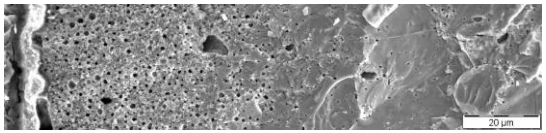
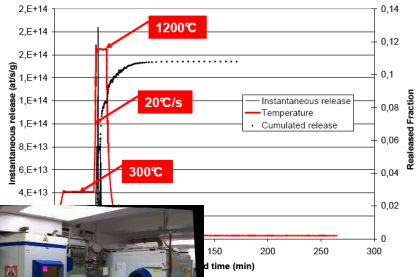
DE LA RECHERCHE À L'INDUSTRIE

Post-Irradiation analysis of fission gases in nuclear fuels

Ch. VALOT, J. NOIROT, Y. PONTILLON





www.cea.fr

MINOS Workshop, Materials Innovation for Nuclear Optimized Systems
December 5-7, 2012, CEA – INSTN Saclay, France

Outline

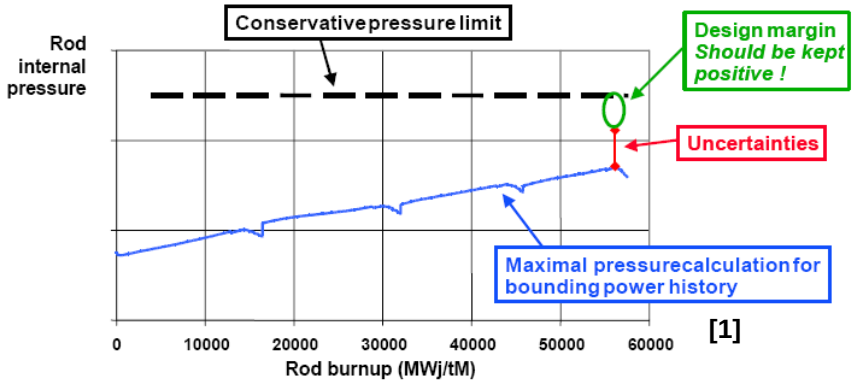
- Fission gases in LWR: a safety issue
- PIE facilities
- Fission gases behavior: nominal operating conditions
- Fission gases behavior: simulated accidental conditions
- Further projects

Fission gas a safety issue in Light Water Reactor

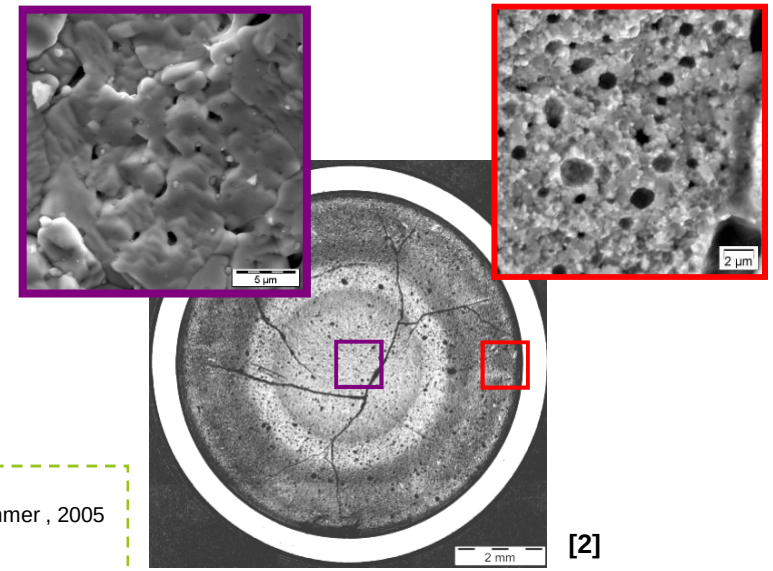
MINOS

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



[1]



[2]

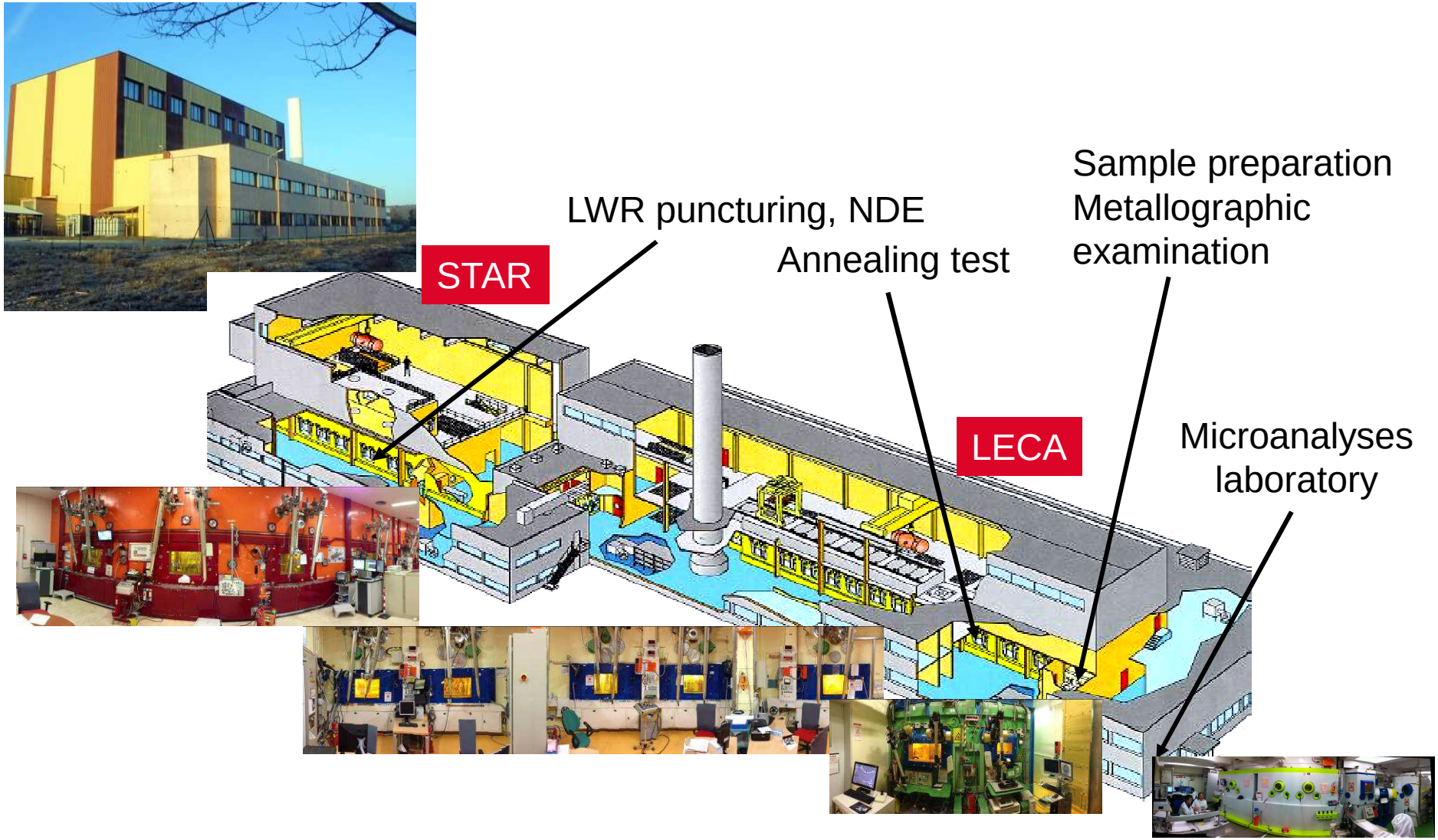
[1]: B. Petitprez, B. Delaye, S. Beguin
Proceedings of EHPGM Enlarged Halden Project Group Meeting, Lillehammer , 2005
[2]: Ch. Valot, J. Noirot, Y. Pontillon
Séminaire MINOS, 12-13 Septembre 2011, Saclay, Neurospin

