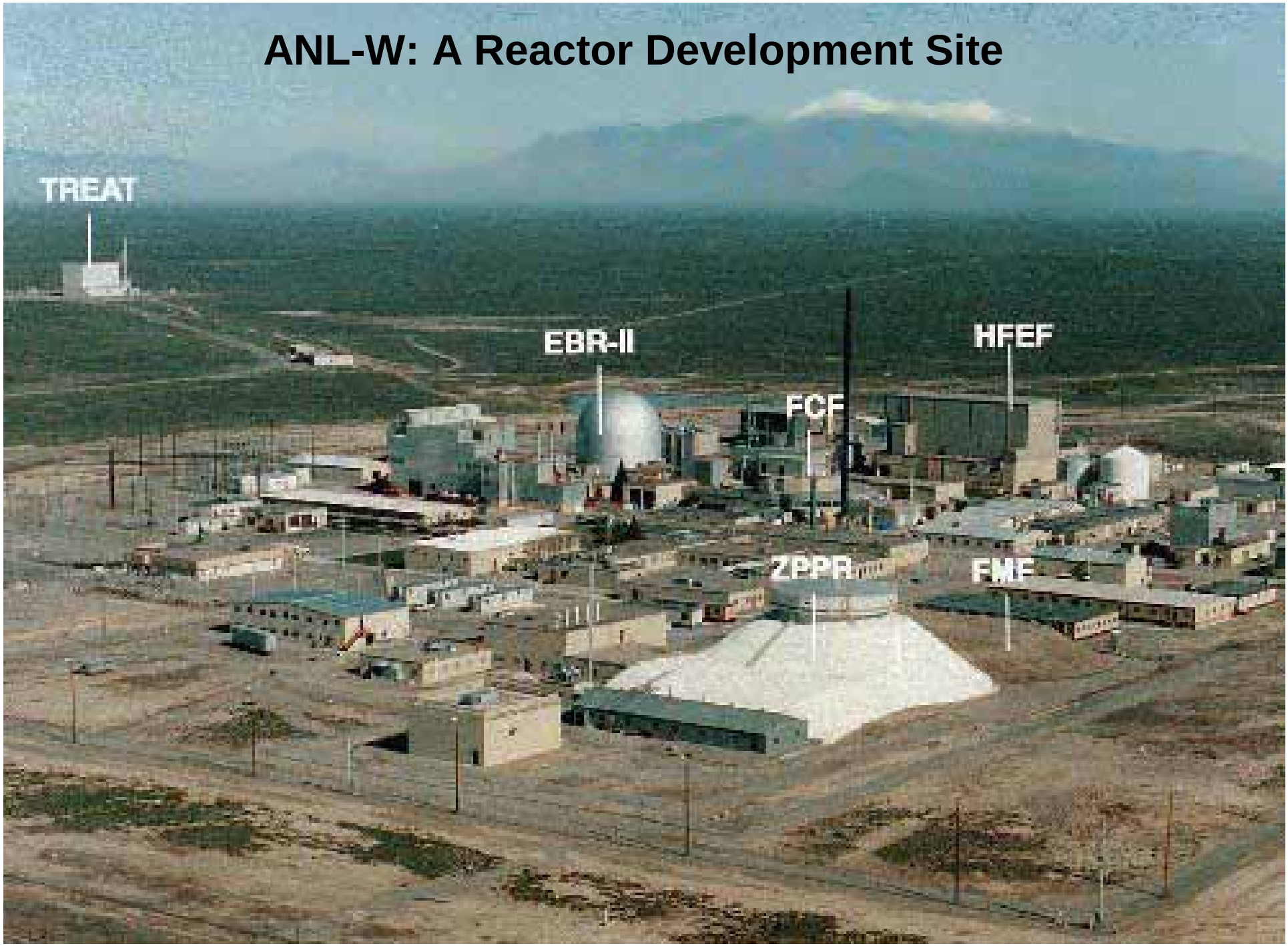
ANL-W: A Reactor Development Site



Drivers for New Fuel Development (1)

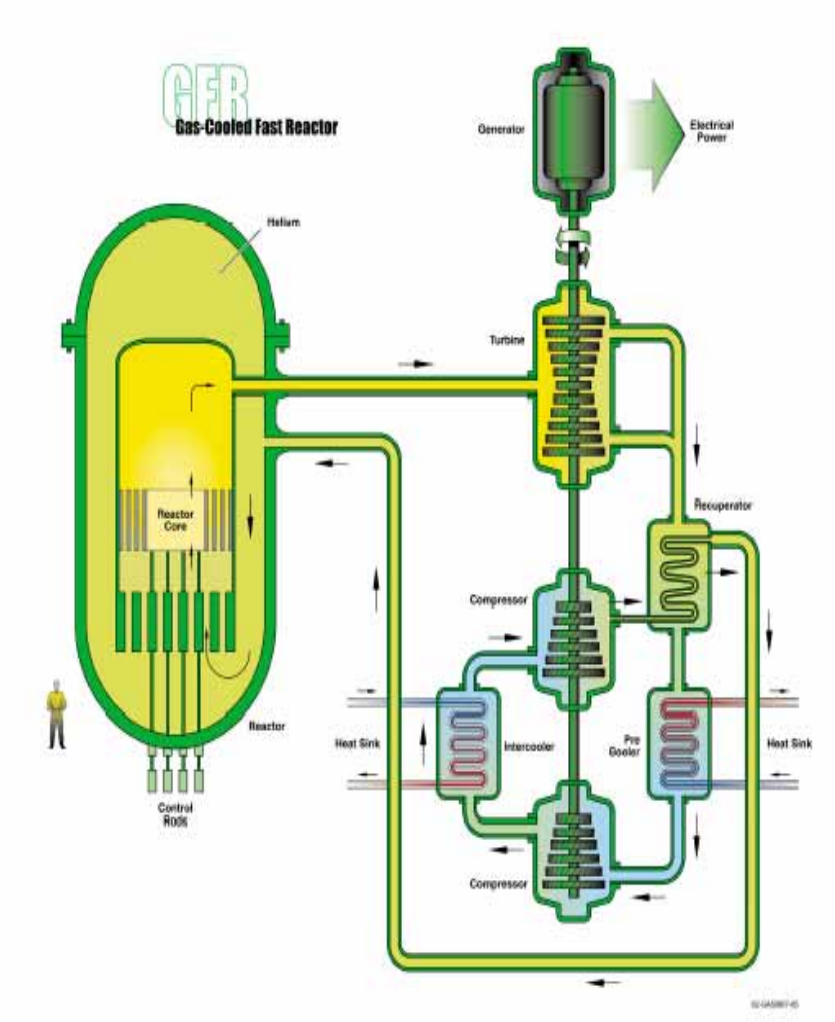
- Advanced Fuel Cycle Initiative
  - Driver – repository benefit
    - Heat load
    - Radiotoxicity
  - Fuels for TRU burning in LWRs
    - Inert Matrix Fuel (IMF)
    - Uranium matrix fuel
  - Fuels for TRU burning in Fast Spectrum Systems
    - Inert Matrix Fuel (IMF) for ADS
    - Uranium matrix fuel for Fast Reactors
- Non-Proliferation
  - Reduced Enrichment for Research and Test Reactors (RERTR)
    - LEU driver fuel for all reactors
    - Fission product Mo-99 targets

Drivers for New Fuel Development (2)

- Generation IV systems
  - Gas-cooled Fast Reactor (GFR)
  - Lead Fast Reactor (LFR, SMR)
  - Sodium Fast Reactor (SFR)
  - Very High Temperature Reactor (VHTR/NGNP)
  - Supercritical Water-cooled Reactor (SCWR)
  - Molten Salt Reactor (MSR)
- Generation IV Goals
  - Safety
  - Sustainability
  - Economics
  - Proliferation Resistance
- Advances in fuel technology play a critical role in achieving 3/4 goals for 5/6 systems.

Example 1: Fuel Development for the Gen IV Gas-cooled Fast Reactor (GFR)

Background: Gas-cooled Fast Reactor



Sustainability

- Conversion ratio ~ 1.0
- Actinide recycle

Economics

- High-temperature, direct cycle
- High efficiency (~48%)
- Hydrogen generation

Proliferation resistance

- Closed fuel cycle, no separation of U, Pu, MA

Safety and Reliability

- Dependent on robust fuels and materials to withstand accident conditions

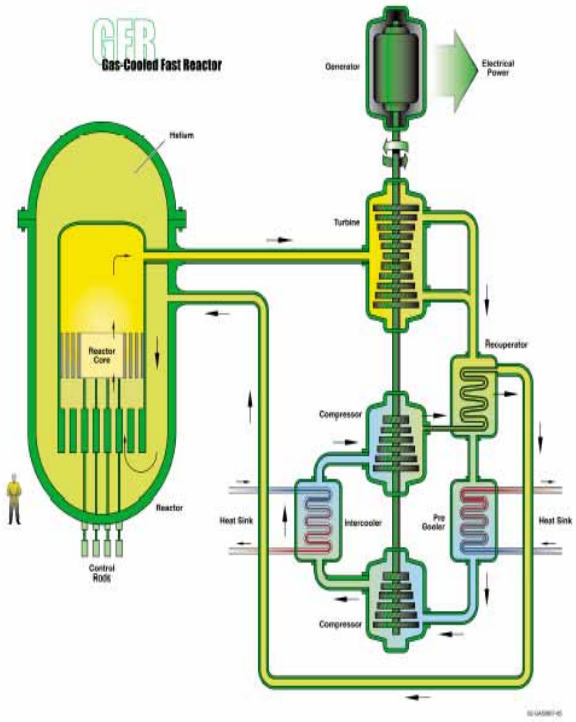
GFR Core Behavior

- GFR core
  - High fuel density, high power density relative to thermal spectrum gas-cooled reactors (10X)
    - High decay heat
  - Large coolant volume, lack of moderator
    - Low core heat capacity
- Behavior during unprotected loss of coolant much different than for VHTR
  - Adiabatic temperature rise at 7% decay heat
    - GT-MHR - 0.2 K/s
    - GFR (50 MW/m^3) – 2.5 K/s
    - GFR (100 MW/m^3) – 5.2 K/s (312 K/minute)
  - Peak fuel temperatures may exceed 1650°C during some scenarios
- Design goal: no core restructuring
  - Accident behavior drives fuel design

