The European Commission’s science and knowledge service

Joint Research Centre
Fuel fabrication and evolution of structural properties under irradiation

Rudy Konings
Content

1. Introduction
2. Properties of Uranium dioxide (and MOX)
3. Fuel Fabrication
4. Radiation effects
5. Behaviour under irradiation

Thursday morning
Thursday afternoon
Friday morning
1

Introduction
Nuclear Fuel

- $^{235}\text{U}$
- $^{239}\text{Pu}/^{241}\text{Pu}$
- $^{233}\text{U}$

The only natural fuel (0.72%)
Requires reprocessing
Breeding from natural $^{232}\text{Th}$
Nuclear Fuel

• Low neutron capture cross section of non-fissile elements
• High fissile density
• No chemical reaction with cladding or coolant
• Favourable physical properties
• High mechanical stability (*isotropic expansion, stable against radiation*)
• High thermal stability (*no phase transitions, no dissociation*)
Nuclear Fuel: Requirements

• During normal operation:
  o Keep its shape
  o No melting should occur
  o Interaction with the cladding should not lead to critical mechanical or chemical interaction

• During accidental condition:
  o No excessive exothermal reactions
  o Limit the amount of volatile species
  o Limited interaction with other core and building materials
Nuclear Fuel: Types and variations

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Melting point (K)</th>
<th>Density (g cm(^{-3}))</th>
<th>U-density (g cm(^{-3}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal</td>
<td>1308</td>
<td>19.05</td>
<td>19.05</td>
</tr>
<tr>
<td>Oxide</td>
<td>3073</td>
<td>10.95</td>
<td>9.6</td>
</tr>
<tr>
<td>Nitride</td>
<td>2798</td>
<td>13.63</td>
<td>12.97</td>
</tr>
<tr>
<td>Carbide</td>
<td>3123</td>
<td>14.32</td>
<td>13.53</td>
</tr>
<tr>
<td>Fluoride (salt)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
# Nuclear Fuel: Types and variations

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Light-water</th>
<th>Heavy-water</th>
<th>Graphite-moderated</th>
<th>High-temperature gas cooled</th>
<th>Sodium-cooled</th>
<th>Carbide</th>
<th>Fluoride (salt)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal</td>
<td>PWR, BWR</td>
<td>CANDU</td>
<td>AGR, RBMK</td>
<td>HTR</td>
<td>SPX, Monju</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Oxide</td>
<td>UO$_2$</td>
<td>UO$_2$</td>
<td>UO$_2$</td>
<td>UO$_2$, (ThO$_2$, UC)</td>
<td>(U,Pu)O$_2$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nitride</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbide</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fluoride (salt)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

- **Light-water**
  - PWR, BWR
  - UO$_2$
- **Heavy-water**
  - CANDU
  - UO$_2$
- **Graphite-moderated**
  - AGR, RBMK
  - UO$_2$
- **High-temperature gas cooled**
  - HTR
  - UO$_2$, (ThO$_2$, UC)
- **Sodium-cooled**
  - SPX, Monju
  - (U,Pu)O$_2$
- **EBR-II**
  - (U,Pu)
  - (U,Pu,Zr)
- **PBTR**
  - (U,Pu)C
- **Molten salt**
  - MSRE
  - LiF/BeF$_2$/ThF$_4$/UF$_4$
### Nuclear Fuel: Types and variations

<table>
<thead>
<tr>
<th>Fuel type</th>
<th>Fuel form</th>
<th>Fuel packing</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal</td>
<td>Single phase</td>
<td>Pellet</td>
</tr>
<tr>
<td>Oxide</td>
<td>Solid solution</td>
<td>Particle</td>
</tr>
<tr>
<td>Nitride</td>
<td>Composite</td>
<td>Liquid</td>
</tr>
<tr>
<td>Carbide</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fluoride (salt)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

- e.g. \( \text{UO}_2 \)
- e.g. \((\text{U,Pu})\text{O}_2\)
- e.g. Mo+\(\text{UO}_2\)
Nuclear Fuel: Light Water Reactors

Source: AREVA S.A., Reproduced with permission
Nuclear Fuel: LWR temperature profile

\[ \Delta T(r) = T(R) - T(r) = \frac{\chi}{4\pi\lambda R^2} \left( R^2 - r^2 \right) \]

Linear heating (W cm\(^{-1}\))

Thermal conductivity

Pellet radius (cm)
Nuclear Fuel: Margin to melting

3130 K  Melting temperature $\text{UO}_2$

2123 K  Melting temperature Zircaloy

1500 K  Central fuel temperature

525 K   Coolant temperature

Margin to melting
Nuclear Fuel: What affects the margins?