Outline

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

Post-Irradiation Analyses: Cadarache LECA-STAR Hot Laboratory

MINOS



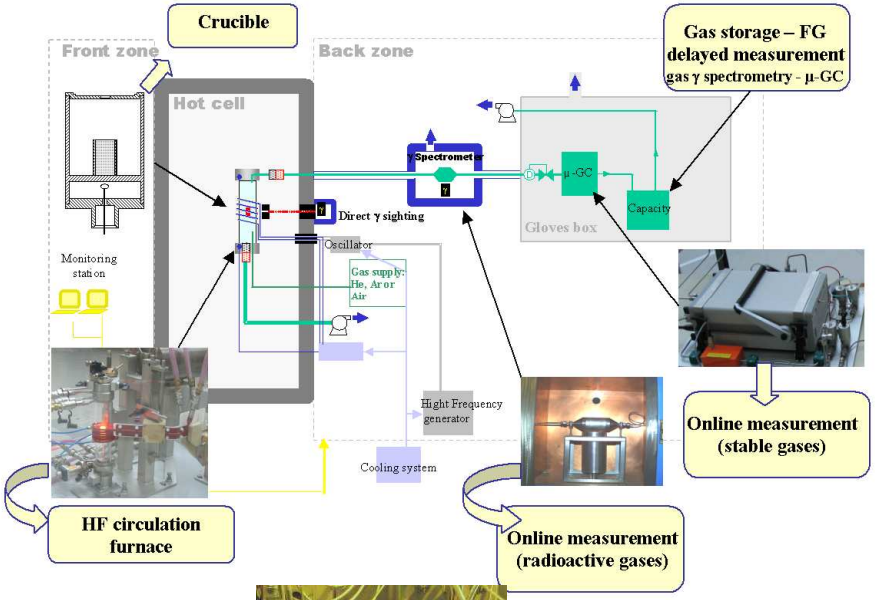
Out of Pile annealing capabilities

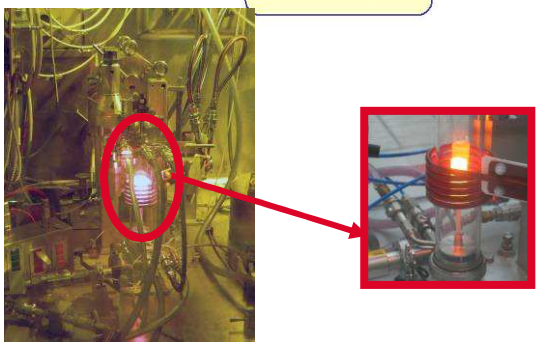
MINOS

- 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℃
    - Max. Temp. ramp: 200℃/s
  - Accurate temperature monitoring
  - Fission gas analysis:
    - Gamma spectrometry for online measurement
    - Gas-chromatography for stable gas analysis (He…)

Annealing device principle [3, 4]



