An auto-catalytic mechanism to explain the fission gas release between 500°C and 800°C on UO_2 irradiated fuel

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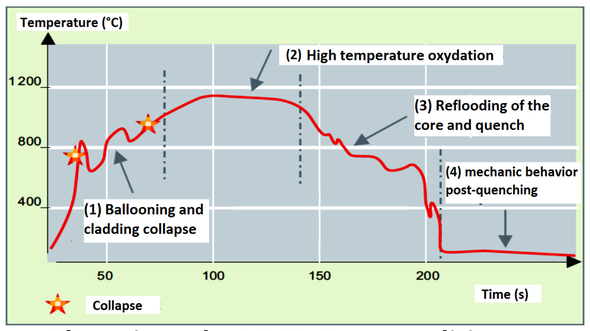
Context

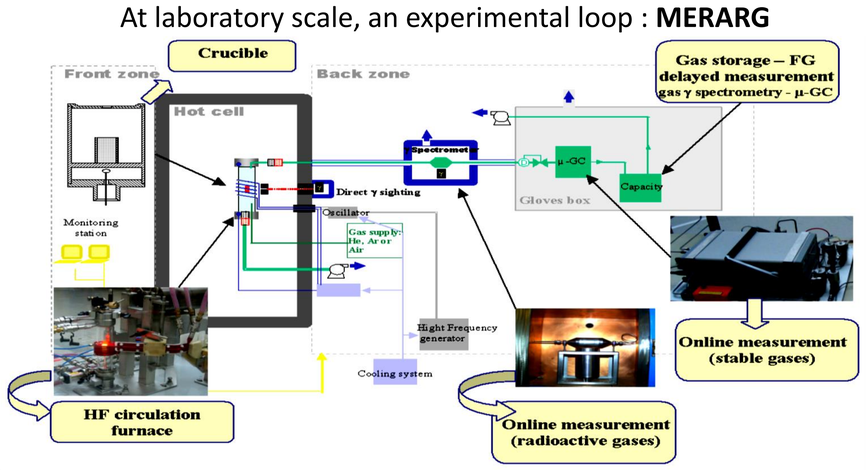
The fission gas release (FGR) is a key point for the nuclear safety that must be accurately assessed under both normal and off-normal conditions. Predicting this release under LOCA type conditions remains an important R&D goal. This work is part of the understanding of the FGR mechanisms.



Thermic under LOCA Type conditions

Which mechanisms promote the source term (i.e. fission gas release) under LOCA type conditions?

At laboratory scale, an experimental loop : **MERARG**



Background

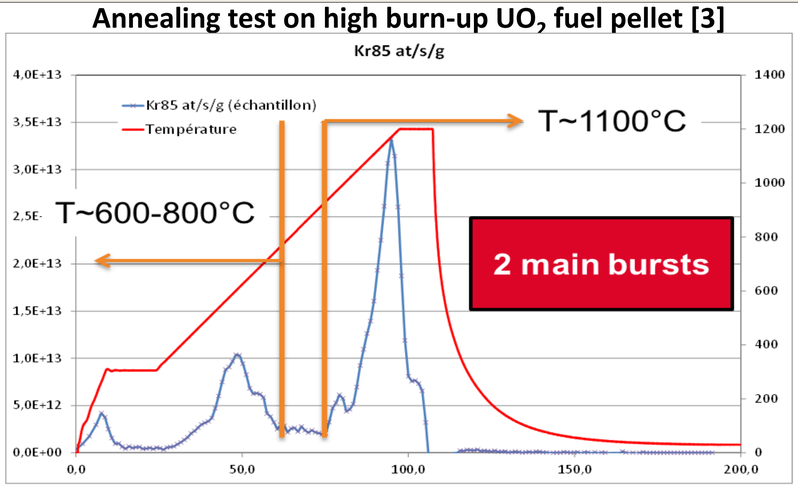
Samples specifications of High burn-up UO_2 Samples

Sample	Burn-up (GWj/t)	Ramp (°C/s)	Annealing Temperature (°C)	Holding time (min)	First burst release (%)	Storage time (d)
1	71.9	0.2	1200	15	4.86	1137
2	76.9	0.2	1200	10	7.04	2627
3	76.9	0.2	1200	10	9.24	3317

- Fission gas release by burst : at about 600°C and at about 1100°C [1].
- The High temperature burst studied a lot : it comes from the edge of the fuel pellet (HBS) [2].
- The release at 600°C cannot be explained by a diffusion mechanism.

Modeling of the first burst data using a **two-step mechanism**: 1) a phase of nucleation of gas clusters (a sort of pre-polymerization) and 2) their growth (similar to a polymerization process).

Annealing test on high burn-up UO, fuel pellet [3]



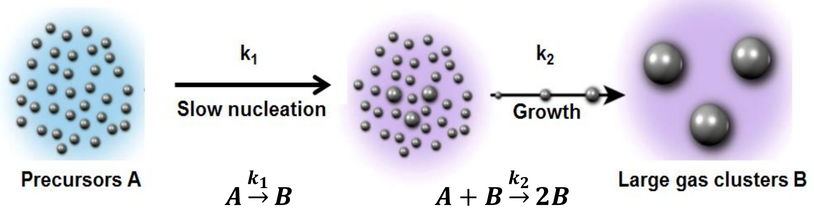
1.M. Marcet, Y. Pontillon, WRFPM/ Top Fuel, 2009.
2. J. Noirot, Y. Pontillon, JNM 446, 163-171 (2014)

What mechanisms could explain the FGR between 500°C and 800°C?