- Fuel Composition
- Microstructure
- Fuel Burn up
- Radiation effects
- Mechanical evolution
2

Properties of Uranium dioxide
Nuclear Fuel: Uranium dioxide

Fluorite-type face-centered cubic structure
Nuclear Fuel: Uranium dioxide

Source: Guéneau et al. (2002), J. Nucl. Mat. 304:161
Nuclear Fuel: Uranium dioxide

\[ \text{UO}_2 = \text{UO}_{2-x} + \frac{x}{2} \text{O}_2(g) \]

O vacancies

\[ \text{UO}_2 + \frac{x}{2} \text{O}_2(g) = \text{UO}_{2+x} \]

O occupying “hole” positions in the fcc lattice
Nuclear Fuel: Uranium dioxide

The oxygen potential of UO$_2$ as a function of the O/U ratio and temperature.

$$2 \left[ O^{2-} \right]_{\text{lattice}} \rightleftharpoons O_2(g) + 4e^-$$

$$\Delta G(O_2) = RT \ln p(O_2)$$

The oxygen potential of UO$_2$ as a function of the O/U ratio and temperature.
Nuclear Fuel: Uranium dioxide

Key thermal properties for safety assessment:

- Thermal expansion
- Melting temperature/Liquidus temperature
- Heat capacity
- Thermal diffusivity

Thermal conductivity

Strongly related to the phonon structure of the material
Phonon theory predicts that for an **ideal crystal** the heat capacity is **zero** at $T = 0$ K and approaches the value $3nR$ (**Dulong-Petit** limit) at high temperature.
Nuclear Fuel: Uranium dioxide

The heat capacity of UO$_2$ as a function of temperature.

Nuclear Fuel: Uranium dioxide

Nuclear Fuel: Uranium dioxide

The thermal expansion of $\text{UO}_2$ expressed as $\Delta L/L$ a function of temperature.
Nuclear Fuel: Uranium dioxide

\[ \lambda = \lambda_{\text{phonon}} + \lambda_{\text{electronic}} \]

- The phonon contribution dominates at low temperature (< 2000 K)

\[ \lambda = \frac{1}{A + BT} \]

- The electronic contribution becomes significant at high temperatures
- The phonon contribution is affected by impurities and microstructure changes
Nuclear Fuel: Uranium dioxide

The thermal diffusivity of UO$_2$ determined by LAF and CLASH instruments in JRC Karlsruhe.

Corrected to 95% theoretical density

Nuclear Fuel: Uranium dioxide

Nuclear Fuel: Uranium dioxide

Nuclear Fuel: Mixed oxide

- Plutonium dioxide also has an fcc structure
- Pu$^{4+}$ has a slightly smaller ionic radius
- Pu$^{4+}$ has a different electronic structure
- PuO$_2$ is very poorly soluble in acids
Nuclear Fuel: Mixed Oxide

The revised UO$_2$-PuO$_2$ phase diagram experimentally determined by laser melting studies and modelled using the CALPHAD approach.

Melting temperature of PuO$_2$ was found to be about 300 K higher than accepted values from the 1960s.

Nuclear Fuel: Mixed Oxide

Nuclear Fuel: Mixed Oxide

The thermal conductivity of (U,Pu)O₂ as a function of the Pu concentration.
Nuclear Fuel: Mixed Oxide

Inset: The thermal conductivity of UO₂ is dependent on porosity

Maxwell-Euken correction:

$$\lambda = \lambda_0 \frac{1 - P}{1 + \beta P}$$

Nuclear Fuel: Mixed Oxide

The thermal conductivity as a function of $x$ in $(U,Pu)O_{2-x}$
Pros and cons of UO$_2$

Pro
- Isotropic expansion (fcc)
- High melting point
- Forms solid solution with PuO$_2$

Con
- Low thermal conductivity
- Low fissile density

3 Fuel fabrication
Fuel Fabrication

Provide a compact that:

- is mechanically stable
- has the required density
- has the right dimensions
- has the right microstructure
- is free from impurities

