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# AN AUTO-CATALYTIC MECHANISM TO MODEL THE FISSION GAS RELEASE BETWEEN 600°C AND 800°C ON UO<sub>2</sub> IRRADIATED FUEL.

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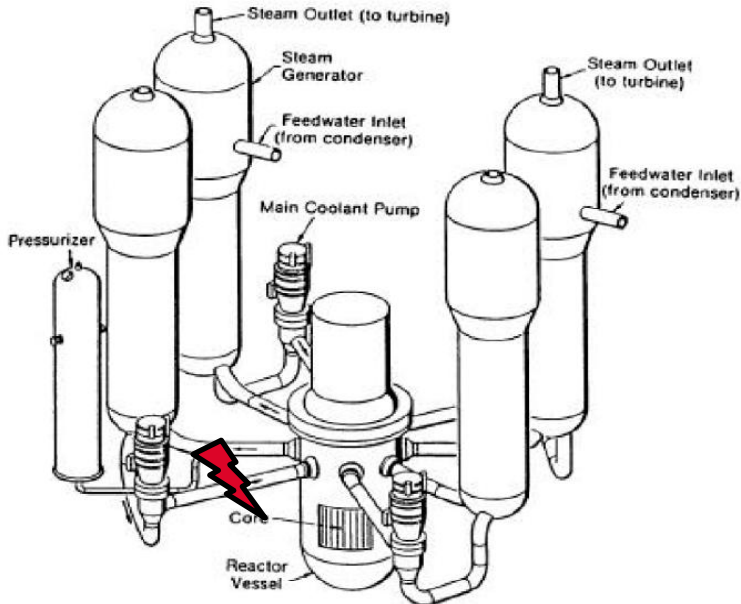
<sup>2</sup>CNRS, SPMS, LRC CARMEN, CENTRALESUPELEC, F-91190 GIF-SUR-

