# 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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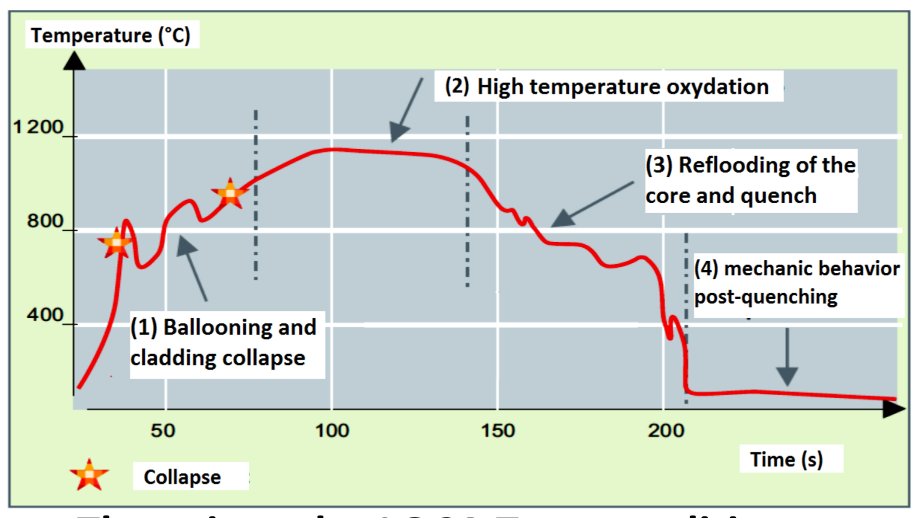
## Context

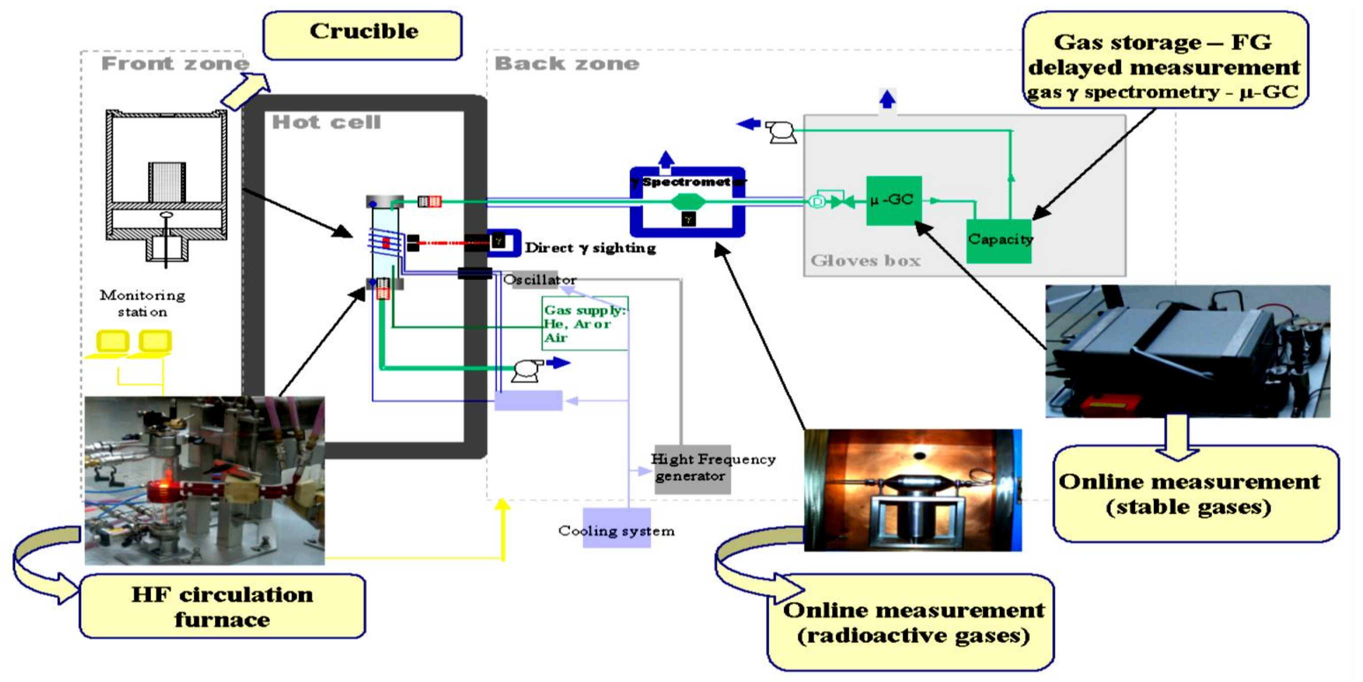
The fission gas release (FGR) is a key point for the nuclear safety that must be accurately assessed under both normal and off-normal conditions. Predicting this release under LOCA type conditions remains an important R&D goal. This work is part of the understanding of the FGR mechanisms.




