Reactor Fuels
Fuel Requirements

- Maintain physical integrity to contain nuclear materials and keep fuel in position
- Compatible with coolant
- Minimum adverse impact on neutron economy
- Transfer heat to coolant
- Allow for control
- Facilitate extended storage and disposal
Outline

• Metal clad fuels: oxide and variants
  – Water-cooled reactors
  – SCRs

• Graphite-based fuels: block and pebble VHTRs

• Effects of irradiation on fuels

• Spent fuel characteristics
Metal-Clad, Water-Cooled Fuels
Pressurized Water Reactor (PWR)

- Dimensions: square, H=4.1m, 21cm x 21 cm
- Weight: 460 kgU, 520 kg UO$_2$, 135 kg hardware
  - Hardware mostly Zircaloy [(Zr with 1.5% Sn, 0.5% (Fe, Cr, Ni)]
  - Grid spacers: Zircaloy, Inconel, stainless steel
  - End pieces: Stainless steel, Inconel
- Fuel element array: 14 x 14 to 17 x 17
- Fuel element size: 1 cm OD, H=3.9m
- Enrichment: 3-5%
- May have separate burnable poison rods
PWR Fuel Pellets
Pressurized Water Reactor

Pressurized-Water Fuel Assembly
Boiling Water Reactor (BWR)

- Dimensions: square, H=4.5m, 14 cm x 14 cm
- Weight: 180 kgU, 210 kg UO$_2$, 110 kg hardware
  - Hardware mostly Zircaloy (Zr with Sn, Fe, Cr)
  - Grid spacers: Zircaloy
  - Channel (aka shroud): Zircaloy
  - End pieces: Stainless steel
- Fuel element array: 8 x 8 to 10 x 10
- Fuel element size: 1.25 cm OD, H=4.1m
- Enrichment: 2.5-4.5%
- May have Gd in some rods and variable enrichment in 3-D
Boiling Water Reactor
LWR Fuel Variants

• Advanced Cladding
  – Zirlo (Zr-Nb alloy) and Duplex (layered) for PWRs
    • Duplex D4: Highly corrosion resistant Zr alloy over Zr-4
  – Tweak Zircaloy composition for BWRs
  – Some potential for clad made of SiC

• Moving to more smaller diameter or annular elements
Advanced LWR Fuels

Annular Fuel Pellet

SiC Duplex Fuel Cladding

Figure 1.2 Schematic of I&EC Sintered Annular Pellet Fuel for a 13x13 Assembly.

Figure 1.3 Sintered Annular Pellets Produced by Westinghouse [Kazimi, 2003].

Figure 1.7 Example SiC Duplex Solid Fuel Rod.
LWR Fuel Variants

• Mixed-oxide (MOX) fuel
  – U- 5-8% Pu
    • Already being done in US (weapons Pu) and elsewhere
  – Mixed actinides from advanced reprocessing
    • U-Np-Pu: modest extension of U-Pu fuel technology
    • Am-Cm: a challenge, probably requires targets

• More PWR fuel complexity
  – Enrichment gradation
  – Burnable poisons
  – Axial blankets

• Thorium oxide fuel
Metal-Clad, Metal-Cooled
SFR Breeder

- Breeder: Makes more fissile material than it consumes
- Dimensions: hexagonal, H=4-5.5m, W (flats)=10-20 cm; HM height ~2m
- Weight: ~60 kg HM, ~65 kg MOX, ~135 kg stainless steel hardware (core + axial blanket)
- Fuel element array: 200-300 pins
- Fuel element size: 0.6-0.9 cm OD, H= 4-5m
- Enrichment: 15-30% Pu
- Axial and radial blanket: All depleted UO$_2$
  - Fewer, larger diameter elements
SCR Core Design

![SCR Core Design Diagram](image)
SFR Fuel Variants

• Designs not settled: considerable variation in number of elements, dimensions, and weights possible

• Reduce breeding/conversion ratio to achieve net destruction of transuranics
  – Eliminate fertile blankets in favor of non-fertile neutron reflectors (e.g., stainless steel)
  – Inert matrix (e.g., ZrO$_2$) fuel