3. Y. Pontillon, J. Bonnin, European Working Group, the Netherlands, 2005.

Model description

Finke-Watzky mechanism [6] [7]



$A \xrightarrow{k_1} B$ $\quad A + B \xrightarrow{k_2} 2B$

Fission gas Release (%)

$$\frac{A_0 - A}{A_0} = 1 - \frac{1 + \frac{k_1}{A_0 k_2}}{1 + \frac{k_1}{A_0 k_2} exp\left[\left(1 + \frac{k_1}{A_0 k_2}\right) A_0 k_2 t\right]}$$

6. A. Watzky, G. Finney JACS 130, 11959-11969 (2008). 7. I. M. Lifshitz, V. V. Slyozov, JPCS 19, 35-50 (1961).

Thermodynamic Driving Force for homogeneous nucleation

Assuming a spherical cluster of radius *r*, an interface energy γ and the specific free energy of a unit volume of the bulk crystal ΔG_v, the total free energy of the crystal can be written :

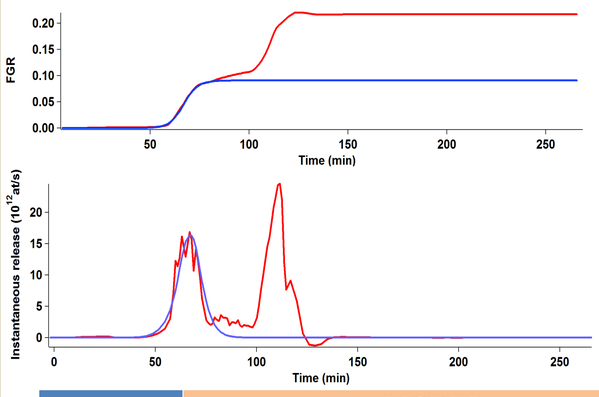
$$\Delta G_v = 4\pi r^2 \gamma + \frac{4}{3}\pi r^3 \frac{k_B T}{v ln\, A_0} \quad \text{then} \quad \Delta G_c = 4\pi\gamma R_c^2 \quad \text{with} \quad R_c = \frac{2\gamma}{\Delta G_V} = \frac{2\gamma v}{k_B T\, ln\, A_0}$$

Therefore, the mechanism of the nucleation rate of *A* gas clusters can be described using an Arrhenius type equation with ΔG_c the total critical free energy :

$$\frac{dA}{dt} \propto exp\left(-\frac{\Delta G_c}{k_B T}\right) \propto exp\left(\frac{16\pi\gamma^3 v^2}{3k_B T^3 (ln\, A_0)^2}\right)$$

It's a function of temperature, of the interface free energy γ and of the gas supersaturation A_0.

Results (model/measurement comparisons)



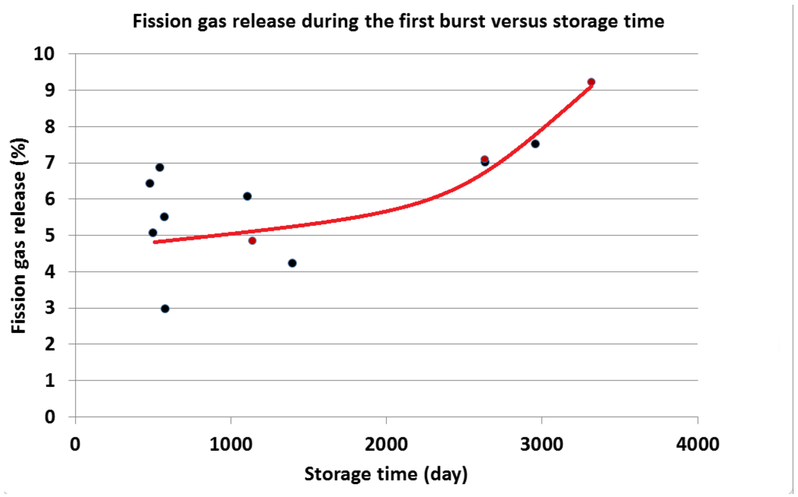
Good agreement between model (blue curves) and experiment (red curves)

- **$[A]_0$ consistent with the experimental first burst release measurements**
- Repeatability on 3 different UO_2 samples : **2 come from the same fuel rod and 1 from another pile**

	MODEL				EXPERIMENT	
Samples	k_1 (10^{-4} s^{-1})	k_2 (s^{-1})	$[A]_0$ (%)	Error (%)	First Burst release (%)	Storage time (d)
1	7.7	3.6	4.4	3.9	4.86	1137
2	8.0	3.0	7.2	1.7	7.04	2627
3	6.2	3.2	9.1	1.8	9.24	3317

Good agreement

FGR vs Storage time?



Red dots are samples use for the modeling

Fission gas release during the first burst in function of the storage time

- Increasing of the first burst FGR (500°C-800°C) with storage time.
- The phenomenon which promote this release **could be the annealing of the defects created by alpha auto-irradiation during the storage** (experiments on model materials in progress to validate this assumption).

Conclusions

A **new mechanism** is proposed to explain **the FGR by burst** in the nuclear fuel. A simplified two-parameter model for describing aggregation kinetics can be borrowed from the Finke-Watzky mechanism. The comparisons between the model and annealing tests show **a good agreement**. This FGR mechanism is possibly connected to **the defects concentration created by alpha self-irradiation**. It's necessary to complete this investigation with other trials using model materials to validate this possible phenomenon.