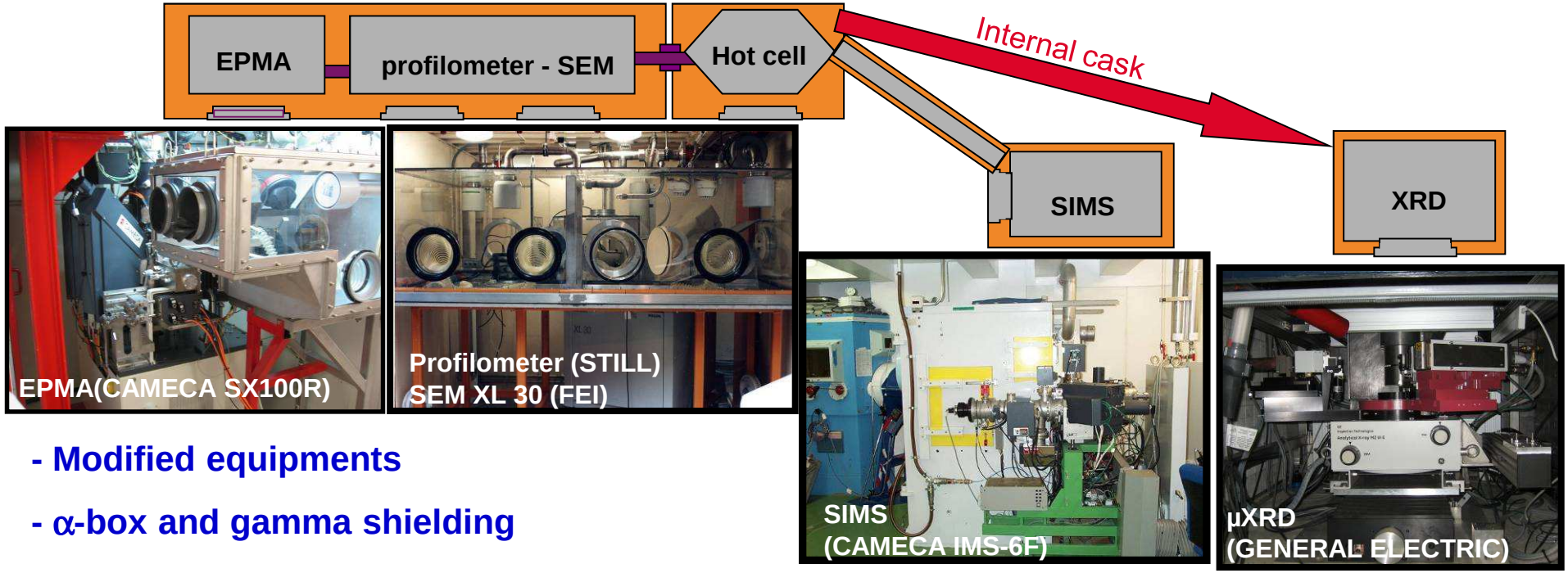


[3]: J. Noirot, Ch. Gonnier, L. Desgranges, Y. Pontillon, J. Lamontagne
IAEA-TECDOC-CD-1635-2009
[4]: Y. Pontillon et al.
Proc. Of the 2004 International Meeting on LWR Fuel Performance, Orlando, Florida, September 1-22, 2004

Post Irradiation Examination capabilities

MINOS

- Actinides and FP:
  - Location
  - Quantification
- Fuel microstructure:
  - Porosity, HBS…
  - Crystalline structure

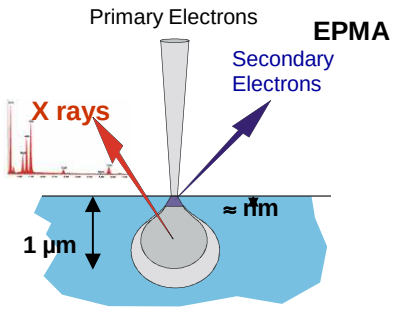


- Modified equipments

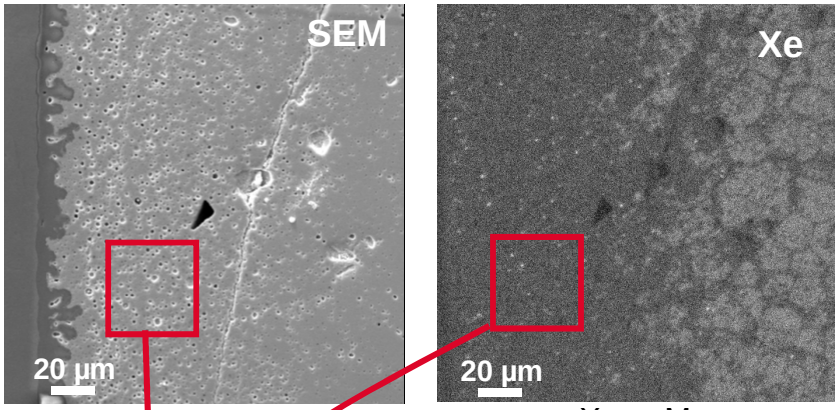
- α-box and gamma shielding

Xenon analysis: Coupling SEM, EPMA and SIMS

MINOS



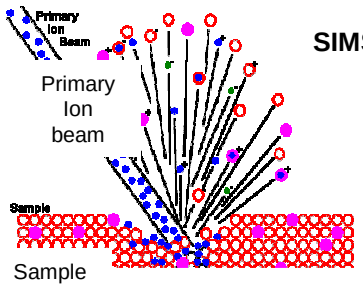
Distribution and content:
Actinides, Fission Products
(Solid, gas)



