Post-Irradiation analysis of fission gases in nuclear fuels

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Fission gases in LWR: a safety issue

PIE facilities

Fission gases behavior: nominal operating conditions

Fission gases behavior: simulated accidental conditions

Further projects
Fission gas a safety issue in Light Water Reactor

- **Industrial issue**
  - Safety criterion: Internal cladding pressure at fuel rods end of life

- **Scientific needs**
  - **Fission gases Formation/Evolution...**
    - Nano-scale bubbles formation
    - From nano-scale to micro-scale bubbles
    - Fission gases features (bubbles internal pressure)
  - **Fission gases vs. fuel microstructure**
    - Link between fission gas release and fuel microstructure
    - Fission gases involved in the HBS (High Burnup Structure) formation
    - Gas release from HBS (normal and off-normal conditions)

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[1]: B. Petitprez, B. Delaye, S. Beguin Proceedings of EHPGM Enlarged Halden Project Group Meeting, Lillehammer, 2005
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Further projects
Post-Irradiation Analyses: Cadarache LECA-STAR Hot Laboratory

Sample preparation Metallographic examination
Microanalyses laboratory
LECA
LWR puncturing, NDE
Annealing test

STAR
Out of Pile annealing capabilities

#### Experimental objectives:
- Better understanding of fuel behavior under nominal condition,
- Out of Pile simulation of fission gas release in case of accidental conditions (LOCA or RIA)

#### Device capabilities:
- Sampling: 10-20 mm segment
- High Frequency induction furnace:
  - Max. Temp.: 2800°C
  - Max. Temp. ramp: 200°C/s
- Accurate temperature monitoring
- Fission gas analysis:
  - Gamma spectrometry for online measurement
  - Gas-chromatography for stable gas analysis (Hel…)

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[3]: J. Noirot, Ch. Gonnier, L. Desgranges, Y. Pontillon, J. Lamontagne
IAEA-TECDOC-CD-1635-2009
[4]: Y. Pontillon et al.
Post Irradiation Examination capabilities

- Actinides and FP:
  - Location
  - Quantification

- Fuel microstructure:
  - Porosity, HBS…
  - Crystalline structure

- Modified equipments
- α-box and gamma shielding
Xenon analysis: Coupling SEM, EPMA and SIMS

**Xe inventory**
- EPMA quantitative analysis
- Sample preparation effect (RIM, fuel center)
- In depth SIMS analysis
- SIMS to EPMA calibration

**Distribution and content:** Actinides, Fission Products (Solid, gas)

**EPMA**
- Secondary Electrons
- Primary Electrons
- X rays

**SIMS**
- Primary Ion beam
- Sample
  - Isotopic analyses
  - Trace elements (ppb)
  - Gas inventory

**SEM**
- Xe inventory
- High Burn-Up fuel
- Before sputtering
- after sputtering

**Graph**
- Xe intensity (c/s)
- Sputtering time (s)
- Baseline (~Gas dissolved in the matrix)
- Peaks (bubbles full of gas)
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Fission gas behavior: nominal operating conditions

67 GWd/tU high burn-up UO₂ PWR fuel

Fuel rod NDE

Axial burn-up distribution

Fission product axial distribution

Fuel pellet μanalysis

Free Volume and Gas pressure measurement
Fission gas inventory (released from fuel column)

\[ F\% = \frac{FGR}{FGC} \]

- FG behavior vs. local μstructure
- Local FGR vs. fuel rod scale FGR
Fission gas: local behavior

Fuel periphery

• SEM:
  • High density of gas bubbles
  • RIM structure

• EPMA Xe profile:
  • << creation
  • FGR ?

• SIMS depth profile:
  • Xe mainly precipitated
  • Baseline (precipitated in nano bubbles) ≈ EPMA
  • total inventory ≈ creation
  • No significant FGR
Fission gas: local behavior

Fuel center

- SEM:
  - High density of gas bubbles
  - EPMA Xe profile:
    - \(<\) creation
    - FGR?

- SIMS depth profile:
  - Part of Xe precipitated
  - nano-precipitated Xe \(\approx\) EPMA
  - total inventory \(<\) creation
  - Local FGR
Local/Total FG release

- Fuel rod FG release: 6.2% (BU ≈ 67 GWd/tU)
- Local FG release: ≈ 8-10% (local BU ≈ 73 GWd/tU)

Fission Gas bubble pressure

<table>
<thead>
<tr>
<th></th>
<th>RIM</th>
<th>0.3-0.4 r/R</th>
<th>Center</th>
</tr>
</thead>
<tbody>
<tr>
<td>W% Xe in bubbles</td>
<td>2</td>
<td>0.2</td>
<td>0.4</td>
</tr>
<tr>
<td>S% Bubbles (&lt; 3.5 μm)</td>
<td>14</td>
<td>1.8</td>
<td>3.5</td>
</tr>
<tr>
<td>At. Vol. (Å³.at⁻¹)</td>
<td>141</td>
<td>182</td>
<td>172</td>
</tr>
<tr>
<td>Bubble pressure (MPa) [4]</td>
<td>13 (300 K)</td>
<td>8 (300 K)</td>
<td>8 (300 K)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>84 (650 K)</td>
<td>130 (1220 K)</td>
</tr>
</tbody>
</table>

[4]: C. Ronchi, JNM, 96 (1981) 314-328
Outline

- Fission gases in LWR: a safety issue
- PIE facilities
- Fission gases behavior: nominal operating conditions
- Fission gases behavior: simulated accidental conditions
- Further projects
Fission gas behavior: simulated accidental conditions

- Lost Of Cooling Accident (LOCA) simulation
- On-line fission gas (85Kr) release
  - Overall release
  - Temperature dependant bursts

63 GWd/tU high burn-up UO₂ PWR fuel

Fission gas behavior: simulated accidental conditions

- RIM cracking
- No significant evolution of nano-precipitated gas
- Sharp decrease of Xe bubbles

- Grain boundaries opening
- No significant evolution of nano-precipitated gas
- No SIMS bubble detection
Fission gas behavior: simulated accidental conditions

Annealing treatment
(on-line FG release)

11.5% $^{85}$Kr

PIE
After heat treatment

12% $^{132}$Xe

50% RIM
35% center
15% mid-radius
Simulated accidental conditions

Importance of coupling separate effect experiments with quantitative PIE

Nominal operating conditions

Multi-scale (Rod to pellet) FG analyses

FGR vs. microstructure
- mainly in hot part of the pellet
- HBS gas retention capacities
- Overpressure of FG

Fuel behavior understanding

New fuel qualification

Safety of nuclear fuel

FGR modeling

FGR features:
- mainly from grain boundaries
- HBS and center release
- bubbles interconnections
- Critical temperature identified

Safety of nuclear fuel
Further projects:

**SEM/FIB/EBSD device**
- FIB column
- EDX detector
- EBSD device
- 3D extraction of porosities and grain boundaries network
- µsample preparation
  - TEM analyses
  - Synchrotron analyses (MARS beamline)

**High pressure annealing loop**
- Impact of fuel rod internal pressure on FGR
- Max Temperature/pressure: 1600°C/1600 bars
- Fission gas behavior: nominal operating conditions
- Standard fuel sample, one pellet
- Main analytical capability: online gamma spectrometry
Thank you for your attention

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