GFR Fuel Matrix Requirements

- Goal: Fuel that will withstand GFR 'blowdown'
- Several factors selected for screening fuel

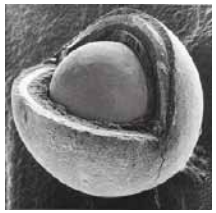
Requirement	Reference Value
Melting/decomposition temperature	**2000°C**
Radiation induced swelling	**<2% over service life (~80 dpa)**
Fracture toughness	**>12 MPa $m^{1/2}$**
Thermal conductivity	**>10 W/m·K**
Neutronic	**Materials allow low core HM density and safety parameters**



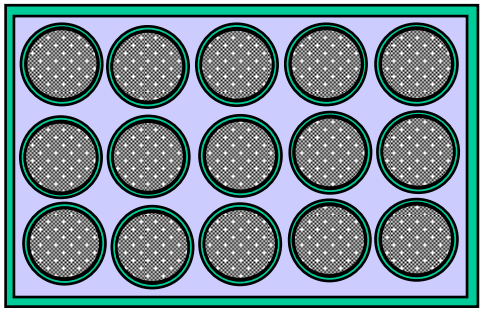
GFR Fuel

- **Pin-type fuel**
  - **No cladding material available that meets GFR requirements**
    - **SiC is possible exception, early stage of development**
    - **Nb-1Zr marginally acceptable neutronically**

- **TRISO fuel technology can't be used**
  - **Fuel density 5-10X lower than required**
  - **Poor irradiation behavior of pyrocarbon at high dose**



- **Refractory matrix dispersion fuels appear to be best candidates for satisfying Gen IV objectives**
  - **Cermets (ceramic fuel in metal matrix)**
  - **Cercer (ceramic fuel in ceramic matrix)**
  - **No data pertaining to GFR conditions**



Matrix Thermal Requirements

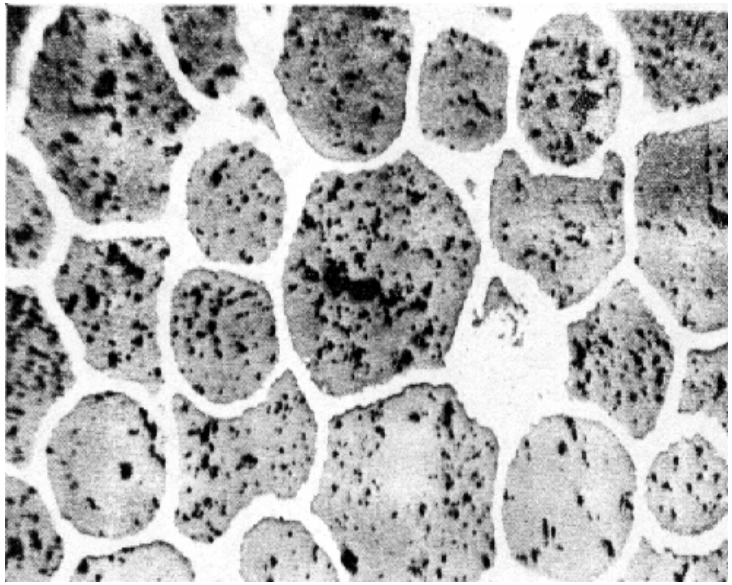
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn

Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

- Elements that meet the requirement for T_m > 2000°C in green
- Several others T_m > 1800°C in light gray

UO_2 Cermet Dispersion

- Nb, Mo matrices
- 80 vol% UO_2 loading
- UO_2 85 ~ 90 % theoretical density
- BOL temperature 1480°C
- Burnup 4+% initial HM
- 1.4% density decrease on irradiation



Postirradiation metallography of 80 vol% UO_2 Nb cermet (Keller, 1963)

GFR Matrix Requirements

H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn

Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu

- All elemental materials can be ruled out on the basis of neutronic criteria, melting temperature, and irradiation performance

Ceramic Matrices for GFR

- Disqualification of metals and metalloids
  - Low melting temperature
  - Poor neutronic properties
  - Inadequate irradiation stability

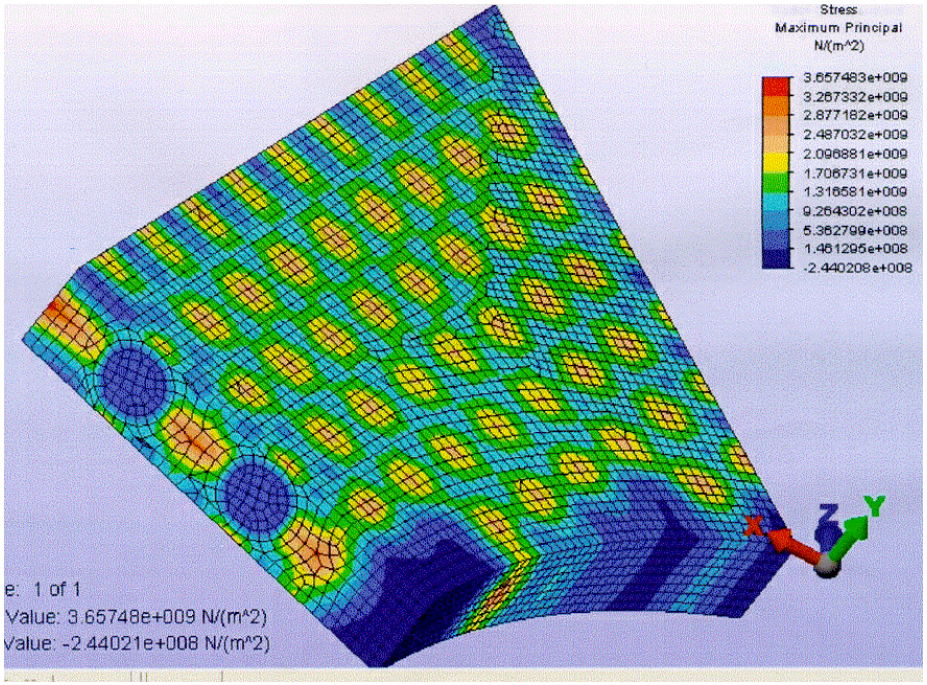
....leads to consideration of ceramics

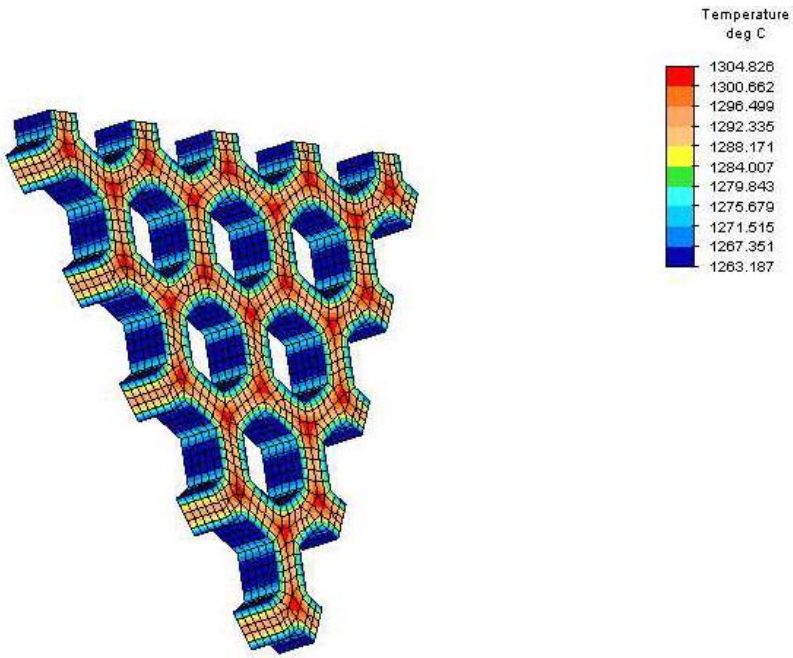
- **Thermal conductivity requirement eliminates most oxides**
- **Irradiation behavior eliminates others**
- **Fracture toughness remains an issue**
- **Ion and reactor irradiations to assess irradiation behavior**

Oxides	Carbides	Nitrides
Al_2O_3	SiC	**AlN**
CeO_2	TiC	**CeN**
$MgAl_2O_4$	**VC**	**Si_3N_4**
MgO	**YC_2**	TiN
Y_2O_3	ZrC	**YN**
ZrO_2		ZrN

Fuel Modeling

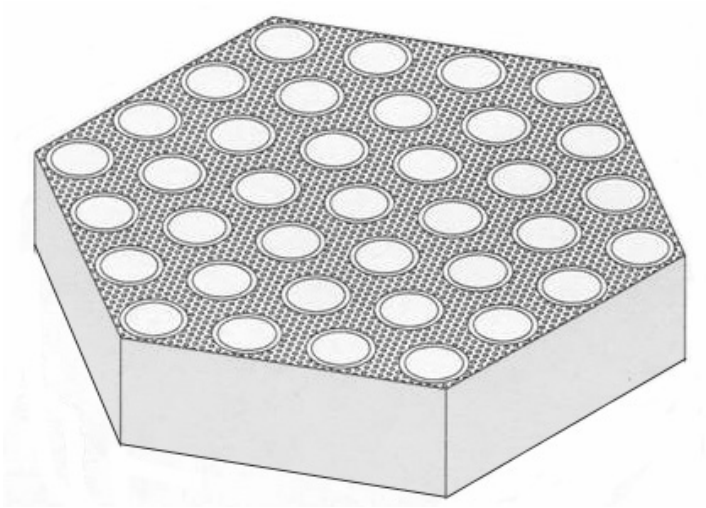
- Finite element analysis of temperature and thermal stress in GFR fuel used in design process
- Low modulus buffer required between particles and matrix