• Carbide, nitride, or metal fuel instead of oxide
Graphite-Based Fuels
HTGR Prismatic Fuel

• Dimensions: hexagonal, H=0.8m, 0.36m (flats)
• Weight: 5-7 kgU, 5.5-7.5 kg UO$_2$
  – Hardware 126 kg C (mostly graphite), 4 kg SiC
• ~1000 blocks for 600 MW(t) reactor
• Fuel element array: 210 on a triangular pitch
  – 108 Coolant channels
• Fuel element size: 1.3 cm OD, H=0.8m
  – Contains 14-15 “compacts” with 350-500µm TRISO particles
• Enrichment: 8-20%
• May have separate B$_4$C burnable poison rods
HTGR Prismatic Fuel

Pyrolytic Carbon
Silicon Carbide
Porous Carbon Buffer
Oxide Fuel (TRUO_{1.68})

\{ TRISO Coating

Tristructural-Isotopic

PARTICLES

COMPACTS

FUEL ELEMENTS
HTGR Prismatic Fuel
Spent Fuel Characteristics
Burnup

- Burnup is a measure of how much energy a unit of spent fuel has produced
  - Gigawatt-days (GWd) per metric ton heavy metal (MTHM)
    - GWd: Thermal energy, multiply specific power (GWt/MTHM) by the number of days in the reactor and the fraction of full power on each day
    - MTHM
      - HM is Th+U+Np+Pu . . .
      - HM is based on what was initially present in the fuel
  - 10 GWd/MTHM = fission of 1% of the heavy metal in the fuel
Effects of Neutron Irradiation

• Elemental and isotopic composition changes
  – Actinides fission to produce energy
    • Fission Products produced in amounts equal to actinides destroyed: 50 GWd/MTHM = 5 wt % fission products
  – Irradiation produces neutron capture products
    • Actinides: $^{236}\text{U}$, Np, Pu, Am, Cm, $^{233}\text{U}$ (from thorium)
    • Hardware and fuel matrix: Activation products

• Physical changes: fuel swelling/cracking, clad embrittlement, fission gas release to plenum
How much burnup?

- PWRs/BWRs, VVERs
  - Now 40-50 GWd/MTHM
  - Climbing but enrichment challenges
- Fast reactors
  - Hope for 100+ GWd/MTHM
- Graphite-fueled reactors
  - Hope for 100+ GWd/MTHM
- SMRs: All over the map
In-Reactor Effects of Burnup-Fuel

- The steep temperature gradient in a fuel pellet can lead to deformation into an hourglass shape
  - Cladding surface ~350 C
  - Centerline ~2350 C

- The corners of the hourglass can dig into the cladding leading to weakening and potentially rupture
In-Reactor Effects of Burnup-Fuel

- The temperature gradient and production of fission products lead to cracking
- Volatile species are released to the gas spaces in the rod
  - Noble gases
  - Other gases: hydrogen
  - Semi-volatile species such as Cs
In-Reactor Effects: Cladding

• Zircaloy cladding becomes increasingly brittle as burnup increases
  – Neutron-induced embrittlement
  – More important: hydride embrittlement
    • External
      – Zr corrodes slowly in water
      – Hydrogen is released at the interface and diffuses into the Zircaloy
    • Internal: hydrogen isotopes from fission and neutron-induced reactions
    • Zirconium hydride precipitates form at grain boundaries which leads to brittleness and susceptibility to failure
Fission Product Distribution: Chain

- $235\text{U}$ Fission Fragments
- Percent yield %
- Mass number $A$ of fission fragment