- no cracks, no chips
- > 95% theoretical density
- very small tolerance (μm’s)
- grain size
- Neutron poisons, halides
### Fuel Fabrication: Typical specification

<table>
<thead>
<tr>
<th>Property</th>
<th>Unit</th>
<th>Target value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Outer diameter</td>
<td>mm</td>
<td>8.05 ± 0.01</td>
</tr>
<tr>
<td>Pellet height</td>
<td>mm</td>
<td>8 ± 1</td>
</tr>
<tr>
<td>Visual aspect</td>
<td>-</td>
<td>No cracks, no chips</td>
</tr>
<tr>
<td>Pellet density</td>
<td>%TD</td>
<td>96 ± 1</td>
</tr>
<tr>
<td>Grain size</td>
<td>µm</td>
<td>5-25</td>
</tr>
<tr>
<td>Pore size</td>
<td>mean µm</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>limit µm</td>
<td>500</td>
</tr>
<tr>
<td></td>
<td>condition</td>
<td>&lt; 10% of the pores &gt; 100 µm</td>
</tr>
<tr>
<td>Stoichiometry</td>
<td>O/M</td>
<td>2.000 ± 0.002</td>
</tr>
<tr>
<td>Thermal stability</td>
<td>-</td>
<td>&lt; 1.5% density change</td>
</tr>
<tr>
<td>Impurities</td>
<td>ppm</td>
<td>B, Cd, Gd &lt; 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Co, N &lt; 75</td>
</tr>
<tr>
<td></td>
<td></td>
<td>F &lt; 10</td>
</tr>
<tr>
<td></td>
<td></td>
<td>C, Ni, Ca &lt; 100</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cl &lt; 15</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Most other elements &lt; 200</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Th &lt; 60</td>
</tr>
<tr>
<td>H₂O content</td>
<td>ppm</td>
<td>&lt; 100</td>
</tr>
</tbody>
</table>
Fuel Fabrication: Powder preparation

The ADU process: aqueous process

\[
2 \text{UF}_6 + 7 \text{H}_2\text{O} + 2 \text{NH}_3 \rightarrow (\text{NH}_4)_2\text{U}_2\text{O}_7 + 12 \text{HF}
\]

\[
(\text{NH}_4)_2\text{U}_2\text{O}_7 \rightarrow \text{U}_3\text{O}_8 \rightarrow \text{UO}_2
\]
Fuel Fabrication: Powder preparation

The AUC process: aqueous process

- **UF₆**
- Precipitation: $\text{UF}_6 + 5\text{H}_2\text{O} + 10\text{NH}_3 + 3\text{CO}_2 \rightarrow (\text{NH}_4)\text{UO}_2\text{(CO}_3\text{)}_3 + 6\text{NH}_4\text{F}$
- Drying
- Ammonium uranyl carbonate: $(\text{NH}_4)\text{UO}_2\text{(CO}_3\text{)}_3$
- Reduction: $(\text{NH}_4)\text{UO}_2\text{(CO}_3\text{)}_3 + \text{H}_2 \rightarrow \text{UO}_2 + 3\text{CO}_2 + 4\text{NH}_3 + 3\text{H}_2\text{O}$
- Milling
Fuel Fabrication: Powder preparation

The IDR process: integral dry route

\[
\begin{align*}
UF_6 \text{ vapor} & \quad \rightarrow \quad UF_6 + 2H_2O \quad \text{(vap)} \rightarrow \quad UO_2F_2(\text{sol}) + 4HF \\
\text{Rotary kiln (+steam)} & \quad \rightarrow \quad UO_2F_2(\text{sol}) + H_2O \rightarrow \quad UO_3 + 2HF \\
\text{Rotary kiln (+H}_2\text{)} & \quad \rightarrow \quad UO_3 + H_2 \rightarrow \quad UO_2 + H_2O \\
\text{Controls} & \\
\text{Screw Blender} &
\end{align*}
\]
## Fuel Fabrication: Powder preparation