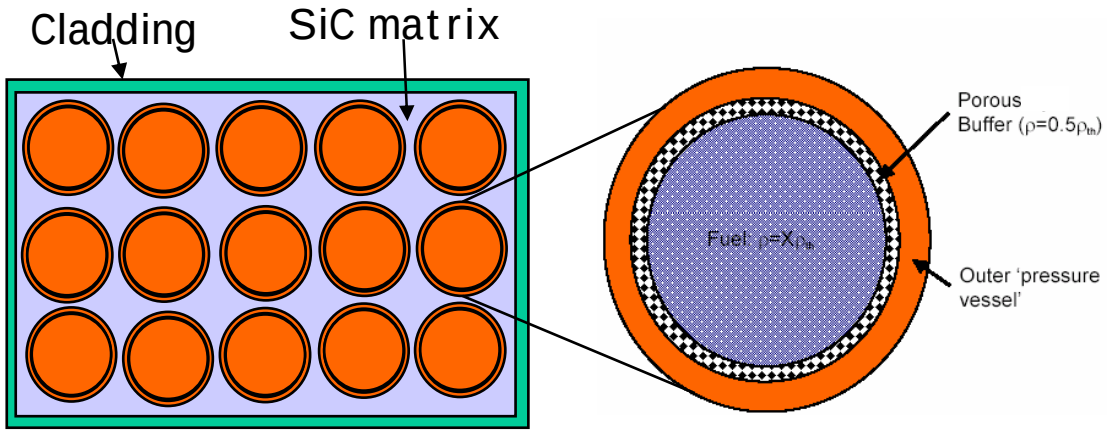




GFR Fuel Design

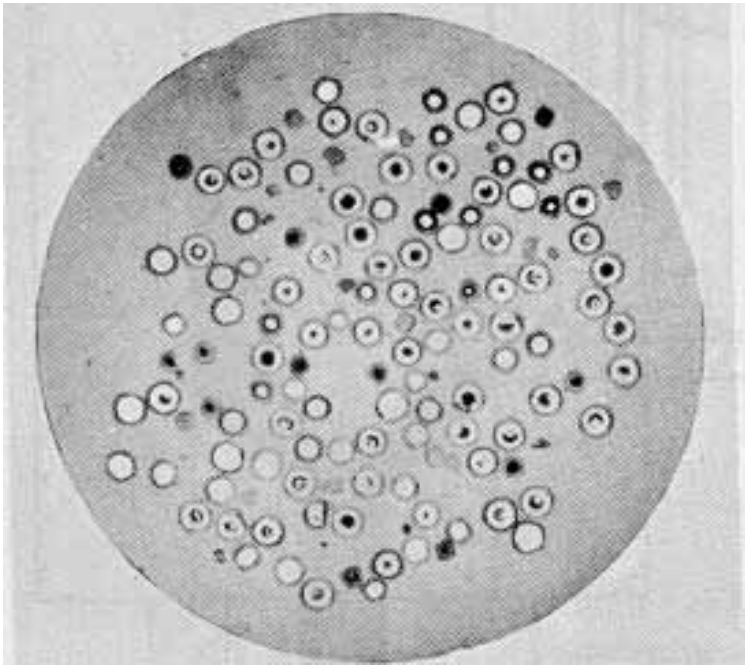
- **Stacked plates**
- **(U,Pu)C fuel particles (0.3 – 0.8 mm diameter)**
- **SiC bi-layer particle coating**
- **SiC matrix**
- **Outer cladding (as yet undefined)**





UC Dispersion in SiC

- Potential fuel for AGR program in the U.K.
- Coated (BISO) particles in reaction bonded SiC matrix
- Two irradiations in U.K.
  - 5% HM burnup
  - Power to 39 kW/m
  - 750° - 1200°C
- No matrix damage
- Xe, Kr R/B ~ 10^{-6}



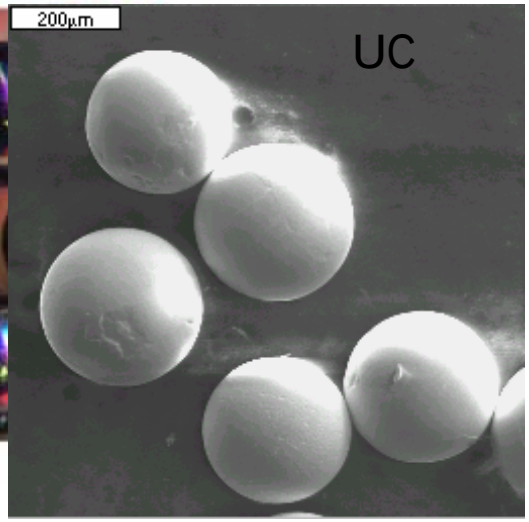
Shennan, 1967

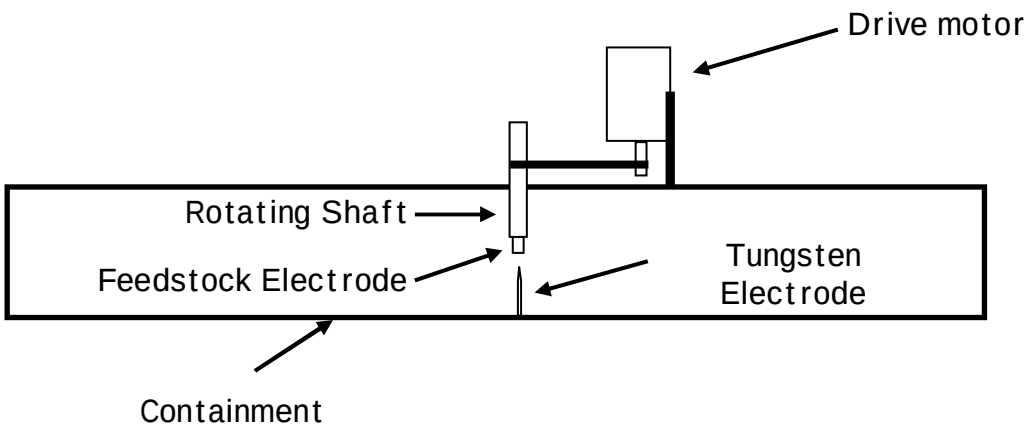
Fuel Fabrication: Particles

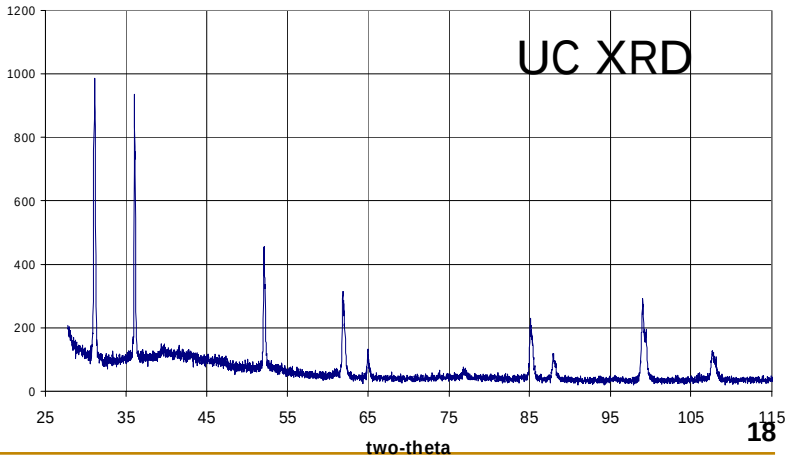
- Fuel fabrication required for reactor testing
  - Particles, particle coating, matrix consolidation
- Several routes possible for fabrication of 300-800 µm diameter spherical carbide particles
  - Sol-gel
    - ➢ Synthesis of oxide precursors followed by conversion to carbide
  - Powder processing
    - ➢ Agglomeration of fine carbide powder into green spheres by rolling
    - ➢ Sintering to desired density
  - Grinding
    - ➢ Irregular dense particles ground to spherical shape and size between two flat platens
  - Melt processing (atomization)

Fuel Fabrication: Particles

- Goal: simple process that lends itself to remotization
- Atomization used to produce UC microspheres
- Particle coating
  - ORNL: chemical vapor deposition
  - ANL: powder coating







Potential Fuel Fabrication Methods

- Sintering
  - Fuel particles act to inhibit densification
  - SiC & ZrC at 2000°C, no consolidation
- Hot Pressing
  - Dense microstructure, good creep resistance
  - High capital and production cost, not easily adapted to closed fuel cycle
  - Process will likely crack coated particles
- Polymer conversion/infiltration/reaction bonding
  - Used for many years to fabricate SiC
  - Recent refinements
  - Can be used for fabricating ZrC