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

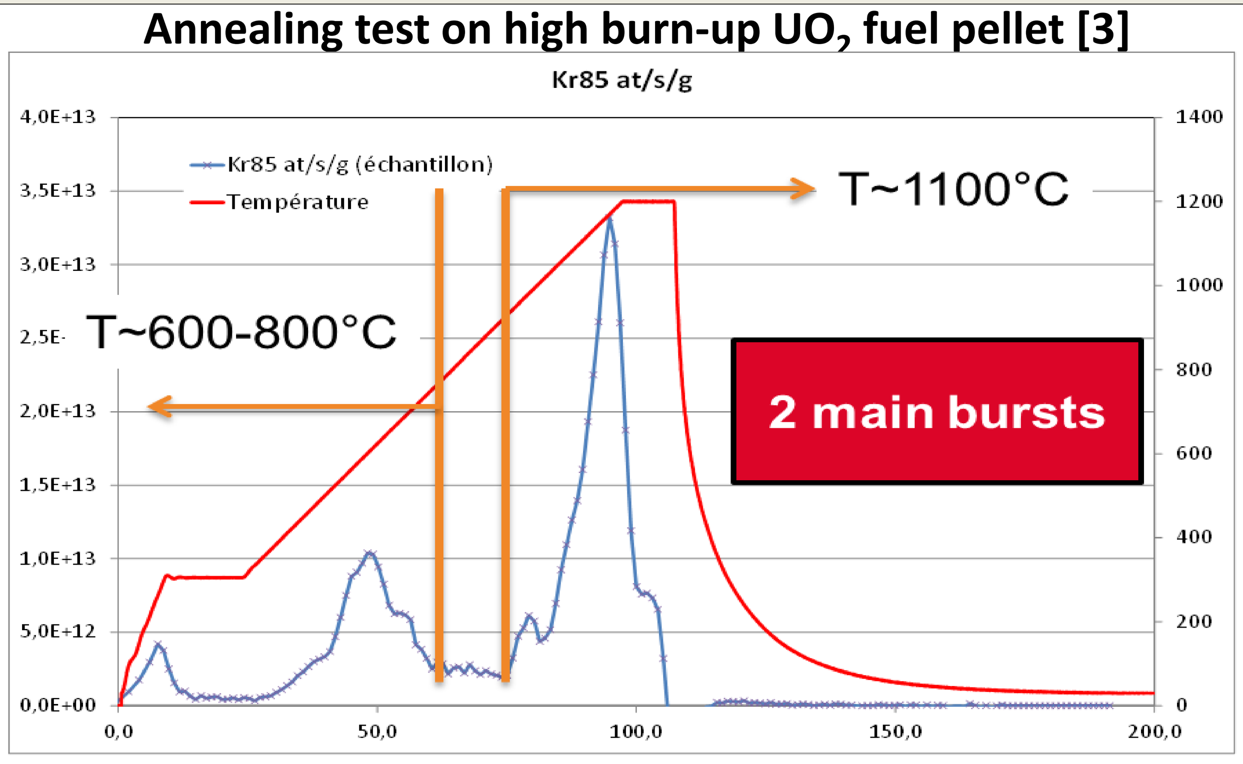
Samples specifications of High burn-up $UO_2$ Samples

| 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 $UO_2$ fuel pellet [3]