YVETTE, FRANCE

Release of Fission products during a LOCA accident

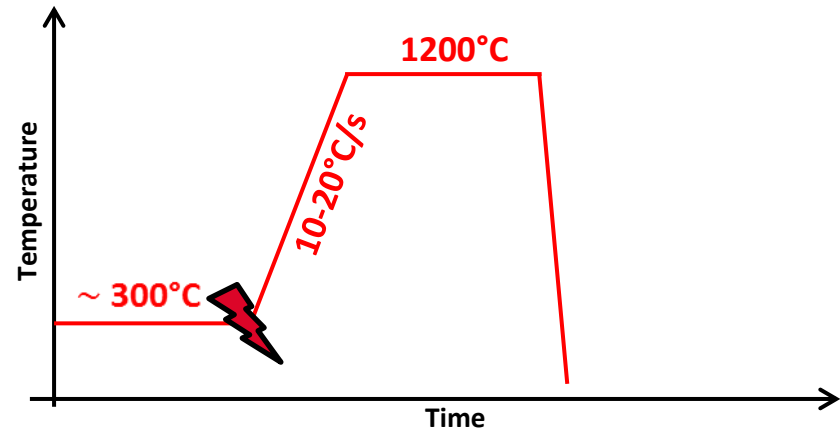
Loss Of Coolant Accident

Breach in primary circuit



Safety assessment requires the inventory of released Fission Products

Average temperature during a LOCA



Fast heating

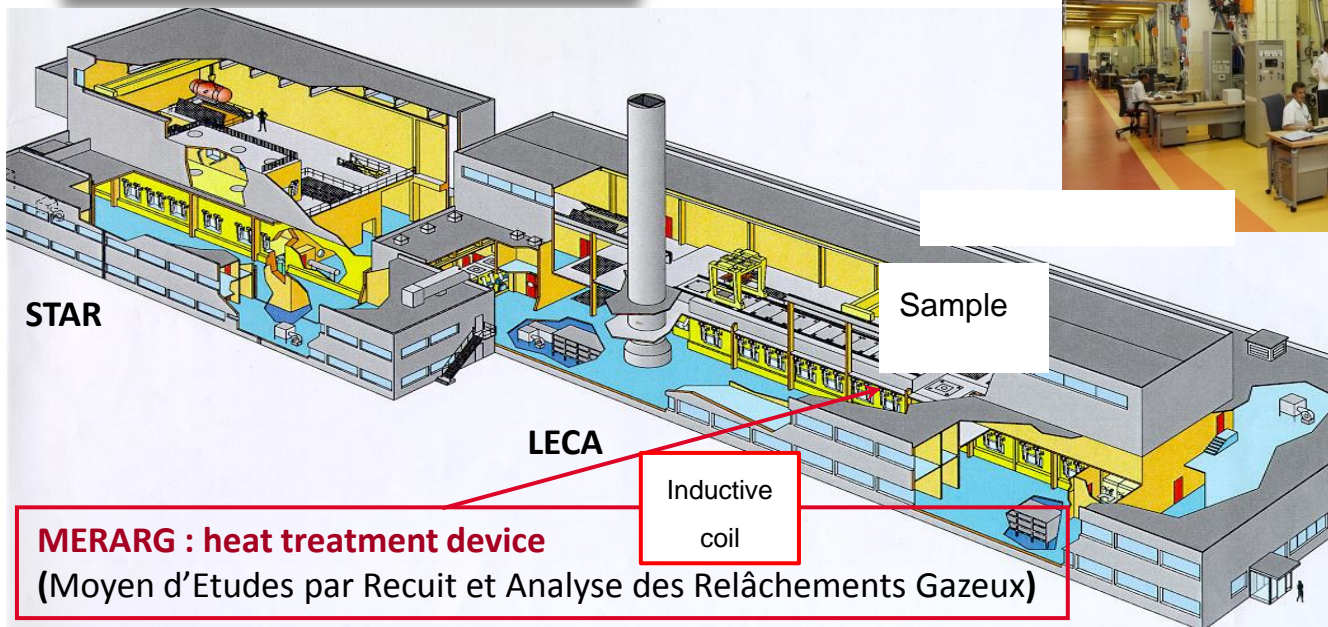
Plateau at  $T_{max} = 1200^{\circ}\text{C}$

Fast cooling due to reflooding

Measurement of Fission Gas release at CEA Cadarache

LECA-STAR  
Hot laboratory

M



Carrier gas and released FPs

Gamma spectrometry  
 $\mu$ -GC



Carrier gas

Fission Gas Release measured using MERARG

Temperature profile for LOCA :