Matrix Fabrication

Ethylene Glycols

Furfuryl alcohol + Furfuryl resin

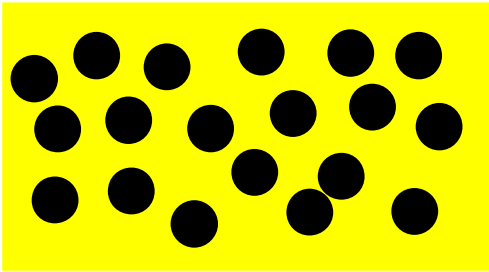
Fuel Particles

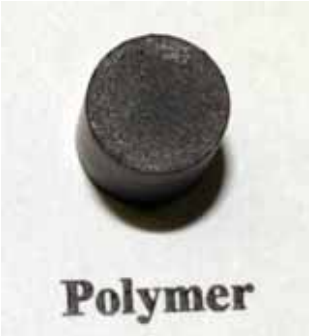
Mixing + Casting

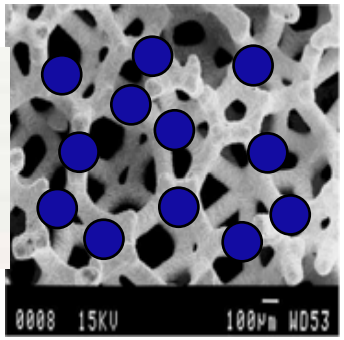
Polymerization (Catalyst)

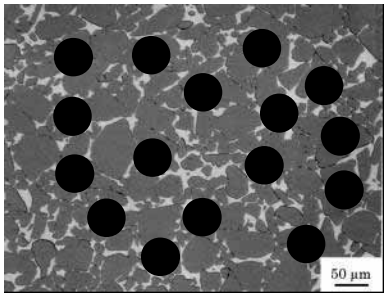
Pyrolysis to Carbon Foam (1000°C)

Metal Infiltration and Reaction

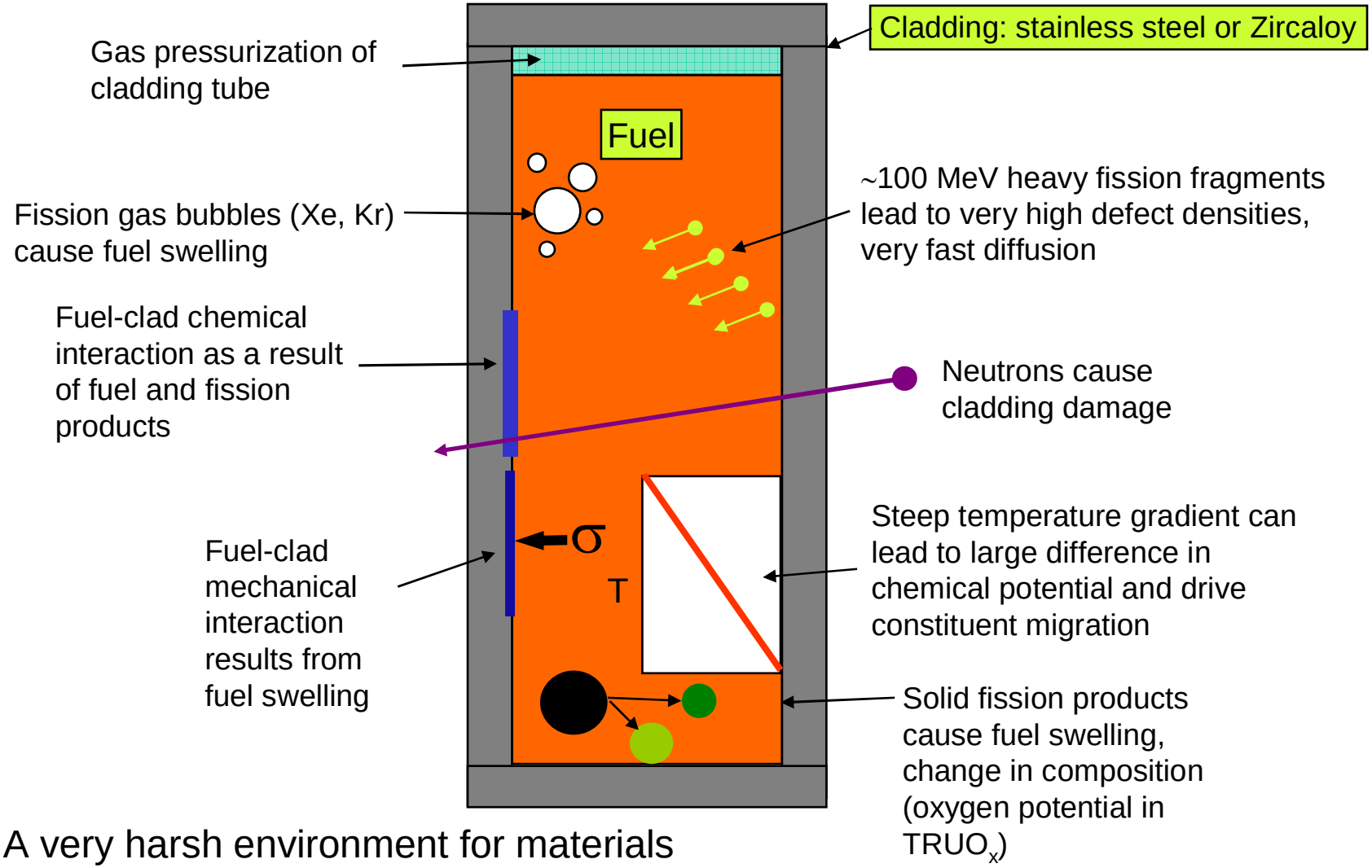








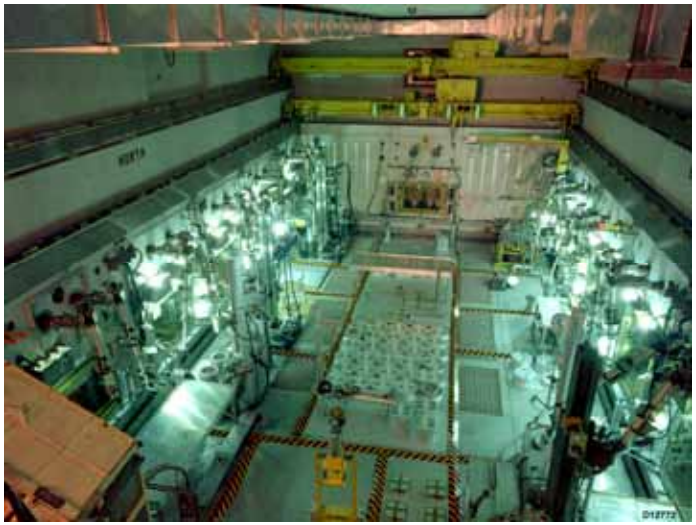
Conditions inside of a fuel rod



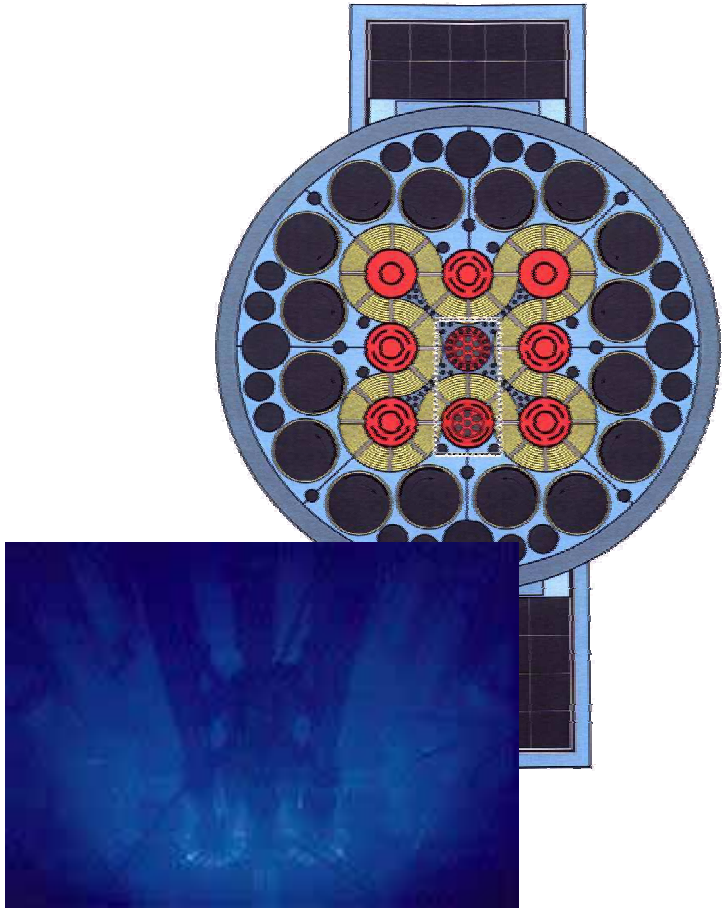
- A very harsh environment for materials

Fuel Irradiation Testing

- Cycles of fuel irradiation testing and postirradiation examination are a necessary part of fuel development