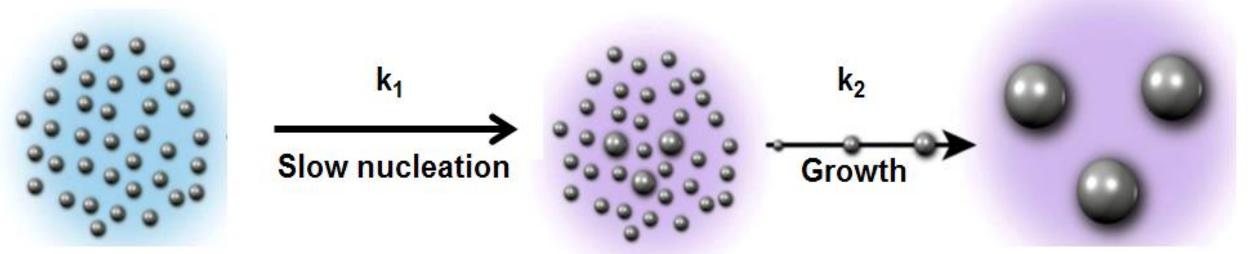

1.M. Marcet, Y. Pontillon, WRFPM/ Top Fuel, 2009.
2. J. Noirot, Y. Pontillon, JNM 446, 163-171 (2014)

**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 $\Delta G_v$, the total free energy of the crystal can be written :

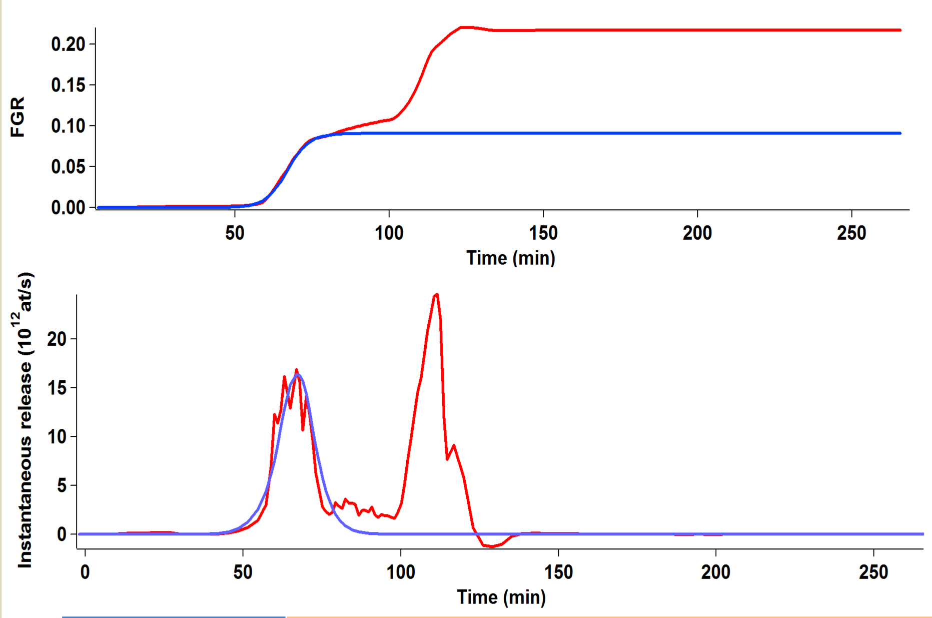
$$\Delta G_v = 4\pi r^2 \gamma + \frac{4}{3}\pi r^3 \frac{k_B T}{v ln A_0} \quad \text{then} \quad \Delta G_c = 4\pi\gamma R_c^2 \quad \text{with} \quad R_c = \frac{2\gamma}{\Delta G_V} = \frac{2\gamma v}{k_B T \ ln A_0}$$