- $A \approx 95$
- $A \approx 137$
- $A \approx 118$

- $\text{Sr}$
- $\text{Xe} \rightarrow \beta \quad T = 14\text{s}$
- $\text{Cs} \rightarrow \beta \quad T = 64\text{s}$
- $\text{Ba} \rightarrow \beta \quad T = 13\text{d}$
- $\text{La} \rightarrow \beta \quad T = 40\text{hr}$
- $\text{Ce} \quad \text{Stable}$
- $\text{Sr} \rightarrow \beta \quad T = 75\text{s}$
- $\text{Y} \rightarrow \beta \quad T = 19\text{min}$
- $\text{Zr} \quad \text{Stable}$
<table>
<thead>
<tr>
<th>Chemical group</th>
<th>$^{235}\text{U}^*$</th>
<th>$^{239}\text{Pu}^*$</th>
<th>15% $^{239}\text{Pu}^\dagger$</th>
<th>85% $^{238}\text{U}$</th>
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<td>0.298</td>
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<tr>
<td>Y + rare earths‡</td>
<td>0.534</td>
<td>0.471</td>
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<td>0.206</td>
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<tr>
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<td>0.516</td>
<td>0.456</td>
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<tr>
<td>Cs + Rb</td>
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<tr>
<td>I + Te</td>
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<tr>
<td>Xe + Kr</td>
<td>0.251</td>
<td>0.248</td>
<td>0.258</td>
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</table>

*All elements with elemental yields greater than 1% are included. The groups shown in the table account for all but about 2% of the fission products. [After L. Burris and J. Dillon, Estimation of Fission Product Spectra in Discharged Fuel from Fast Reactors, USAEC Report ANL-5742, Argonne National Laboratory, June (1957).]


‡ Lanthanum, cerium, praseodymium, neodymium, promethium, samarium, europium, and gadolinium.
FPs Distribution and Effects

- FP distribution varies with element chemistry
  - More noble metals precipitate at grain boundaries: embrittlement
  - Others form stable oxides in the pellet: swelling
  - Some noble gases released (pressure) or cause swelling in pellet
  - Cs and Mo are volatile at operating temperature and migrate into the cladding
Effects of Fission Products on Fuel

• Affect oxidation state of fuel
  – \( \text{UO}_2 \): uranium is in the +4 valence state
  – Most fission products have lower valences
  – Result: uranium is oxidized to +5 or +6 and various chemical compounds form
  – Affects fuel pellet integrity, melting point, etc.

<table>
<thead>
<tr>
<th>Chemical group</th>
<th>Physical state</th>
<th>Probable valence</th>
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<tr>
<td>Zr and Nb*</td>
<td>Oxide in fuel matrix; some Zr in alkaline earth oxide phase</td>
<td>4+</td>
</tr>
<tr>
<td>Y and rare earths†</td>
<td>Oxide in fuel matrix</td>
<td>3+</td>
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<tr>
<td>Ba and Sr</td>
<td>Alkaline earth oxide phase</td>
<td>2+</td>
</tr>
<tr>
<td>Mo</td>
<td>Oxide in fuel matrix or element in metallic inclusion</td>
<td>4+ or 0</td>
</tr>
<tr>
<td>Ru, Te, Rh, and Pd</td>
<td>Elements in metallic inclusion</td>
<td>0</td>
</tr>
<tr>
<td>Cs and Rb</td>
<td>Elemental vapor or separate oxide phase in cool regions of fuel</td>
<td>1+ or 0</td>
</tr>
<tr>
<td>I and Te</td>
<td>Elemental vapor; I may be combined with Cs as CsI</td>
<td>0 or 1−</td>
</tr>
<tr>
<td>Xe and Kr</td>
<td>Elemental gas</td>
<td>0</td>
</tr>
</tbody>
</table>
Effects of Fission Products on Fuel

• Some fission products are noble gases: Kr, Xe
  – A portion (a few to several percent) is released from the fuel
    • Increased pressure in the rod
    • Decreased thermal conductivity across the gap between the pellet and cladding
    • Released if cladding fails
Irradiation: Buildup of Actinides

- Irradiation of uranium produces heavier actinides
  - $^{235}\text{U} \rightarrow ^{237}\text{Np}, ^{238}\text{Pu}$
  - $^{238}\text{U} \rightarrow ^{239-242}\text{Pu}, ^{241,243}\text{Am}, ^{244}\text{Cm}$