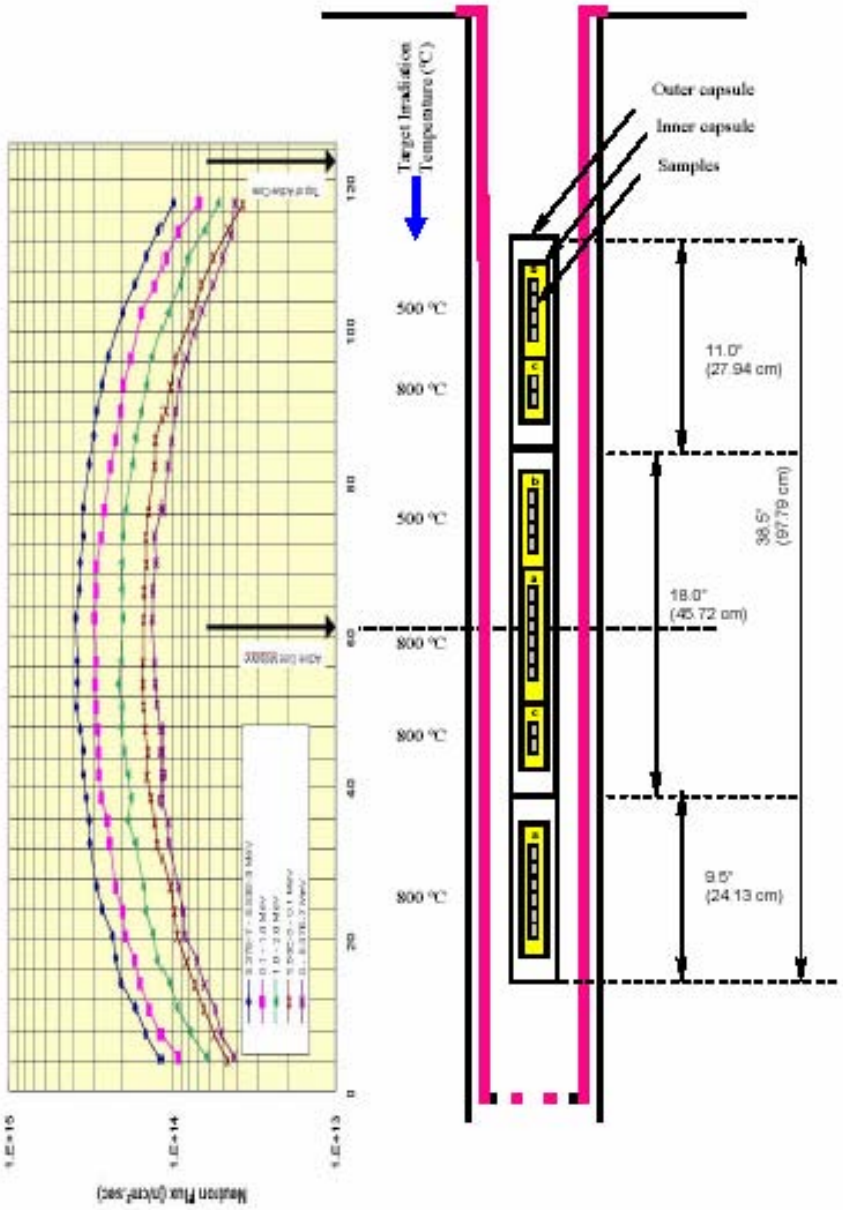
HFEF hot cell



Advanced Test Reactor

GFR Fuel Irradiation Testing in ATR

- In the case of GFR fuel, a simple screening test is appropriate to provide basic viability information on fuel behavior
- Test based on existing AFC-1 hardware design
  - Fuel specimens in HT-9 alloy cladding tubes
  - Outer 316L capsule
  - Cd filtered thermal spectrum (Cd also serves as dosimetry)
  - No active temperature control or monitoring



Example 2: Inert Matrix Fuel for LWR transmutation

Advanced Fuel Cycle Initiative

- Mission:
  - Develop and demonstrate technologies that enable transition to a stable, long-term, environmentally, economically and politically acceptable advanced fuel cycle
- Goals:
  - Reduce the quantity and radiotoxicity of high-level nuclear waste bound for geologic disposal
  - Enable more effective use of the currently proposed geologic repository and reduce the cost of geologic disposal.
  - Reduce the inventories of civilian plutonium in the U.S.
  - Enable recovery of the energy value from commercial spent nuclear fuel

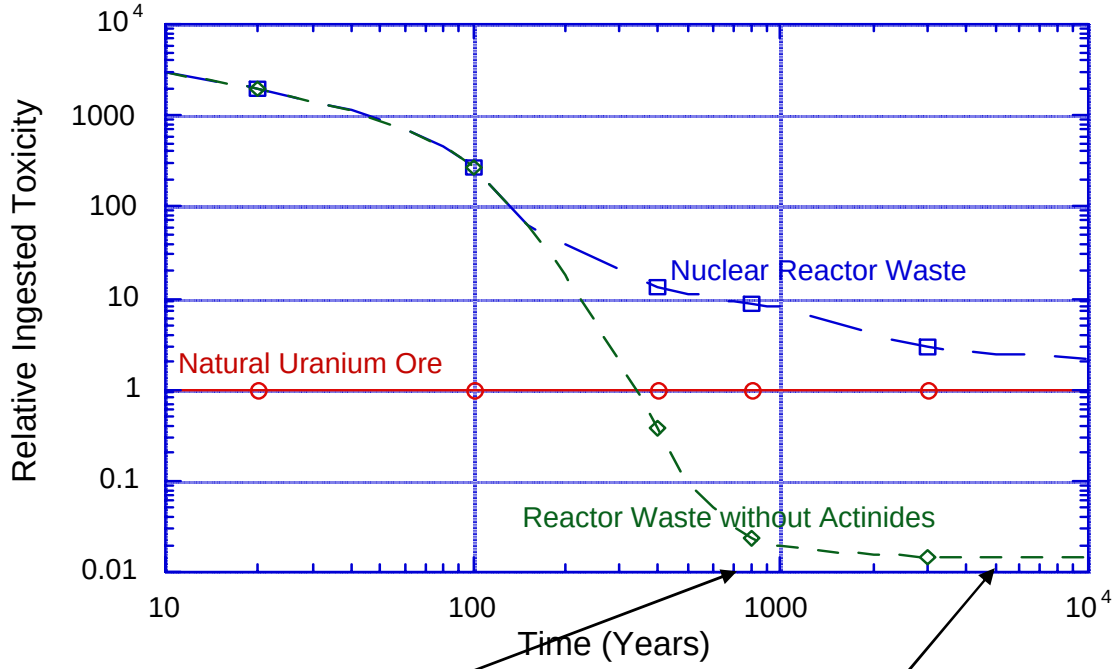
January 2003

Report to Congress
on
Advanced Fuel Cycle Initiative:
The Future Path for Advanced
Spent Fuel Treatment and
Transmutation Research

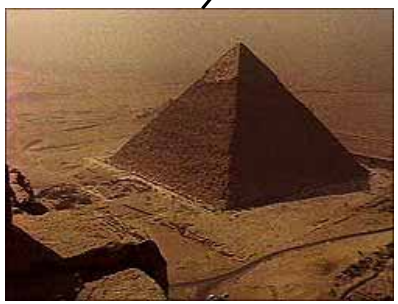
Prepared by
U.S. Department of Energy
Office of Nuclear Energy,
Science, and Technology
January 2003

http://www.nuclear.gov/AFCI_RptCong2003.pdf

Nuclear Waste Timeline



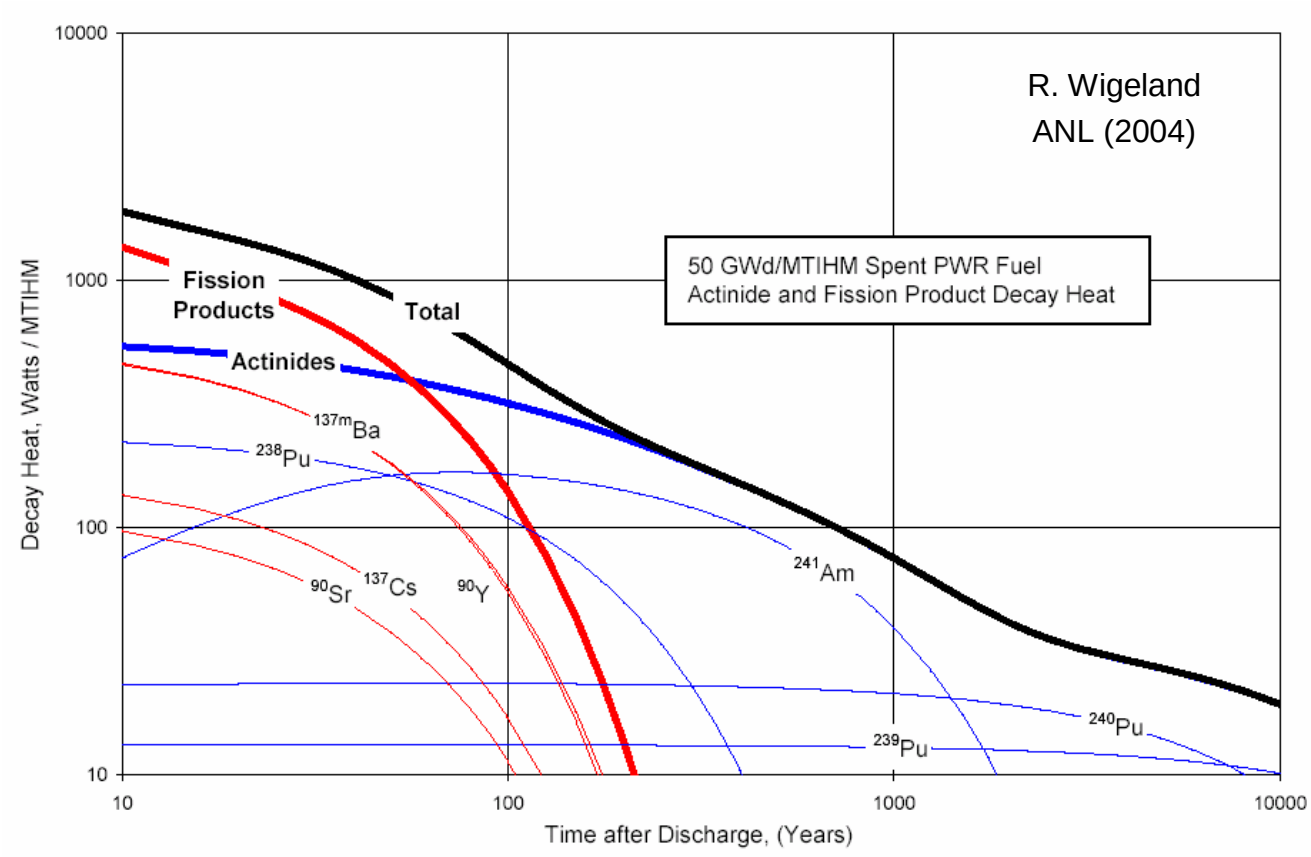
Notre Dame



Pyramids at Giza

- Plutonium and minor actinides are responsible for most of repository hazard beyond ~600 years
- ^{237}Np from ^{241}Am decay is long-term driver