<table>
<thead>
<tr>
<th></th>
<th>ADU</th>
<th>AUC</th>
<th>IDR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific surface (m²/g)</td>
<td>2.8-3.2</td>
<td>5.0-6.0</td>
<td>2.5-3.0</td>
</tr>
<tr>
<td>Raw density (g/cm³)</td>
<td>1.5</td>
<td>2.0-2.3</td>
<td>0.7</td>
</tr>
<tr>
<td>Tap density (g/cm³)</td>
<td>2.4-2.8</td>
<td>2.6-3.0</td>
<td>1.65</td>
</tr>
<tr>
<td>Mean size (microns)</td>
<td>0.4-1.0</td>
<td>8</td>
<td>2.4</td>
</tr>
<tr>
<td>Morphology</td>
<td>spheroids</td>
<td>Porous aggl.</td>
<td>dendrites</td>
</tr>
<tr>
<td>O/U ratio</td>
<td>2.03-2.17</td>
<td>2.06</td>
<td>2.05</td>
</tr>
<tr>
<td>Fluor (ppm)</td>
<td>30-50</td>
<td>30-70</td>
<td>&lt;25</td>
</tr>
<tr>
<td>Carbon (ppm)</td>
<td>40-200</td>
<td>120</td>
<td>20</td>
</tr>
<tr>
<td>Iron (ppm)</td>
<td>70</td>
<td>10-20</td>
<td>10</td>
</tr>
<tr>
<td>Boron (ppm)</td>
<td>0.2</td>
<td>0.1</td>
<td>&lt;0.05</td>
</tr>
</tbody>
</table>
Fuel Fabrication: Powder preparation

- **Powder preparation**
  - **Matrix**
  - **Additives**
  - **UO$_2$**
- **Compaction**
- **Sintering**
- **Grinding**
- **Controls**

**Source:** European Communities
Fuel Fabrication: Powder preparation

- UO₂
- Blending
- Additives
- Compaction
- Sintering
- Grinding
- Controls

Scraps

Dish

Chamfer
Fuel Fabrication: Sintering

- Transforms a compacted powder to a dense object
- Homogenizes composition (partially)
- Based on thermally activated diffusion
- Principally surface diffusion

Schematic representation of sintering. The dotted lines indicated the as-fabricated particles, the solid lines the necking as a result of mass transport.

https://www.youtube.com/watch?v=Yz2FgkJFjHY
Fuel Fabrication: Sintering

- **Atmosphere:** Ar+H$_2$
- **Temperature:** 1600-1700 °C
- **Duration:** 6 hours at max

Density changes from
\[ \rho_{\text{green}} \approx 50 \% \text{ TD} \] to
\[ \rho_{\text{final}} \approx 95 \% \text{ TD} \]

TD = theoretical density
Fuel Fabrication: Grinding

- Pressed pellets have an hours glass shape → Needs to be corrected
- To meet the specification and tolerances
Fuel Fabrication: Controls

- Dimensions
- Density
- O/U
- Microstructure
- Impurities
- Stability

- Visual, Optical
- Archimedes, geometric
- X-ray, thermal gravimetric analysis
- Ceramography
- Chemical analysis (ICP-MS)
- Re-sintering test
Fuel Fabrication: Microstructure

Grain size UO$_2$: 8 micro-meter

Source: European Communities
MOX Fabrication: Traditional

- UO₂
- PuO₂

1. Ball milling
2. Forced Sieving
3. Compaction
4. Sintering
5. Controls

- Pu-rich islands
  - Hot spots
  - Local high burnup
  - Insoluble

Source: European Communities
MOX Fabrication: MIMAS - industrial

- UO₂
- PuO₂
  - Ball milling
  - Forced Sieving
  - Secondary blender
    - Compaction
      - Sintering
        - Grinding
          - Controls
    - Scraps and rejects

Source: ORANO
MOX Fabrication: MIMAS - industrial

- UO₂
- PuO₂

Ball milling

Forced Sieving

Secondary blender

Compaction

Sintering

Grinding

Controls

Scraps and rejects

MOX Fabrication: JMOX

1. U & Pu co-extraction
2. Conversion to (U,Pu)O₂
3. Ball milling
4. Compaction
5. Sintering
6. Grinding
7. Controls
8. Scraps and rejects
Pros and cons of UO$_2$

**Pro**
- Isotropic expansion (fcc)
- High melting point
- Forms solid solution with PuO$_2$
- Straightforward fabrication
- ...