- first plateau at 300°C
- Ramp 20°C/s
- Plateau at 1200°C

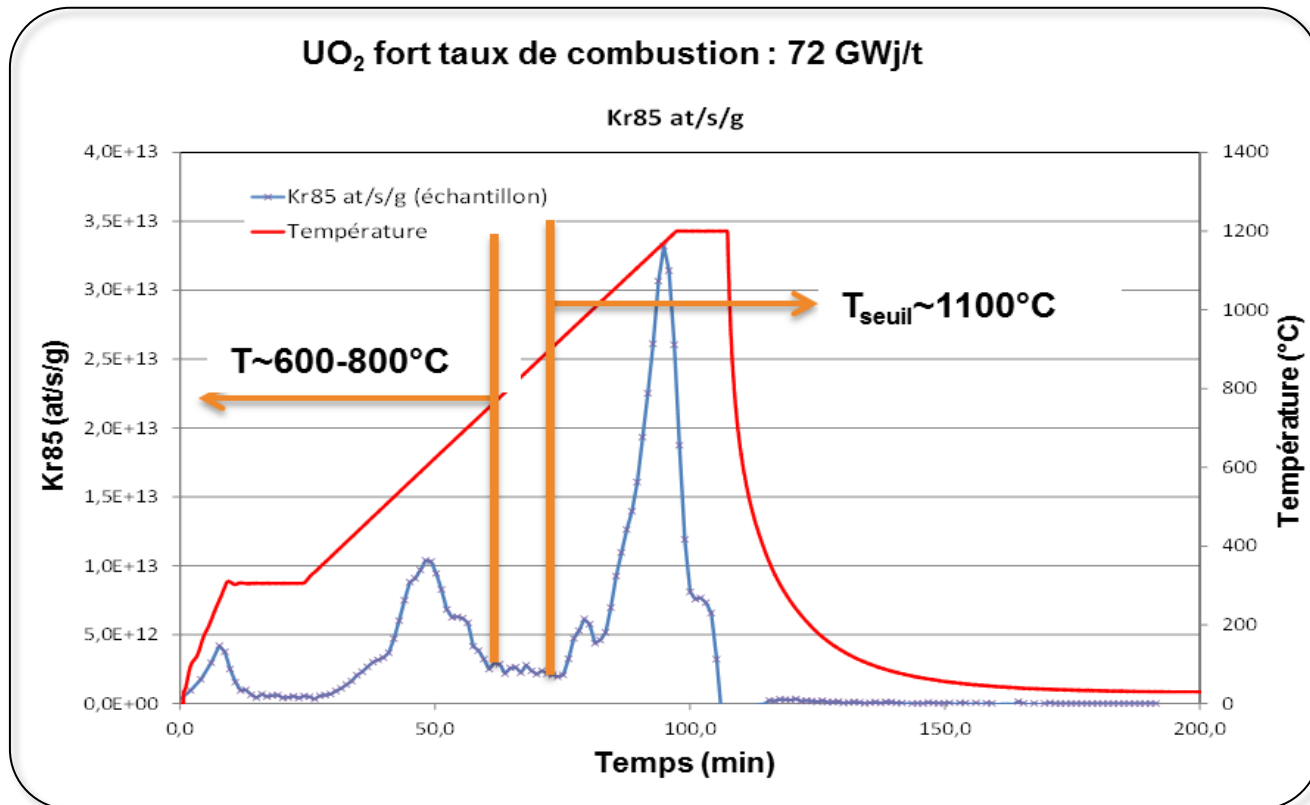


*Too fast in MERARG for mechanism decoupling*



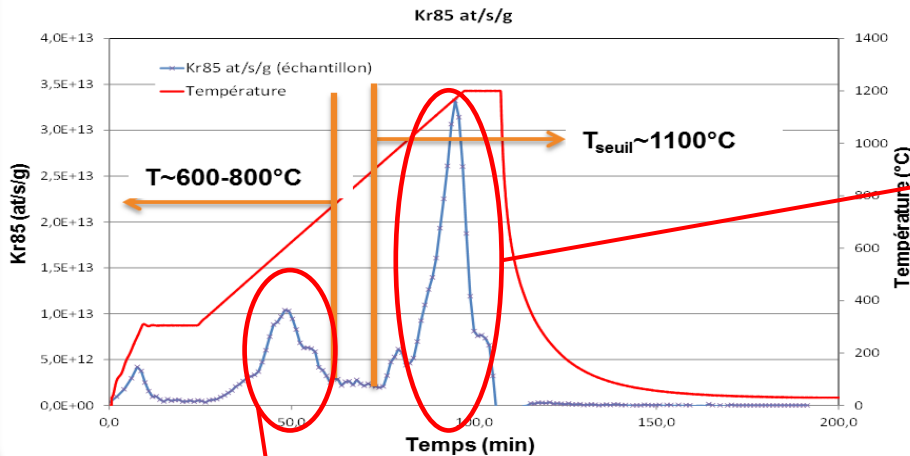
Understanding temperature profile:

- Ramp 0,2°C/s



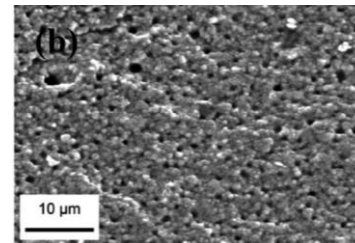
## Bursts of Fission Gas Release

UO<sub>2</sub> fort taux de combustion : 72 GWj/t



### Comes from High Burn-up Structure

J. Noiroot , Y. Pontillon, *JNM 446,163-171, 2014*



Proposed mechanism:

- Highly pressurized bubbles of HBS release the gas they contain

Up to now no validated mechanism is proposed for the burst at 600-800°C

An analysis of the experimental data set obtained with MERARG in order to evidence some trends :

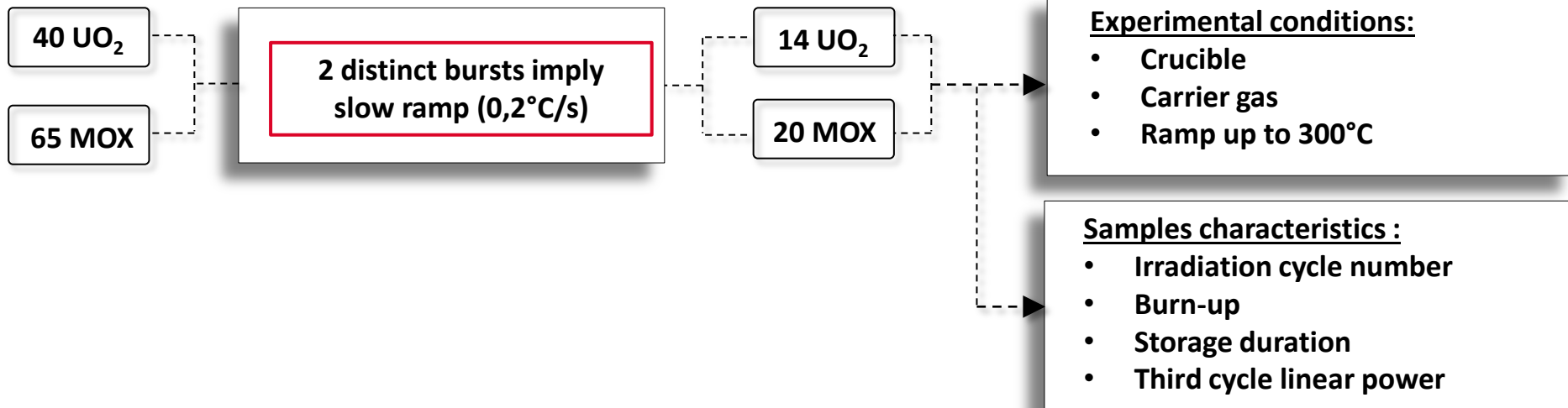
**Correlation between the burst at 600-800°C and experimental parameters ??**

## Parameters of MERARG data set

➤ From November 2000 up to nowadays more than 200 heat treatments in MERARG

Which ones are relevant?

- Temperature profiles related to LOCA
- First plateau at 300°C and then temperature ramp in inert atmosphere.





14 UO<sub>2</sub>

~~20 MOX~~

Experimental conditions:

- Crucible
- Carrier gas
- Ramp up to 300°C



No influence of experimental conditions

Samples characteristics :

BU (GWj/tM)	Storage (d)	1 <sup>st</sup> burst (%)
76,94	1137	4,8
	2627	6,0
	3317	9,2

no correlation for MOX

Gas release in %

:

5

Nombre de cycles

6

7

8

0

2

50

taux de combustion (GWj/tM)

60

70

80

0

12

Gas rel

4

2

Strong correlation between FGR and storage duration for UO<sub>2</sub>

Storage time in days

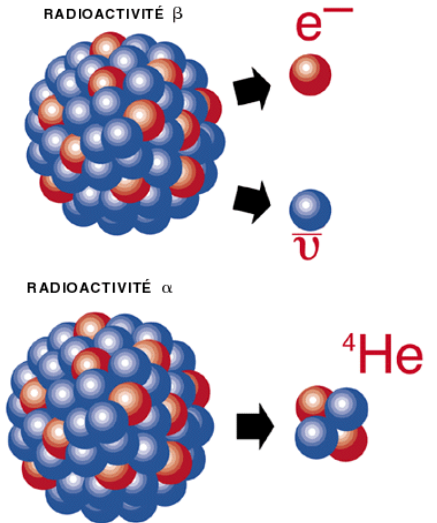
0 195 200  
N/cm)



What happen during storage duration ?