Repository Heat Load



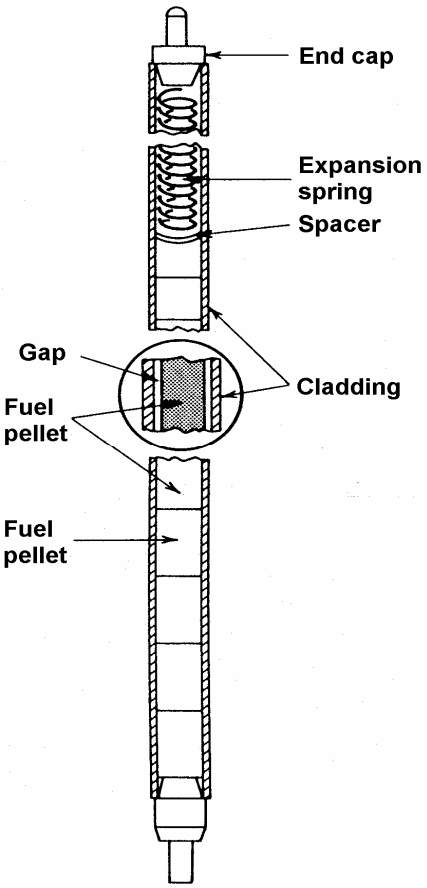
- After about 80 years, actinides are major contributors to total decay heat
- ^{241}Am, ^{238}Pu, and ^{240}Pu are largest contributors

MA Transmutation

- Fast Reactors and closed fuel cycle are optimum for repository benefit (Gen IV or dedicated burner) ~ 40X

- LWRs – Use of currently available infrastructure
  - Inert Matrix Fuels (no additional breeding)
    - ~ 1.8X decrease in net heat load in one pass
  - Uranium-matrix fuels
    - ~ 1.8X decrease in net heat load in 5-7 'MOX' passes
  - Emplacement of TRU in fuel from last cycle dominates heat load
  - Continuous recycle to keep MA out of repository ~ 20X

Operator Acceptance of Inert Matrix Fuel



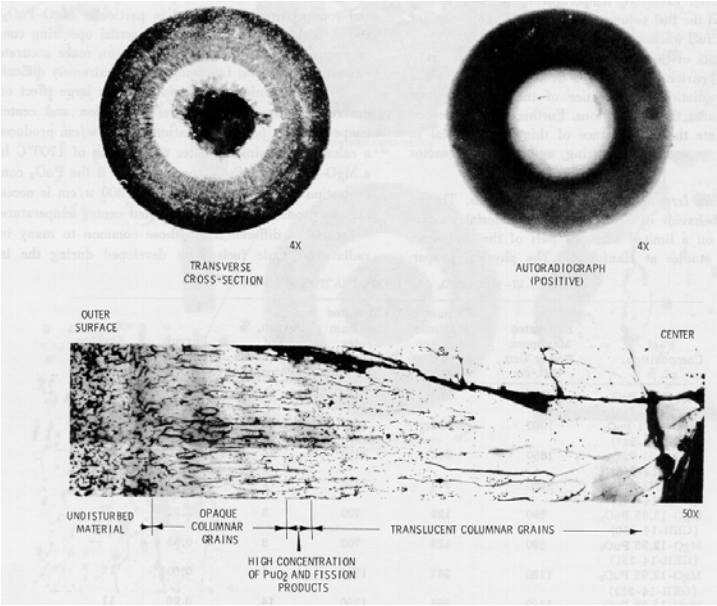
- Deployment of any new fuel in the U.S. LWR fleet depends on operator acceptance
  - Acceptance is unlikely without significant economic or performance advantages
- Goal – Inert Matrix Fuel for Pu disposition that is as close as possible to a direct replacement for UO_2 pellet fuels
  - Looks like UO_2 (initially)
  - Behaves like UO_2 (or better)
    - Thermal conductivity/ fuel temperature
    - Burnup limits
    - Response to power transients
    - Response to clad failure

Candidates for IMF Recycle Scenarios

- Al_2O_3 (poor neutron irradiation behavior at high dpa)
- $MgAl_2O_4$ (high swelling due to fission fragments, operating restrictions)
- SiC (difficult to recycle using current commercial technology)
- ZrO_2 (difficult to recycle, poor K_{th})
- Zr (difficult to recycle using current commercial technology, acceptance)
- NiAl (long development time, operator acceptance)
- ***MgO (good irradiation behavior but poor corrosion resistance)***

MgO-PuO_2 Fuels: U.S. Experience

- Zircalloy clad specimens in ETR
  - 4 MgO-2.71 wt%Pu, 4 MgO 12.95 wt% Pu, 1.44 cm OD
  - Peak power 59-165 kW/m, burnup 5-72%
  - Peak Temperatures 700 – 2450°C (±20%)
    - ➢ Central void and major Pu redistribution at 165 kW/m
- Zircalloy clad specimens in PRTR
  - 19 1.43 cm OD x 251 cm rods, 2.1 wt% PuO_2
  - Swage compacted –6 mesh MgO + -325 mesh PuO_2
  - Failure 3 hours after full power (60 MW)
    - ➢ Cause: high local temps, F contamination of Pu, water in MgO caused cladding breach
    - ➢ Irradiation continued 8 days, 23 cm fuel lost
    - ➢ USAEC HW-SA-3127, USAEC HW-76300



165 kW/m,
2450°C,
72% burnup

MgO-based Inert Matrix Fuel

- $MgO + H_2O \rightarrow Mg(OH)_2$
  - Up to 117% volume expansion due to lower density of $Mg(OH)_2$
  - Fuel pin rupture and fuel washout

As fabricated MgO pellet

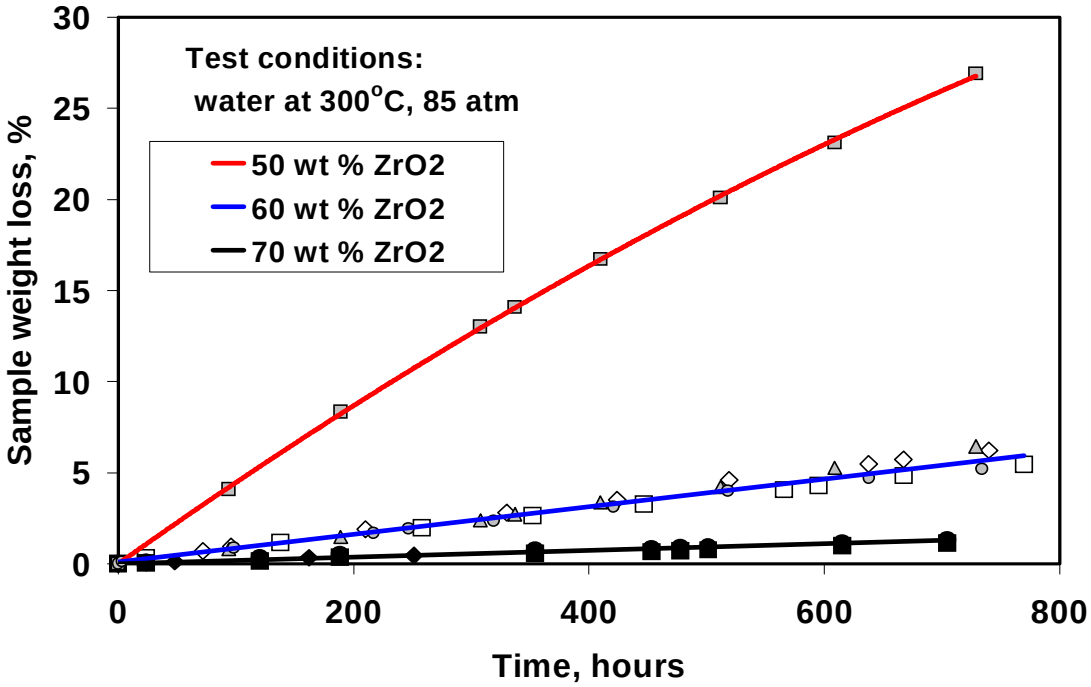
MgO pellet after 3 hr in boiling water

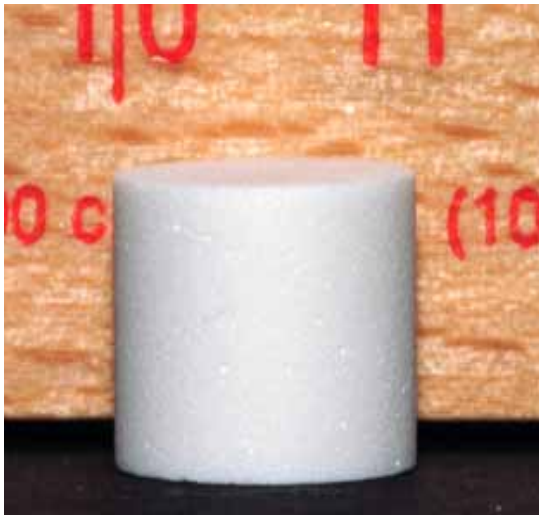
Possible solutions to MgO Corrosion

- MgO microstructure
  - Larger grain size
  - Fewer grain boundaries reduce susceptibility to corrosion attack
- Surface coating
  - A surface layer acting as a diffusion barrier
- Use of ‘additives’
  - Formation of an insoluble grain boundary phase
  - Formation of a hydration-resistant solid solution
  - Formation of a hydration-resistant multi-phase system