- Significant contribution to radiation, especially neutron and alpha
Irradiation: Activation Products

• Neutrons convert initially stable nuclides to radionuclides: activation products
  – Structural metals: $^{59,63}$Ni, $^{60}$Co, $^{93,95}$Zr
  – Coolant: tritium, $^{24}$Na
  – Trace contaminants in structural materials and fuel
    • U produces transuranics
    • N produces $^{14}$C
    • Li, H produce tritium
Spent Fuel Composition

Uranium fuel

3% (Uranium235)
97% (Uranium238)

Emission products
3% (high-level radioactive waste)
1% (Uranium235)
1% (Plutonium)

95% (Uranium238)
Reuseable material
97%
Post-Irradiation Situation

• Complex mix of elements to be considered in separations (~entire periodic table)
• Fuel and cladding are physically degraded
• Greatly increased radioactivity
  – Photons
  – Neutrons
  – Alpha
  – Beta
Post-Irradiation Situation

• Impacts
  – Penetrating radiation $\rightarrow$ need radiation shielding
  – Particles $\rightarrow$ material damage
  – Decay heat $\rightarrow$ provisions for heat removal
Material Damage From Radiation

• Water and organic materials
  – Radiolysis produces hydrogen which can accumulate and explode

• Radiation degrades equipment and chemicals
  – Rubber and plastics are embrittled
  – Organic compounds are degraded to dysfunctional or dangerous species
Radioactivity of PWR Spent Fuel
Decay Heat, PWR, 33 GWd/MT

Spent Fuel

Spent Fuel less most actinides
Backup
Metal-Clad, Water-Cooled
CANDU Fuel

- Cylindrical, 50 cm x 11.3 cm
- 21.8 kg UO$_2$, 19.1 kg U
  - Hardware is Zircaloy-4
- Fuel element array
  - 37: Center pin and 3 rings of 6, 12, and 18
  - 1.3 cm OD, 48 cm long
  - Have new design (CANFLEX) that has 43 pins with a few smaller pins in the center
- Enrichment: natural to ~2%
VVER Fuel

- Hexagonal: 4.6 m x 23.4 cm across flats
  - Size can vary substantially depending on VVER size

- Weight
  - Hardware Zr- 1% Nb
  - Has a shroud like a BWR

- Fuel Element: 312 pins, 0.9 cm OD

- Enrichment: 1.6% - 5%
SMR Fuels

• Many are similar to large reactor fuels: PWR, graphite block, pebble
• Fuel design is still very fluid for more esoteric designs, e.g., Hyperion
SCWR Fuel Concept
Metal-Clad Fuel Fabrication
UO₂ Fuel Fabrication
UO₂ Fuel Fabrication
UO$_2$ Fuel Fabrication
UO$_2$ Fuel Fabrication

- Material cutting
- Manufacture of components
- Welding of bare end-fitting
- Machining of bare end-fitting
- Electron beam welding
- Deburring
- 3D inspection
- Plug assembly
UO$_2$ Fuel Fabrication

Assembly facility

- Fuel assembly envelope and water channel inspection
- Verticality check
- Visual inspection
- Loading into containers
- Fuel assembly bench
- Cage check
- Cage assembly bank
MOX Fuel Fabrication

• MOX defined earlier

• The same as UO$_2$ fuel manufacture however…. 
  – Need to make sure the oxides are very homogenous and have required proportions
    • MOX fuel fabrication from powder usually dilutes a high-concentration “master blend” to the proper Pu enrichment

• Conventional MOX fuels are being tested in the US and are being used in reactors elsewhere
  – Facility using weapons Pu is being built at SRS
MOX Fuel Fabrication

UF₆
Steam Heating

Precipitation

Filtering

Drying

Calcining

Milling

UO₂ Powder

Steam + HF

NH₃OH

Fluidized Bed Reacting

Reducing

H₂ + Heat

UO₂ or MOX Powder

Continue as with UO₂ Fuel

Dry Blending

MOX Powder

U Compound

Pu Compound

Dissolution in HNO₃

Coprecipitation

Filtering

Calcining

Milling

MOX Powder

NH₃
Sol-Gel/Sphere-Pac Fabrication

• Used to prepare oxide fuels (U and MOX) without mixing powders, grinding, and dust