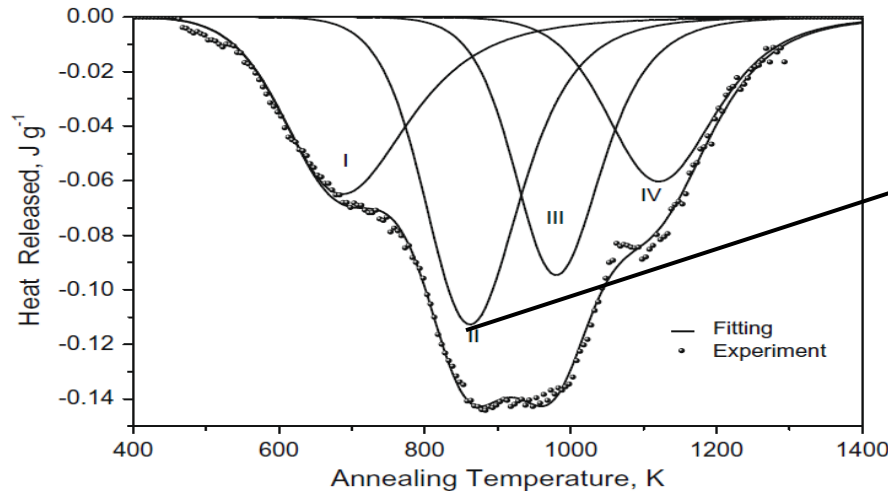
Storage at low temperature :

- No thermally activated diffusion
- Creation of defects due to self-irradiation
- Defects that can be annealed by heat treatment



Annealing kinetics of self irradiation defects in Pu doped  $UO_2$

Staicu & al., JNM 397, 2010



600-800°C :  
Annealing of U vacancies [1][2]



Fission gas release :  
A mechanism derived from  
autocatalytic of uranium vacancies ?

[1] H. Matzke, *Radiation-Damage in Nuclear-Materials*, Mar. 1992.

[2] H. Labrim & al. Aug. 2007.

## A tentative modeling of Fission Gas release in MERARG

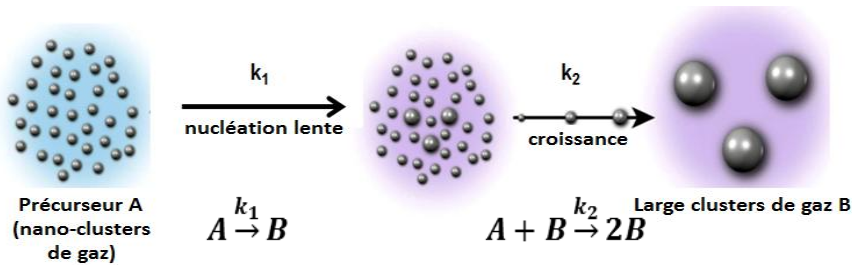
Simplified mechanism with 2 steps (Finke-Watzky type)