Corrosion Test Results

- Long-term stability in water has been demonstrated

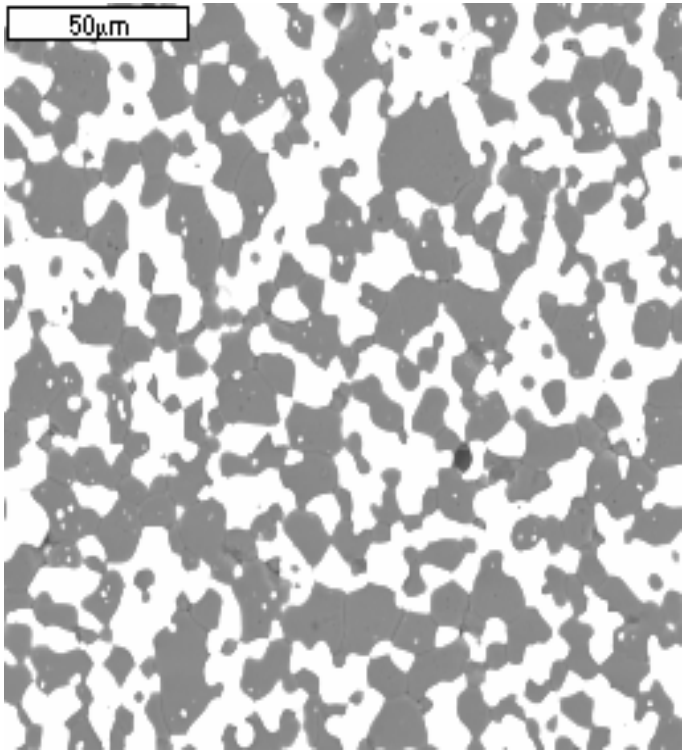




$MgO\text{-}ZrO_2$ ceramic pellet after a 400 hr. exposure to the water at 300°C, 85 atm

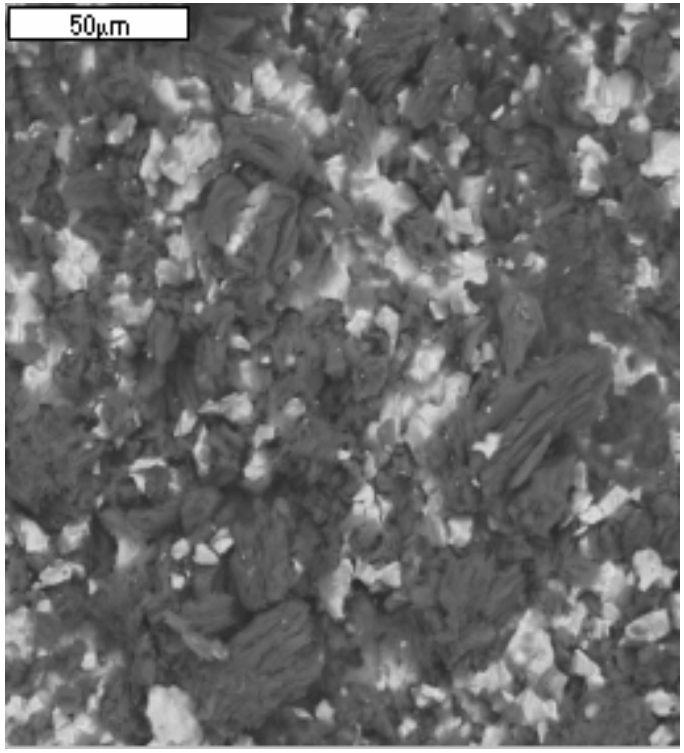
Microstructure before and after corrosion testing

As-fabricated, polished and etched surface



White phase: ZrO_2-MgO(ss); grey phase: MgO

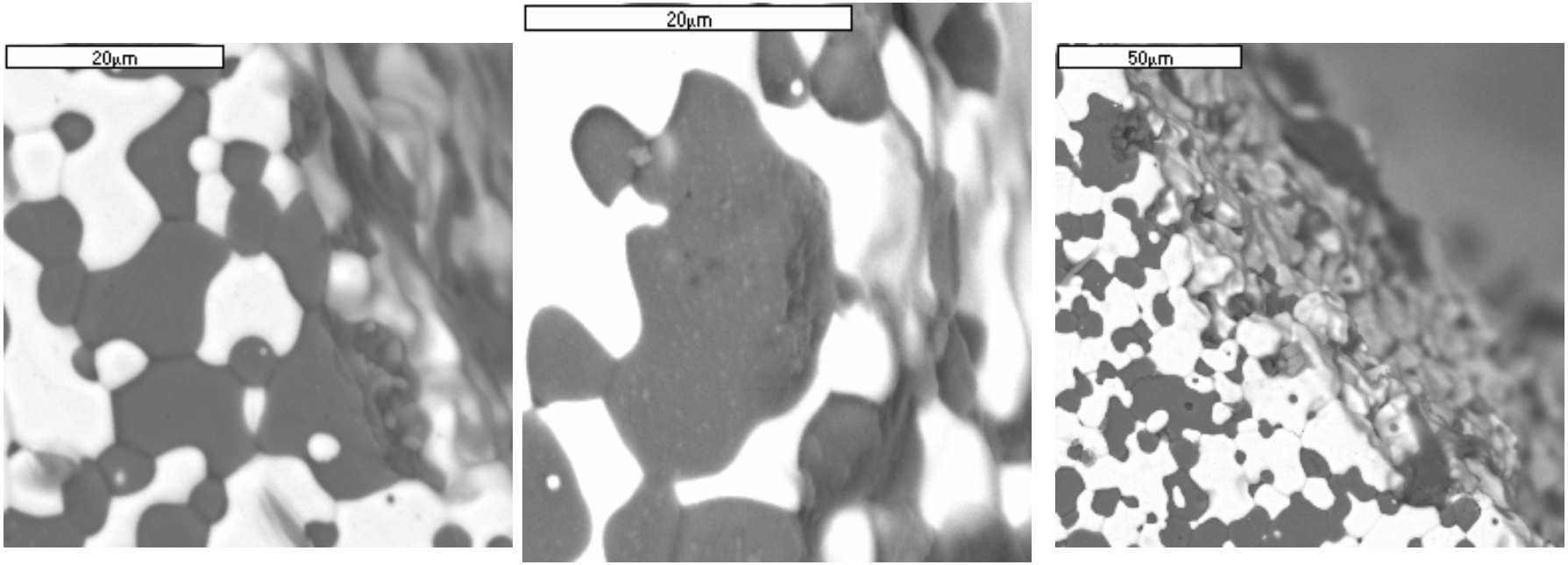
Surface after 700 hr in deionized water at 300°C



White phase: ZrO_2-MgO(ss); grey phase: $Mg(OH)_2$+MgO

SEM of $MgO-ZrO_2$ cross-section after exposure to water

Hydration reaction is confined to the surface layer
Bulk of the MgO is encapsulated by ZrO_2



$MgO-ZrO_2$ ceramics after 700 hr in deionized water at 300˚C
White phase: ZrO_2-MgO(ss); grey phase: MgO