**Con**
- Low thermal conductivity
- Low fissile density
- Dirty process (dust)
- Many steps (MOX)
- Proliferation risk (MOX)
4 Radiation effects
Radiation effects: Alpha decay

$^{235}\text{U} \rightarrow ^{231}\text{Th} + ^4\text{He}$

$E \approx 5.5 \text{ MeV}$

$^{231}\text{Th} \rightarrow$ 

$^{235}\text{U}$

~ 1500 defects

$^{235}\text{U}$

~ 0.02 μm

~ 100 keV

~ 15 μm

~ 5.5 MeV

$^{4}\text{He}$

~ 200 defects
Radiation effects: Fission

\[ ^{235}\text{U} + n \rightarrow X_1 + X_2 + 2\frac{1}{2}n \]

\[ E \approx 200\ \text{MeV} \]

\[ ^{235}\text{U} \rightarrow ^{89}\text{Kr} + ^{144}\text{Ba} \]

\[ \approx 60000 \text{ defects} \]

\[ ^{144}\text{Ba} \rightarrow ^{89}\text{Kr} + ^{56}\text{Ba} \]

\[ \approx 7\ \mu\text{m} \]

\[ \approx 70\ \text{MeV} \]

\[ \approx 9\ \mu\text{m} \]

\[ \approx 95\ \text{MeV} \]

\[ \approx 40000 \text{ defects} \]
Particles passing through matter loose energy via two interaction processes:

- **Nuclear Collisions** (Rutherford scattering): direct collision with atoms (nucleus)

- **Electronic Collisions** (Born scattering): dissipating the energy to the electrons
Radiation effects: Energy loss

Radiation sources to be considered

- Neutrons
- β-decay
- α-decay
- Fission fragments

minimum displacement energy \(~ 100 \text{ eV}\) in LWR’s neutrons are moderated (< 1eV)

electronic scattering

Atomic displacements
Radiation effects: Alpha decay
Radiation effects: Alpha decay

- Radiation defects lead to expansion of the lattice
- Equilibrium between creation and annihilation occurs after about 1 dpa

Radiation effects: Lattice defects

• Point defects

• Extended defects

• Dislocations (line, loop)
Radiation effects: Lattice defects

(a) interstitial impurity atom
(b) edge dislocation
(c) Self-interstitial atom
(d) Vacancy
(e) precipitate of impurity atoms
(f) vacancy-type dislocation loop
(g) interstitial-type dislocation loop
(h) Substitutional impurity atom
Radiation effects: Alpha decay

TEM micrograph of 10 wt% $^{233}$U-doped UO$_2$ showing the presence of prismatic loops resulting from the alpha-damage

Source: European Communities
Radiation effects: Alpha decay

Source: European Communities
Radiation effects: Alpha decay

TEM micrograph of 10 wt% $^{233}$U-doped UO$_2$ after annealing at 1100 K showing the presence of Helium gas bubbles

Source: European Communities
Radiation effects: Fission product recoil

Oxygen

Uranium

Recoil atom path

15 Å
Radiation effects: Fisson product recoil

Calculated radial temperature distribution of a fission track in UO\textsubscript{2} as a function of time. Calculations were made for the first nm of the material.

Radiation effects: Fission product recoil

<table>
<thead>
<tr>
<th></th>
<th>Energy (keV)</th>
<th>Range (µm)</th>
<th>((de/dx)_{n})</th>
<th>((de/dx)_{e})</th>
<th>No. of defects</th>
</tr>
</thead>
<tbody>
<tr>
<td>Light fission product</td>
<td>95,000</td>
<td>9</td>
<td>0.03</td>
<td>0.97</td>
<td>40,000</td>
</tr>
<tr>
<td>Heavy fission product</td>
<td>70,000</td>
<td>7</td>
<td>0.06</td>
<td>0.94</td>
<td>60,000</td>
</tr>
<tr>
<td>α particle</td>
<td>5,500</td>
<td>15</td>
<td>0.01</td>
<td>0.99</td>
<td>200</td>
</tr>
<tr>
<td>α recoil atom</td>
<td>95</td>
<td>0.02</td>
<td>0.90</td>
<td>0.10</td>
<td>1,500</td>
</tr>
</tbody>
</table>
Radiation effects: Fisson product recoil

Displacements cascades produced by LFPs, HFPs and α-particles

\[ 95\text{Zr} \]
\[ 127\text{I} \]

α-particles

\[ \text{d}E/\text{d}x_{\text{nuclear}} \text{ for recoil atom of } \alpha\text{-decay, e.g. } 94 \text{ keV } ^{234}\text{U} \]

\[ \text{d}E/\text{d}x_e \]

