Centre of Excellence for Nuclear Materials

Workshop

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CEA (France)

Post-Irradiation Analysis of Fission Gases in Nuclear Fuels

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Post-Irradiation Analysis of Fission Gases in Nuclear Fuels

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Researches about irradiated fuel are conducted in order to investigate fuel properties and fuel behavior. These works cover various nuclear fuel operating conditions (i.e. Nominal operating conditions, off normal and severe accidents conditions, storage conditions...). Experimental investigations may be carried out through hot cell characterization and/or analytical tests [1-3], (Fig. 1). This talk is focused on the Fission Gas (FG) behavior which is a leading parameter in the overall fuel rod performance. Several parameters promote fission gas release (FGR) from nuclear fuels. Temperature is the most important one, but the local fuel microstructure has also to be considered as a key parameter. With the increase in fuel burn-up, the formation of a FG rich microstructure so-called High Burn-up Structure (HBS) has been evidenced in the fuel pellet periphery. Thus, it is a key issue to assess the contribution of the different parts of the fuel pellet to the total FGR using Post-Irradiation analyses.

Through an example of experimental program performed on a standard PWR UO₂ fuel, the benefits of a coupled approach "Post-Irradiation analyses" and "out-of-pile annealing test" is shown.

(a)

(b)

(c)

Fig. 1: LECA-STAR Hot-cell capabilities; (a) μ -analysis laboratory; (b) puncturing facility; (c) Annealing tests facility (MERARG II).

References

- [1] J. Noirot et al., LWR fuel gas characterization at CEA Cadarache LECA-STAR Hot Laboratory. Post-Irradiation Examination and In-Pile Measurement Techniques for Water Reactor Fuels, IAEA-TECDOC-CD-1635-2009.
- [2] J. Noirot et al., High burnup changes in UO₂ fuels irradiated up to 83 GWd/t in M5® claddings. Journal of Nuclear Engineering and Technology, Vol. 41, n°2-3 (2009).
- [3] Y. Pontillon et al., Experimental and Theoretical Investigation of Fission Gas Release from UO₂ up to 70 GWd/t under Simulated LOCA type Conditions: the GASPARD Program. Proceeding of the 2004 International Meeting on LWR Fuel Performance, Orlando, Florida, September 19-22, 2004.

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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 μ structure
- Fission gases involved in the HBS (High Burnup Structure) formation
- Gas release from HBS (normal and off-normal conditions)

[1]: B. Petitprez, B. Delaye, S. Beguin
Proceedings of EHPGM Enlarged Halden Project Group Meeting, Lillehammer, 2005
[2]: Ch. Valot, J. Noirot, Y. Pontillon
Séminaire MINOS, 12-13 Septembre 2011, Saclay, Neurospin

■ Fission gases in LWR: a safety issue

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Post-Irradiation Analyses: Cadarache LECA-STAR Hot Laboratory

- 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 (He...)

Annealing device principle [3, 4]

[3]: J. Noirot, Ch. Gonier, L. Desgranges, Y. Pontillon, J. Lamontagne
IAEA-TECDOC-CD-1635-2009

[4]: Y. Pontillon et al.
Proc. Of the 2004 International Meeting on LWR Fuel Performance, Orlando, Florida, September 1-22, 2004

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

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Fuel rod NDE

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

$$F\% = \frac{FGR(\text{Fission Gas Release})}{FGC(\text{Fission Gas Creation})}$$

Fuel pellet μ analysis

- FG behavior vs. local μ structure
- 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) $\approx$ EPMA
 - total inventory $\approx$ creation
 - **No significant FGR**

- 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 $\approx$ 67 GWd/tU)
- Local FG release: $\approx$ 8-10%
(local BU $\approx$ 73 GWd/tU)

Fission Gas bubble pressure

	RIM	0.3-0,4 r/R	Center
W% Xe in bubbles	2	0.2	0.4
S% Bubbles ($< 3,5 \mu\text{m}$)	14	1.8	3.5
At. Vol. ($\text{\AA}^3.\text{at}^{-1}$)	141	182	172
Bubble pressure (MPa) [4]	13 (300 K) 84 (650 K)	8 (300 K) 130 (1220 K)	8 (300 K) 150 (1280 K)

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

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

[5]: M. Marcet et al.
Proc. Top Fuel 2009 conference, Paris, France, September 6-10 2009

RIM

Center

Before annealing test

After annealing test

- 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

Annealing treatment (on-line FG release)	<div> <div>11.5% (⁸⁵Kr)</div> <div>12% (¹³²Xe)</div> <div>50% RIM</div> <div>35% center</div> <div>15% mid-radius</div> </div>
PIE After heat treatment

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

Contribution:

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