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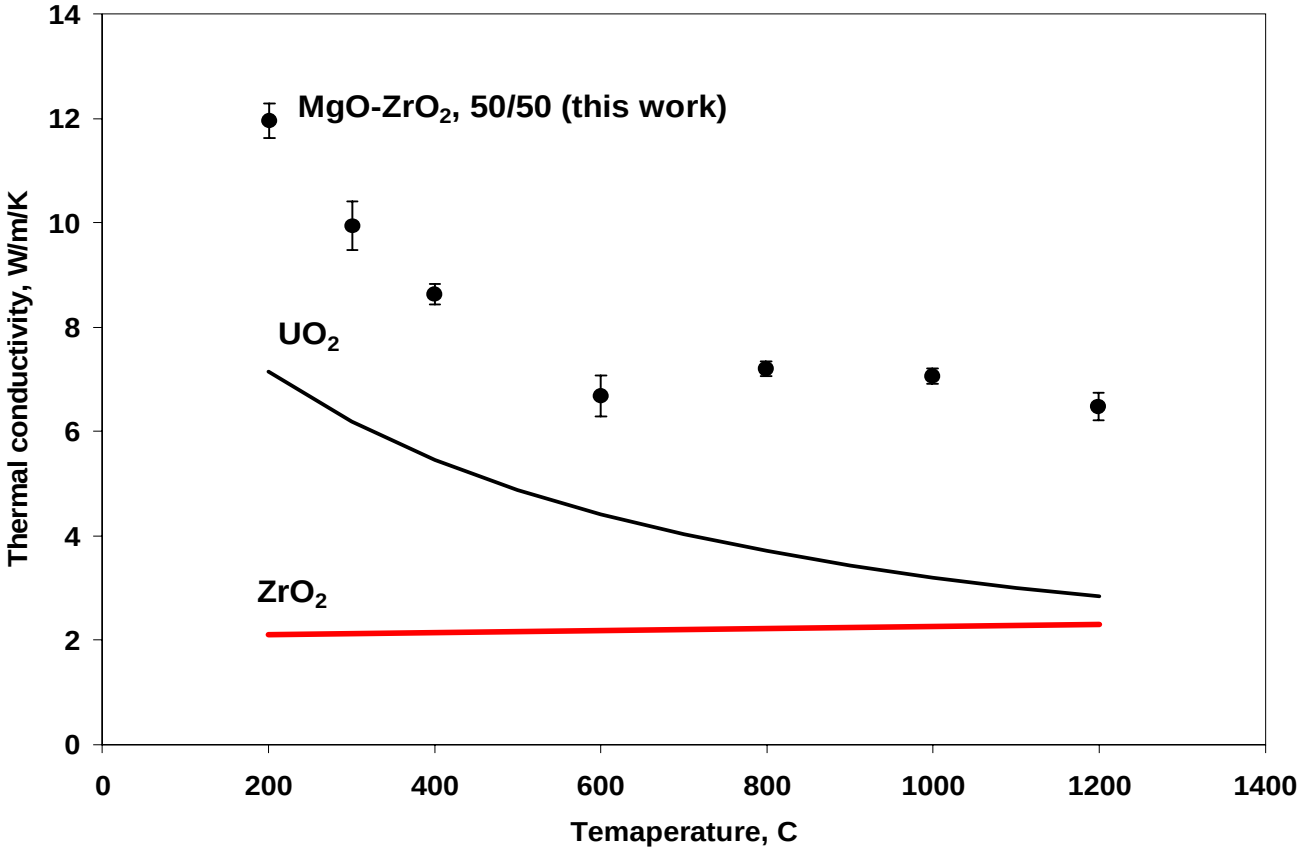
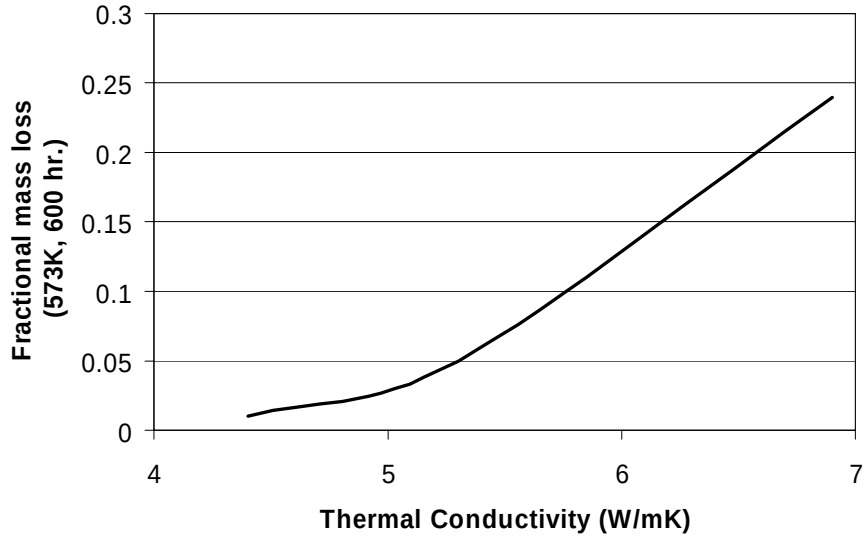
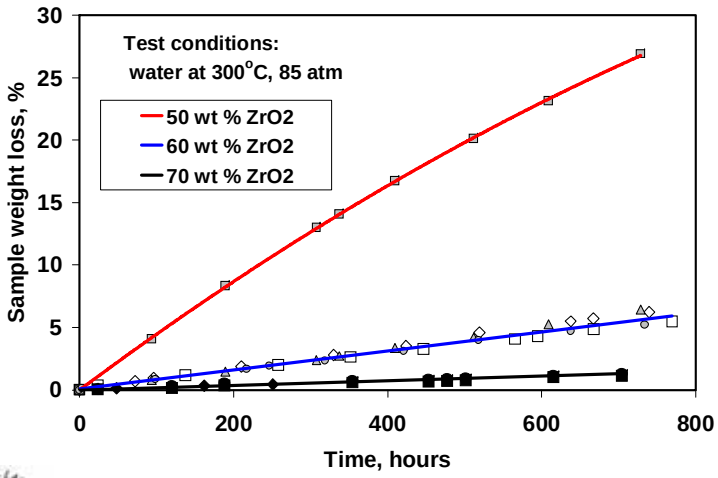
Thermal Conductivity Data

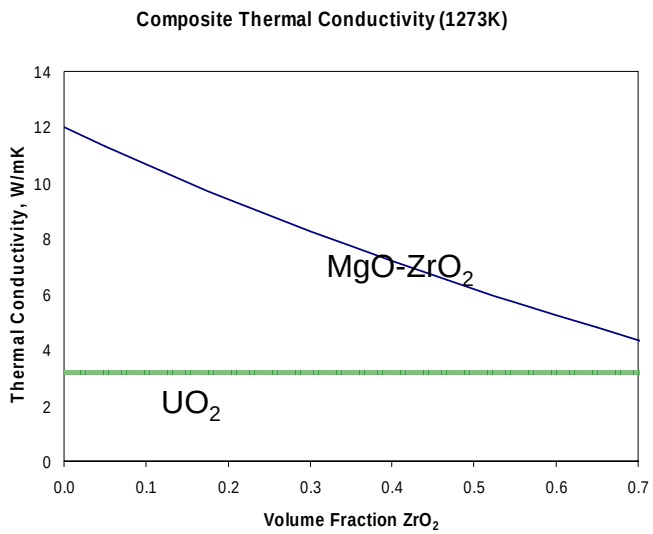


- **Preliminary: Still need heat capacity data, cooling corrections, thermal expansion corrections**

Optimization of Two-phase Matrices







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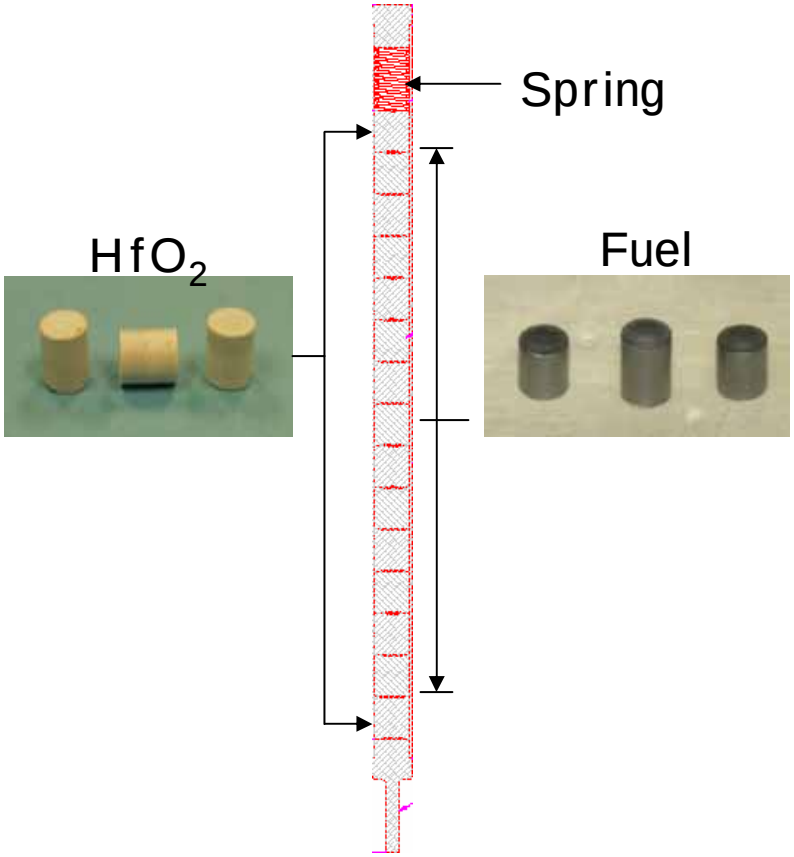
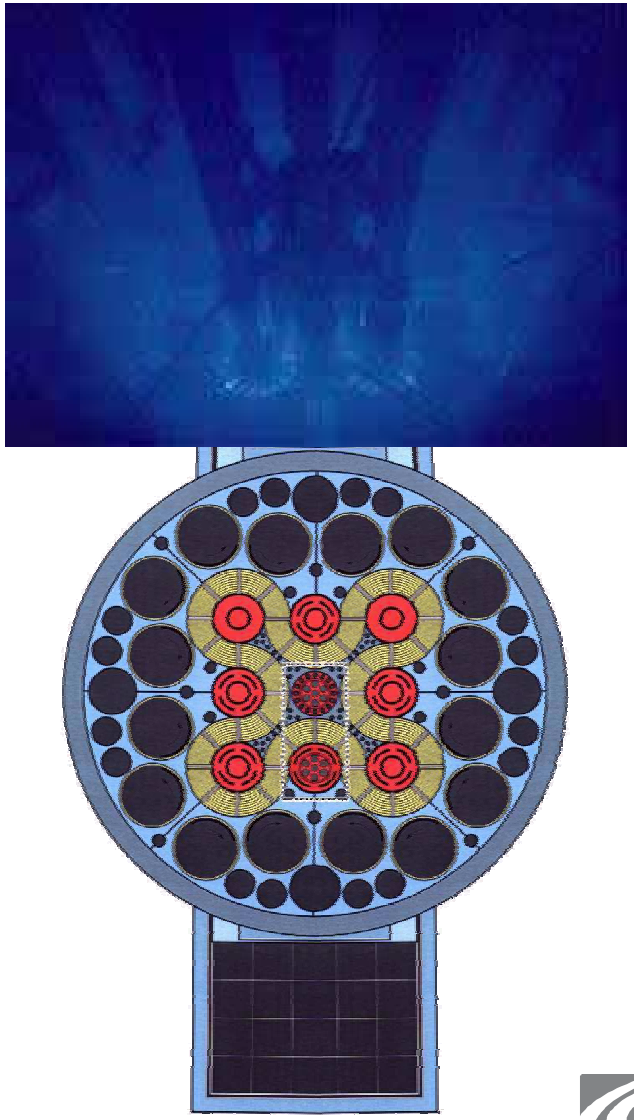
IMF Irradiation Testing

- **Planned for insertion of LWR-2 irradiation test in ATR in mid-2006**





Advanced Test Reactor

THE END