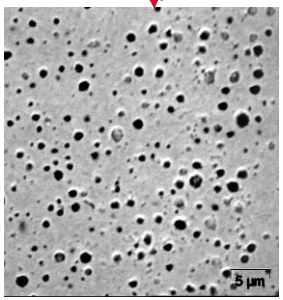
X-ray Map

Xe inventory

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

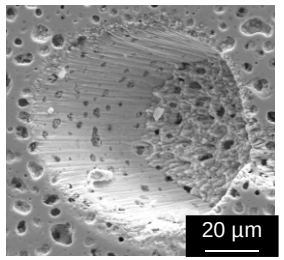


- Isotopic analyses
- Trace elements (ppb)
- Gas inventory

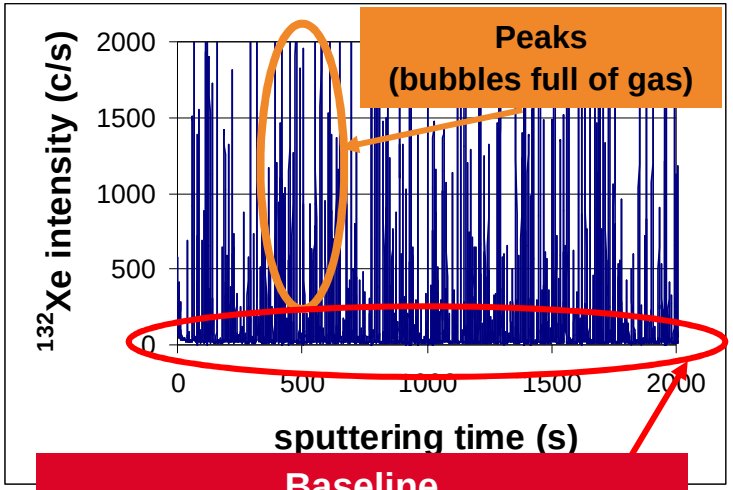


Before sputtering

High Burn-Up fuel



after sputtering



Outline

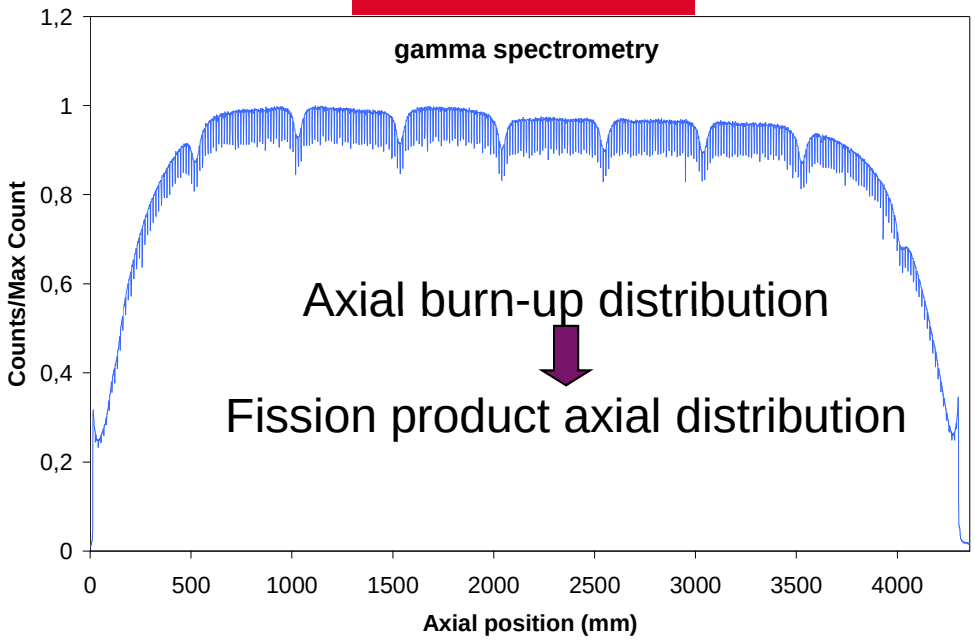
- Fission gases in LWR: a safety issue
- PIE facilities
- **Fission gases behavior: nominal operating conditions**
- Fission gases behavior: simulated accidental conditions
- Further projects

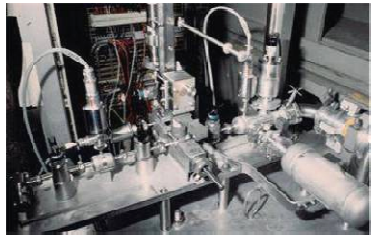
Fission gas behavior: nominal operating conditions

MINOS

67 GWd/tU high burn-up UO_2 PWR fuel

Fuel rod NDE

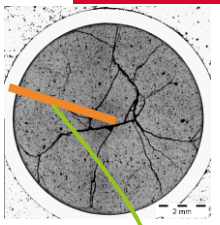


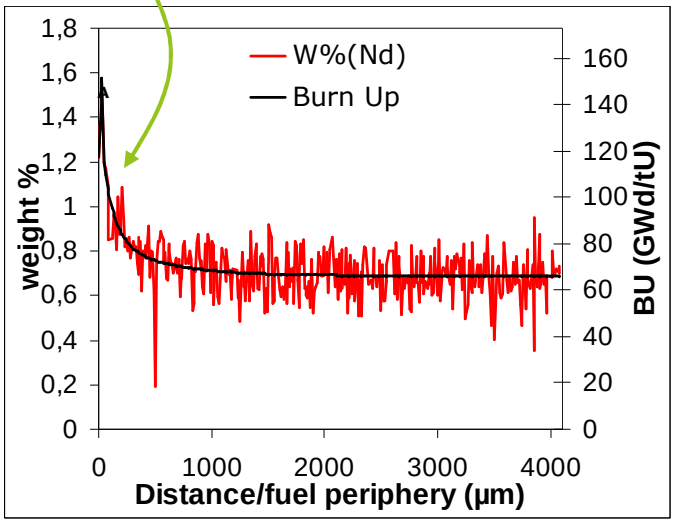


Free Volume and Gas pressure measurement
Fission gas inventory (released from fuel column)