Range, µm

\[ E_d = 40 \text{ eV for U and 20 eV for O} \]
Total number of displacements:
- 40000 for \( ^{95}\text{Zr} \)
- 60000 for \( ^{127}\text{I} \)
- 200 for \( ^4\text{He} \)

Pros and cons of UO$_2$

**Pro**
- Isotropic expansion (fcc)
- High melting point
- Forms solid solution with PuO$_2$
- Straightforward fabrication
- Stable against irradiation

**Con**
- Low thermal conductivity
- Low fissile density
- Dirty process (dust)
- Many steps (MOX)
- Proliferation risk (MOX)
5

Behaviour under irradiation
Irradiation behaviour: Swelling & Cracking

- The UO$_2$ expands with temperature (dilatation)
- During irradiation UO$_2$ swells about 1 vol% per 10 MWd/kgU as results of fission product accumulation
- Volume increase partially compensated by
  - closing of porosity
  - closing of pellet “dishes”
  - closing of gap
Irradiation behaviour: Swelling & Cracking
Irradiation behaviour: Swelling & Cracking

$\text{UO}_2$, 53 GWd/t

$\text{UO}_2$, 66 GWd/t

Source: European Communities
Irradiation behaviour: Cracking & Cracking
Irradiation behaviour: Swelling & Cracking

Source: AREVA S.A., Reproduced with permission
Irradiation behaviour: Swelling & Cracking

- Difference in thermal expansion $\text{UO}_2$ and cladding
- The cladding creeps down (shrinks) due to the accumulation of radiation damage
- Once the gap is closed “Pellet Cladding Mechanical Interaction” occurs
Irradiation behaviour: Plutonium distribution

The Pu and Nd concentration profiles for irradiated UO$_2$ of 97.8 MWd/kgU pellet average burnup measured by EPMA.

Irradiation behaviour: High Burnup Structure

HBS at $r/r_0 = 0.99$. Local burnup is about 210 MWd/kgHM

Irradiation behaviour: High Burnup Structure

Irradiation behaviour: High Burnup Structure

Irradiation behaviour: High Burnup Structure

The Cs and Xe concentration profiles for irradiated UO$_2$ of 97.8 MWd/kgU pellet average burnup measured by EPMA. (After Manzel and Walker, 2002).
Irradiation behaviour: High Burnup Structure

Elemental distribution maps of fission products in the High Burnup Structure near pellet rim.

Source: Walker et al. (2009), J. Nucl. Mat.393:212
IRRADIATION BEHAVIOUR: HIGH BURNUP STRUCTURE

Irradiation behaviour: High Burnup Structure

High Burnup Structure formation in Pu-rich island in irradiated Mixed Oxide fuel

Source: European Communities ©
Irradiation behaviour: Fission products

Fissions yields of $^{235}\text{U}$ (blue) in a thermal neutron spectrum and $^{239}\text{Pu}$ (red) in a fast neutron spectrum.
Irradiation behaviour: Fission products

1) **Dissolved** in the matrix: Rb, Sr, Y, Zr, Nb, Te, Cs, Ba, La, Ce, Pr, Nd, Pm, Sm, Eu

2) **Oxide precipitates at grain boundaries**: Rb, Sr, Zr, Nb, Mo, Se, Te, Cs, Ba

3) **Metallic precipitates**: Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, Se, Te

4) **Gases**: Kr, Xe

5) **Volatile**: Br, Rb, I, Cs, Te
Irradiation behaviour: Fission products
TEM micrograph of a UO₂ fuel irradiated to high burnup showing a large fission product inclusion, dislocation loops, fission product precipitates sometimes pinning dislocation lines.

Source: European Communities ©
Irradiation behaviour: Fission products

TEM micrograph of a UO$_2$ fuel irradiated to high burnup showing a large fission product inclusion, dislocation loops, fission product precipitates sometimes pinning dislocation lines.