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

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

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

## Results (model/measurement comparisons)



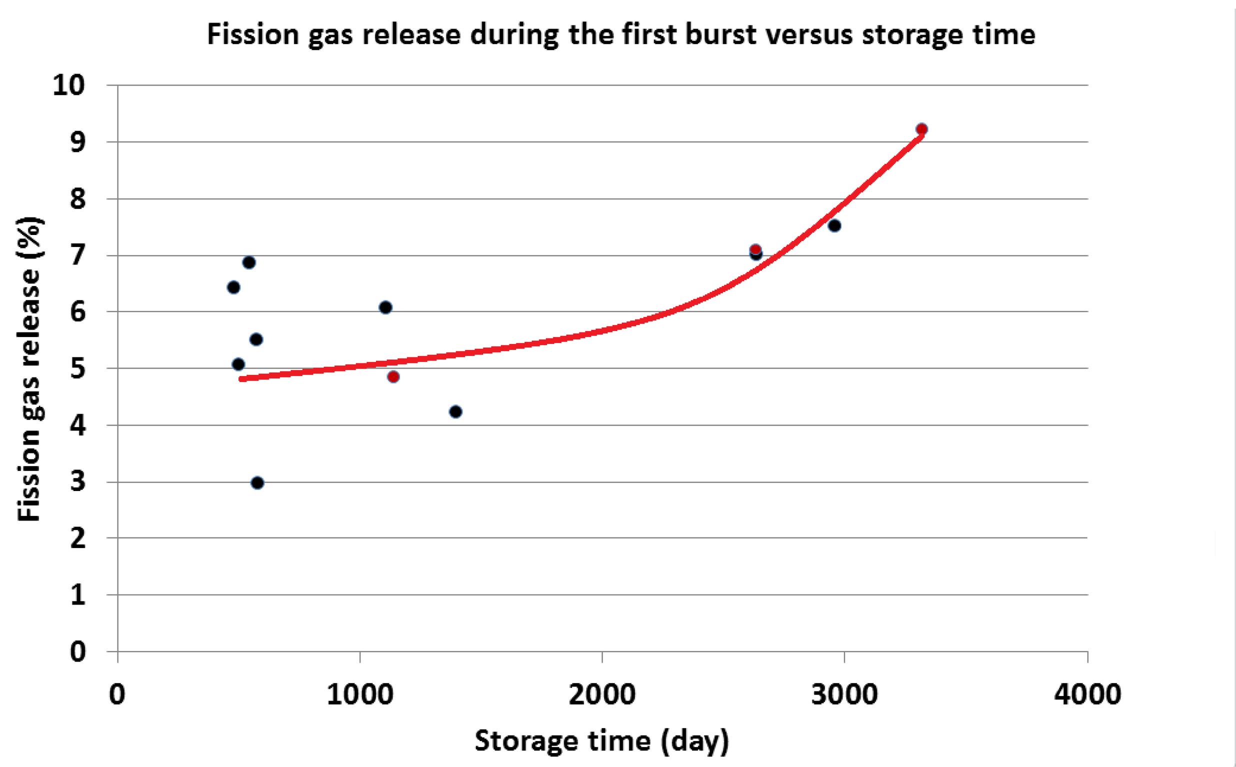

**Good agreement between model (blue curves) and experiment (red curves)**

- **$[A]_0$ 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**

| Samples | MODEL | | | | EXPERIMENT | |
|---|---|---|---|---|---|---|
| | $k_1$ ($10^{-4}$ $s^{-1}$) | $k_2$ ($s^{-1}$) | $[A]_0$ (%) | 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.