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

Fuel pellet µanalysis

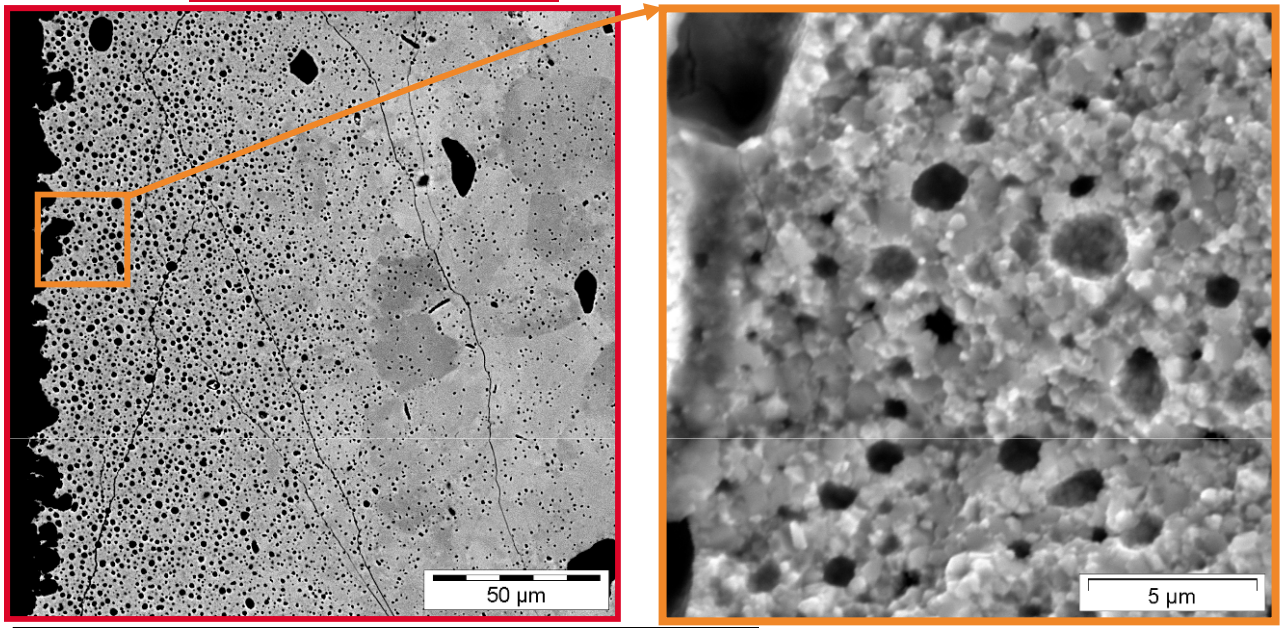




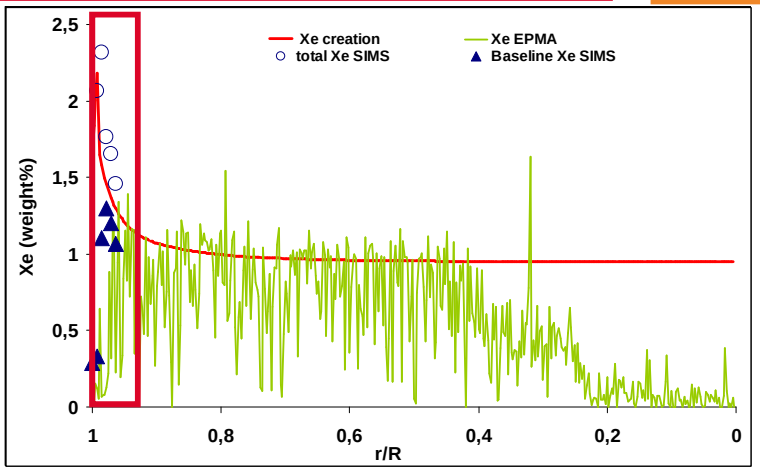
- FG behavior vs. local µstructure
- Local FGR vs. fuel rod scale FGR

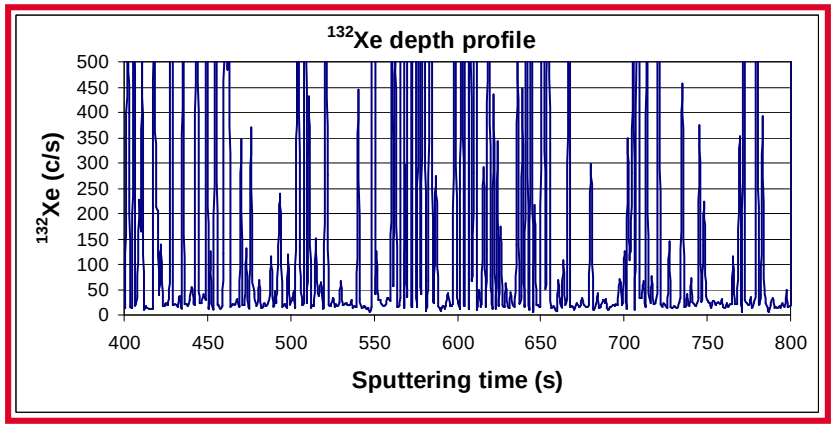
Fission gas: local behavior

Fuel periphery



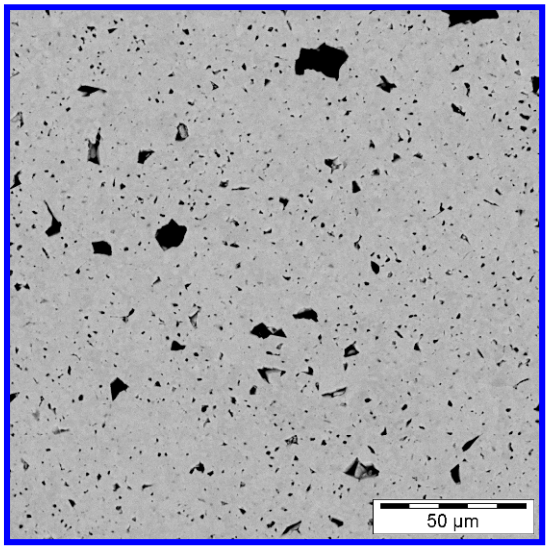
- SEM:
  - High density of gas bubbles
  - RIM structure
- EPMA Xe profile:
  - << creation
  - FGR ?
- SIMS depth profile:
  - Xe mainly precipitated
  - Baseline (precipitated in nano bubbles) ≈ EPMA
  - total inventory ≈ creation
  - **No significant FGR**