• Involves two steps:
  – Gelation: form kernels of U, Pu, and/or Th oxides by forming spheres
    • Based on ammonia precipitation of hydrated heavy metal oxides
    • 30, 300, and 1200µm optimal for Sphere-Pac (85% T.D.)
    • External gelation seems to be the current preference
  – Spheres are then washed and dried at ~200°C
  – Sphere-Pac: Calcine and then sinter the spheres and load them into clad tubes
Metal Fuel Fabrication

• Begin with a furnace containing a molten mixture of actinides, some fission products, and alloying constituents
• Make a mold having an array of quartz tubes
• Insert mold in furnace, seal, and evacuate
• Lower mold into melt and increase pressure to force metal melt into tubes
• Raise mold, cool, break to yield metal fuel “pellets” ~0.5m long.
• Insert in metal clad as with oxide fuels
Metal Fuel Fabrication
Fabrication Scrap Recycle

• All fabrication processes internally recycle off-specification fuel
  – Essentially dissolution and U/Pu/MOX purification steps used in a reprocessing plant
  – Off-spec metal fuels simply go back into the melt furnace or electrorefiner (more later)
DUPIC

• **Direct Use of PWR Fuel in CANDU Reactors**
  – Direct use = no reprocessing

• Being developed by the Korean Atomic Energy Research Institute (KAERI) and Atomic Energy of Canada Limited (AECL)

• Status: Pellets and elements have been fabricated and are undergoing in-reactor testing in Korea and Canada
DUPIC Process

1. **Spent PWR Fuel** → **PWR Rods**
2. **Decladding** → **Cut to size**
3. **Oxidation/Reduction**
4. **Pelletization**
5. **Sintering**
6. **Parts** → **DUPIC rod** → **DUPIC Bundle**

- **Structural waste**
- **Volatile waste**
- **Cladding waste**
- **Volatile/Semi-volatile waste**
Graphite Fuel Fabrication: Pebble Bed
Pebble Bed Reactor Fuel

• Dimensions: Spherical, D= 6.0 cm
• Weight: 9 g U, 10 g UO$_2$
  – “Hardware” 194 g C (mostly graphite), ~6 g SiC
• ~360,000 pebbles for 400 MW(t) reactor
• Fuel element array: random pile
• Fuel element size:
  – 900µm TRISO particle
  – ~15,000 particles per pebble
• Enrichment: 7-10%
Pebble Bed Reactor Fuel

**Fuel Element Design for PBMR**

- **Fuel Sphere**
  - Dia. 60mm

- **Section**
  - 5mm Graphite layer
  - Coated particles imbedded in Graphite Matrix

- **TRISO Coated Particle**
  - Dia. 0.92mm
  - Inner Pyrolytic Carbon 40/1000mm
  - Silicon Carbide Barrier Coating 35/1000mm
  - Pyrolytic Carbon 40/1000mm
  - Porous Carbon Buffer 95/1000mm

- **Fuel Kernel**
  - Dia. 0.5mm
  - Uranium Dioxide
Pebble Bed Reactor
Graphite Fuel Fabrication
Graphite Fuel Fabrication

Kernel Coating

U/Pu/Th Oxide Kernels from Sol-Gel
- Deposit Porous Carbon
- Deposit Inner Pyrolytic Carbon
- Deposit SiC
- Deposit Outer Pyrolytic Carbon

Ethane/Argon

Propane/Argon

MTS/Hydrogen/Argon

Propane/Argon

HTGR Prism Fuel

Coated Kernel (Fuel Particle)
- Mix with matrix former
- Graphite Powder and Additives
- Carbonize at ~800 C in N₂
- Sinter at ~1800 C In vacuum
- Insert 14-15 Compacts In fuel holes and plug