Source: European Communities ©
Irradiation behaviour: Fission products

After Grimes and Catlow, Phil. Trans. R. Soc. London A, 335 (1991) 609
Irradiation behaviour: Fission gases

1. Atomic diffusion in the lattice (thermal and radiation induced)
2. Capture in intergranular bubbles
3. Migration of bubbles to grain boundaries
4. Resolution of gas
5. Aggregation to closed porosity
6. Venting via open porosity channels

Irradiation behaviour: Fission gases

1. Small nano-meter sized bubbles
2. Density $10^{23}-10^{24}$ m$^{-3}$
3. Reduce $D_{\text{eff}}$ by reducing the amount of gas available for migration
4. Nucleated in lines in the wake of fission fragments

Source: European Communities ©
Irradiation behaviour: Fission gases

Halden threshold: the relation between the fuel centerline temperature and burnup for a (arbitrary) 1% fission gas release, based on the in-pile experiments performed in the Halden test reactor (Norway).

\[ T_c(\degree C) = \frac{9800}{\ln\left(\frac{bu}{0.005}\right)} \]
Irradiation behaviour: Thermal conductivity

\[ \lambda = \frac{1}{A(T_{\text{irr}}, T_{\text{an}}, bu) + B(T_{\text{irr}}, T_{\text{an}}, bu)T} \]

- \( T_{\text{ann}} \): maximum temperature (700-1450 K) reached during annealing
- \( bu \): burn-up (0 to 100 GWd/t)
- Adaptation to in-pile thermal conductivity: \( T_{\text{irr}} = T_{\text{ann}} = T \)

Irradiation behaviour: Thermal conductivity

Irradiation behaviour: Thermal conductivity

Dominant effects (Ronchi et al., 2004b):
1. Soluble, non-volatile fission products
2. Fission gas and Cs content and its state
3. Irradiation defects (both present at end-of-life and created during subsequent storage by self-irradiation)
4. Precipitation of the fission gasses
5. Annihilation of irradiation defects for thermal recovery conditions

Irradiation behaviour: Melting point

Change in melting temperature of irradiated fuel as a function of burnup (Yamanouchi et al.)

Irradiation behaviour: Fission gas release
Irradiation behaviour: Fission Product Release


Irradiation behaviour: Fission Product Release

SEM of before KEMS measurement

SEM after heating at T = 1800 K in KEMS

Burnup systematically reduces the onset of release
- higher fission gas content
- grain boundary opening

Irradiation behaviour: Fission Product Release

Oxidation significantly reduces the onset of release
- grain boundary oxidation

Irradiation behaviour: Fission Product Release

SEM after annealing at T = 1900 K
Un-oxidised

SEM after annealing at T = 1900 K
Pre-oxidised

Irradiation behaviour: CsI formation?

\[ \text{Cs} + \frac{1}{2}\text{I}_2(\text{g}) = \text{CsI} \]

Thermodynamically stable

\[ 2\text{Cs} + 2\text{Te} = \text{Cs}_2\text{Te} \]

Can form at low oxygen potential

\[ 2\text{Cs} + \text{MoO}_2 + \frac{1}{2}\text{O}_2(\text{g}) = \text{Cs}_2\text{MoO}_4 \]

\[ 2\text{Cs} + \text{UO}_2 + \frac{1}{2}\text{O}_2(\text{g}) = \text{Cs}_2\text{UO}_4 \]
Irradiation behaviour: CsI formation?

Cesium iodide

- Fragments: $\text{Cs}^+$, $\text{I}^+$, $\text{CsI}^+$ (monomer), $\text{Cs}_2\text{I}^+$, $\text{Cs}_2^+$, $\text{I}_2^+$ (dimer)
- $\text{I}^+ / \text{CsI}^+ \approx 3:1$
- Parallel release profiles

Simulated Fuel ($\text{UO}_2 + \text{CsI}$)

- Fragments: $\text{Cs}^+$, $\text{I}^+$, $\text{CsI}^+$
- $\text{I}^+ / \text{CsI}^+ \approx 3:2$
- Parallel release profiles

Source: European Communities ©

Zappey et al., to be published
Irradiation behaviour: CsI formation?

Irradiated fuel (BWR, 55 MWd/kg)

- Absolute intensities similar to Simfuel
- No CsI⁺ ions (but higher background)
- Some temperature regions with parallel release

**Conclusion:** No clear evidence for CsI formation in irradiated fuel
- Insufficient reaction sites?
- Gamma radiation?

Source: European Communities ©
Nuclear Fuel: What affects the margins?

- Fuel Composition
- Microstructure
- Radiation effects
- Fuel Burn up
- Mechanical evolution
Further reading


Thank you

Any questions?
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