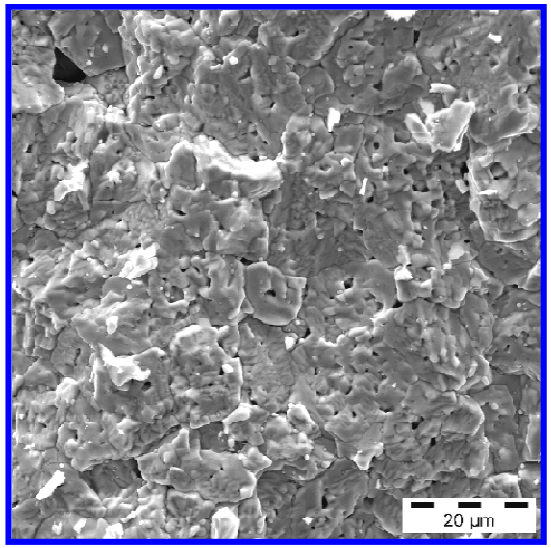




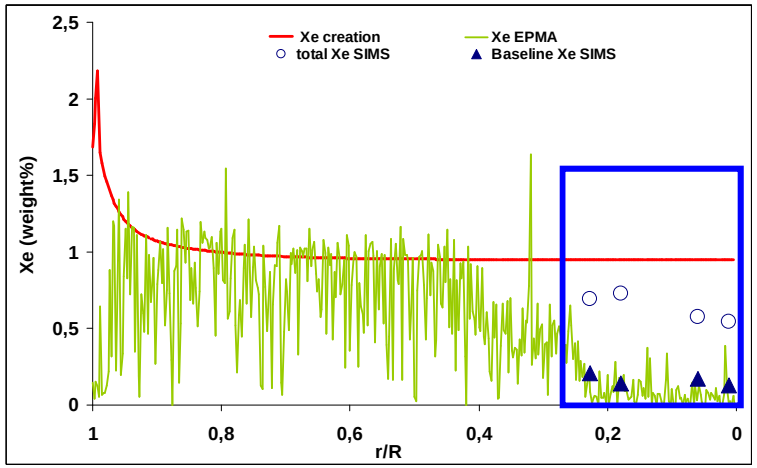
Fission gas: local behavior

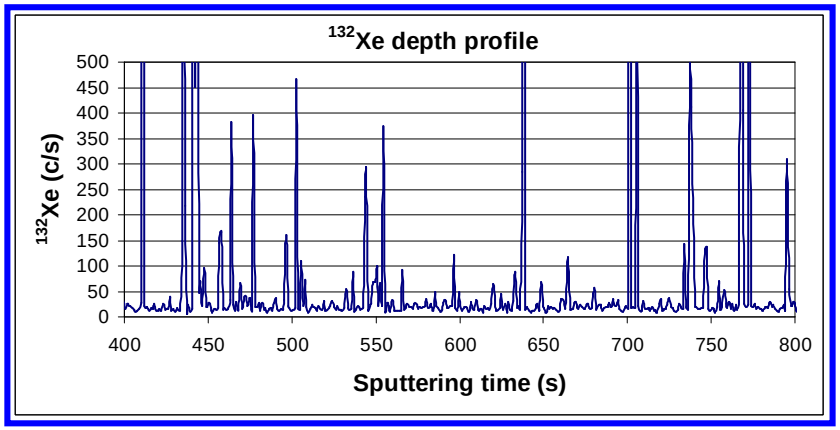
Fuel center





- SEM:
  - High density of gas bubbles
  - EPMA Xe profile:
  - << creation
  - FGR ?
- SIMS depth profile:
  - Part of Xe precipitated
  - nano-precipitated Xe ≈ EPMA
  - total inventory < creation
  - **Local FGR**



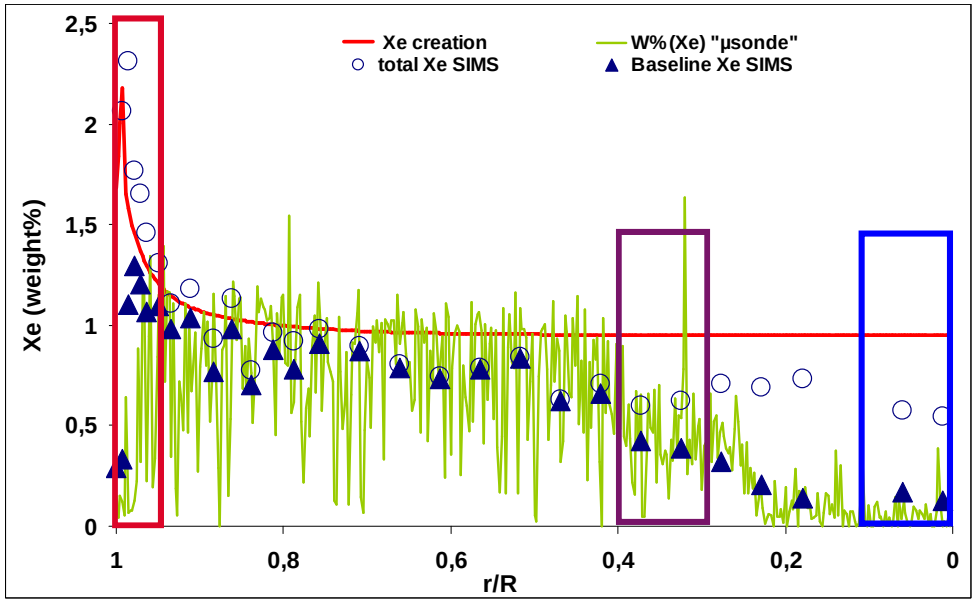


Quantitative analysis

MINOS

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

	RIM	0.3-0,4 r/R	Center
W% Xe in bubbles	2	0.2	0.4
S% Bubbles (< 3,5 µm)	14	1.8	3.5
At. Vol. ($Å^3.at^{-1}$)	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

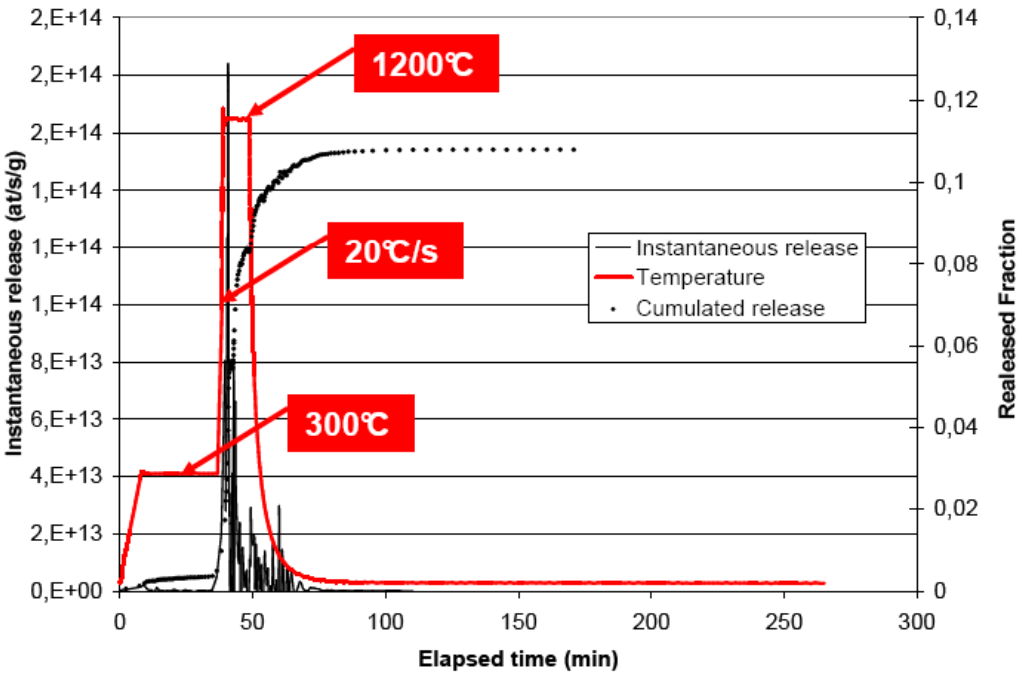
Outline

- 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

63 GWd/tU high burn-up UO_2 PWR fuel [5]

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



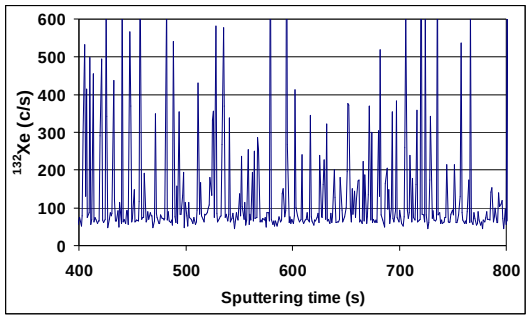
[5]: M. Marcet et al.
Proc. Top Fuel 2009 conference, Paris, France, September 6-10 2009