PBMR Pebble Fuel

Overcoat Particles
- Mix with matrix former
- Mold spherical pebble core (low-pressure)
- Mold graphite pebble shell (high-pressure)
- Machine
- Carbonize at ~800 C in N₂
- Sinter at ~1800 C In vacuum

Graphite Powder and Additives
- Mix with matrix former
- Mold cylindrical fuel compact (low-pressure)
- Carbonize at ~800 C in N₂
- Sinter at ~1800 C In vacuum
- Insert graphite block
Pebble Bed Reactor Fuel

- Dimensions: Spherical, D= 6.0 cm
- Weight: 9 g U, 10 g UO$_2$
  - “Hardware” 194 g C (mostly graphite), ~6 g SiC
- ~360,000 pebbles for 400 MW(t) reactor
- Fuel element array: random pile
- Fuel element size:
  - 900µm TRISO particle
  - ~15,000 particles per pebble
- Enrichment: 7-10%
Pebble Bed Reactor Fuel

Fuel Element Design for PBMR

Fuel Sphere

Diameter: 60mm

Section

5mm Graphite layer
Coated particles imbedded in Graphite Matrix

TRISO
Coated Particle

Dia. 0.92mm

Uranium Dioxide
Fuel Kernel

Dia. 0.5mm

Pyrolytic Carbon 40/1000mm
Silicon Carbide Barrier Coating 35/1000mm
Inner Pyrolytic Carbon 40/1000mm
Porous Carbon Buffer 95/1000mm
Pebble Bed Reactor
Graphite Fuel Fabrication
Graphite Fuel Fabrication

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U/Pu/Th Oxide Kernels from Sol-Gel
- Deposit Porous Carbon
- Deposit Inner Pyrolytic Carbon
- Deposit SiC
- Deposit Outer Pyrolytic Carbon

Ethane/Argon

Propane/Argon

MTS/Hydrogen/Argon

Propane/Argon

HTGR Prism Fuel

Coated Kernel (Fuel Particle)
- Mix with matrix former
- Graphite Powder and Additives
- Mold cylindrical fuel compact (low-pressure)
- Carbonize at ~800°C in N₂
- Sinter at ~1800°C in vacuum

Insert 14-15 Compacts In fuel holes and plug

Graphite Powder and Additives

Graphite Powder

PBMR Pebble Fuel

Overcoat Particles
- Mix with matrix former
- Mold spherical pebble core (low-pressure)
- Mold graphite pebble shell (high-pressure)
- Machine
- Carbonize at ~800°C in N₂
- Sinter at ~1800°C in vacuum

Mold graphite pebble shell (high-pressure)

Graphite block
Spent Fuel Characteristics
Dose Rate from 10y-old PWR Fuel
Radiotoxicity of PWR Fuel
Hazards and Wastes
Fuel Fabrication Hazards

• Chemical
  – Hydrogen, hydrofluoric acid, fluorine

• Organic chemicals
  – Fire, explosion when degraded

• Radiological
  – U: Enough air flow to keep concentration low
  – Pu: Inhalation hazard, need tight containment
  – Np, Am, Cm: like U$^{233}$
Radioactive Wastes from Fuels

• Uranium fuels
  – Low-level wastes
  – Disposal by packaging and disposal in licensed near-surface burial ground

• Pu and minor actinide fuels
  – Will produce wastes containing substantial concentrations of long-lived transuranics
  – Disposal by packaging and disposal in a licensed deep geologic repository or equivalent

• More on waste disposal later
Source of Spent Fuel Characteristics

• Computer codes that solve the Bateman equations
  – ORIGEN2, ORIGEN-S, ORIGEN-ARP
    • Calculationally irradiate fuel or other material to yield nuclide concentrations (gram-atoms) as a function of time
    • Decay composition of irradiated material
    • Contains conversion factors to give output in various units: grams, Ci, watts, radiotoxicity
    • Contains data bases to give source terms for photons and neutrons
**ORIGEN2 Output Sample**

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<thead>
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<th>ELEMENT</th>
<th>Activity</th>
<th>Radioactivity</th>
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