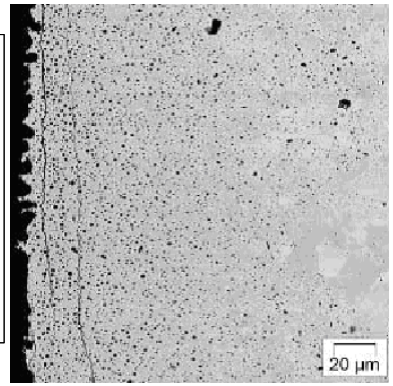
Fission gas behavior: simulated accidental conditions

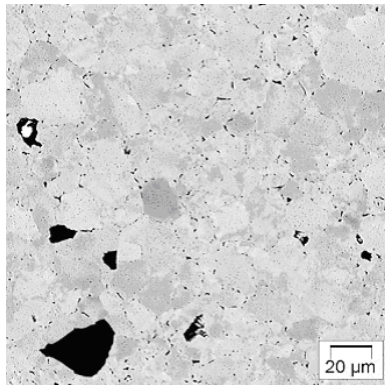
MINOS

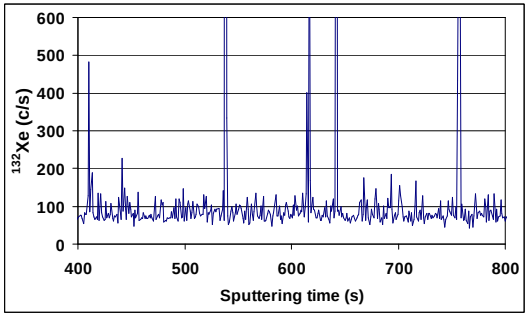
RIM | **Center**

Before annealing test

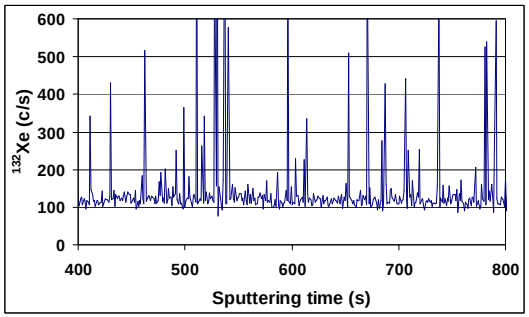


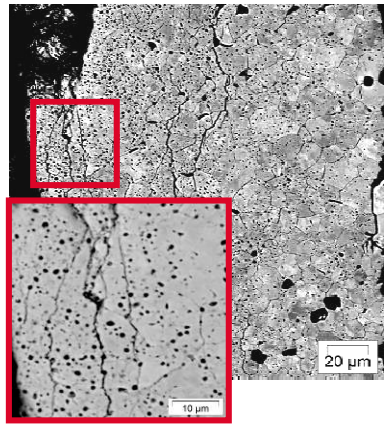


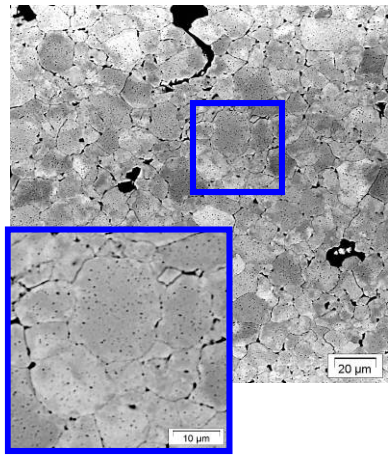


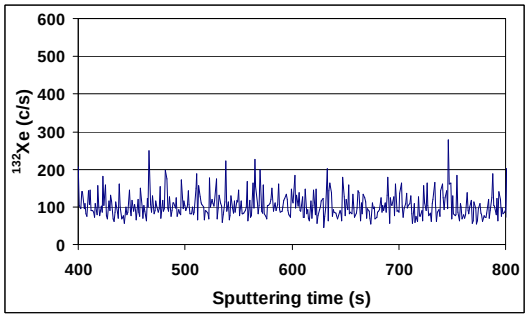


After annealing test





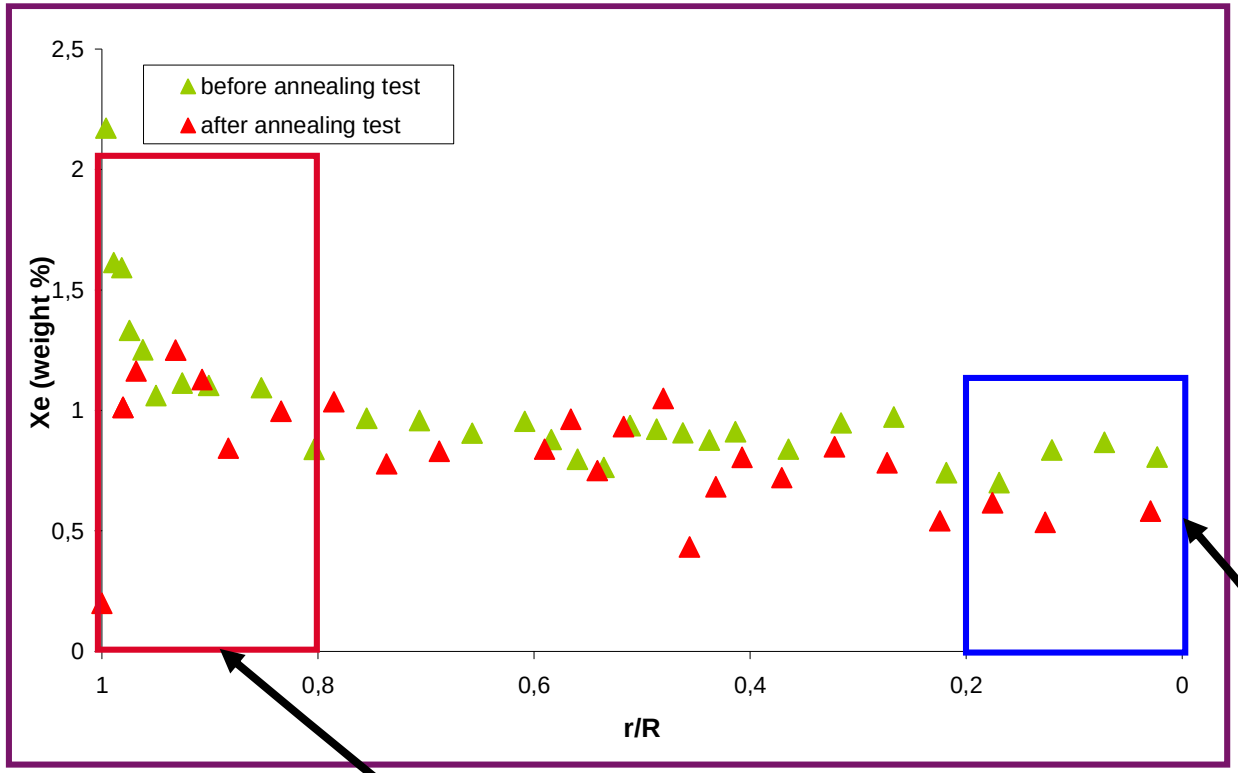




- RIM cracking
- No significant evolution of nano-precipitated gas
- Sharp decrease of Xe bubbles

- Grain boundaries opening
- no significant evolution of nano-precipitated gas
- No SIMS bubble detection

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

Summary

MINOS

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

Simulated accidental conditions

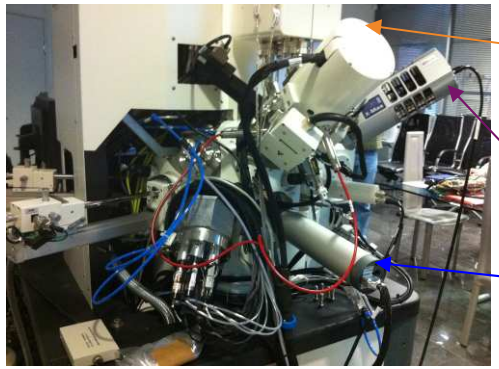
Importance of coupling separate effect experiments with quantitative PIE

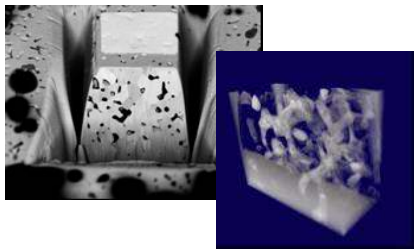
FGR features:
- mainly from grain boundaries
- HBS and center release
- bubbles interconnections
- Critical temperature identified

Further projects:

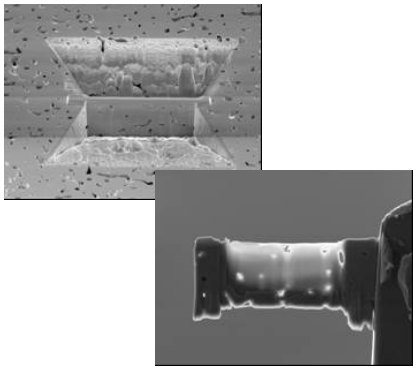
MINOS

SEM/FIB/EBSD device



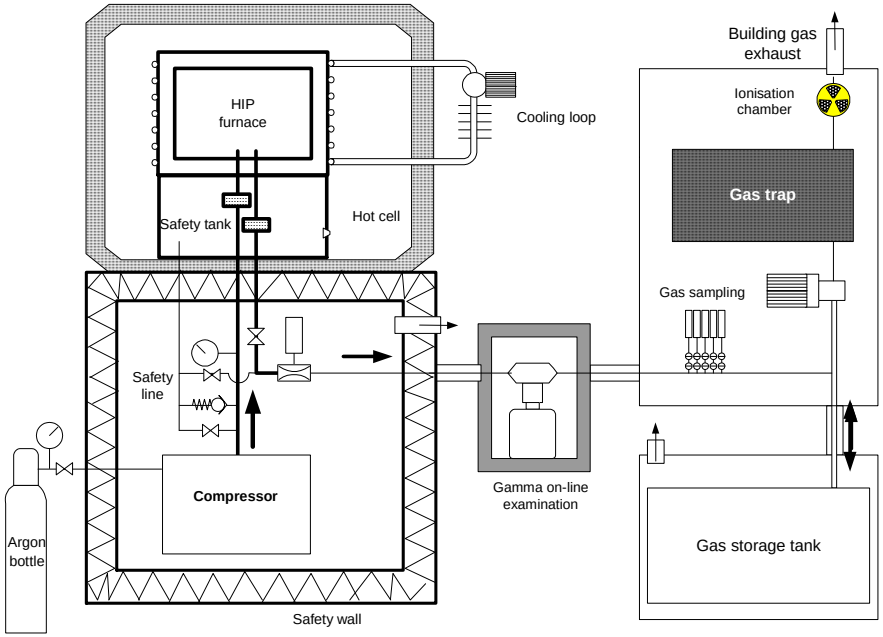


- 3D extraction of porosities and Grain boundaries network



- µsample preparation
  - TEM analyses
  - Synchrotron analyses (MARS beamline)

High pressure annealing loop



- Impact of fuel rod internal pressure on FGR
- Max Temperature/pressure: 1600℃/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:

I. Aubrun, Ph. Bienvenu, S. Clément, B. Lacroix,
J. Lamontagne, B. Petitprez, C. Pozo
Th. Blay, E. Brochard, P. Delion, I. Roure

Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Cadarache | 13108 Saint Paul Lez Durance
T. +33 (0)4 42 25 37 02 | F. +33 (0)4 42 25 36 11
Christophe.valot@cea.fr

DEN
DEC
SA3C
LEMCI