Formation of nano-clusters in steels

Evolution of nano-particles in colloidal solutions<sup>2</sup>

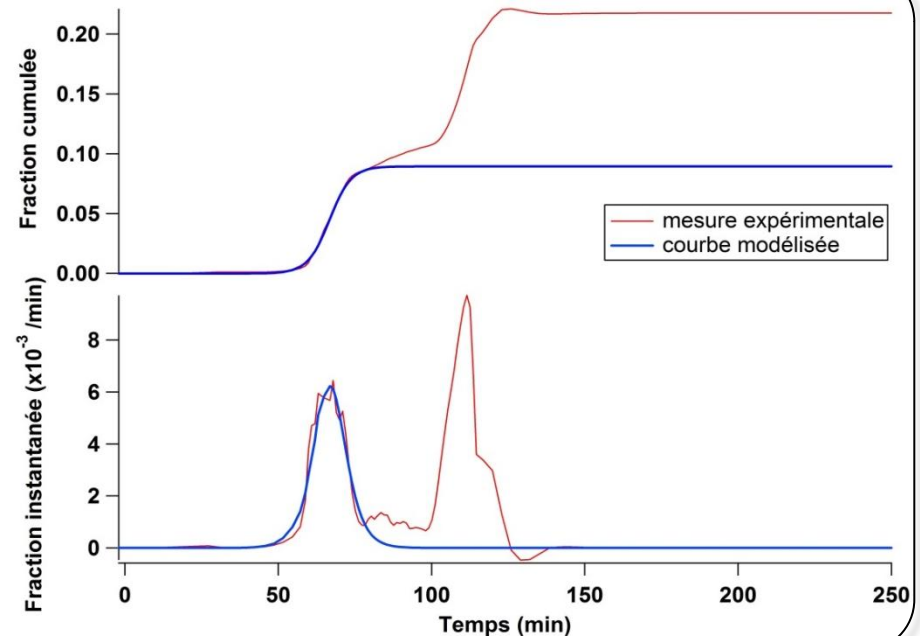
### Modeling for auto-catalytic kinetics

Brindelle, Numat2016



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

$A_0$ : available gas capacity for release



$$cumulated\ fraction = \frac{\text{released gas quantity}}{\text{initial gas quantity}}$$

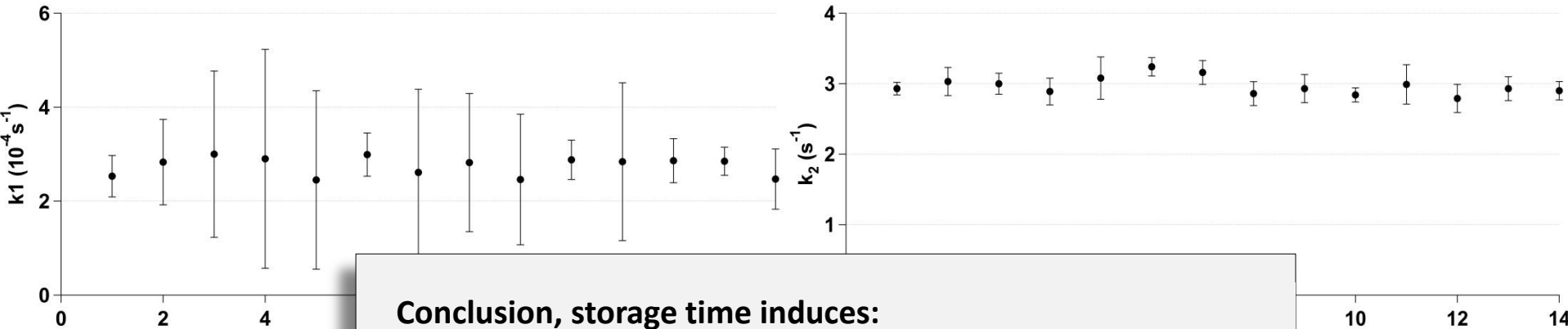
<sup>1</sup>M. A. Watzky & al., *Am. Chem. Soc.*, vol. 130, no. 36, pp. 11959–11969, Sep. 2008.

<sup>2</sup>D. V. Talapin & al., *J. Phys. Chem. B*, vol. 105, no. 49, pp. 12278–12285, Dec. 2001.

Comparison modeling/measurements

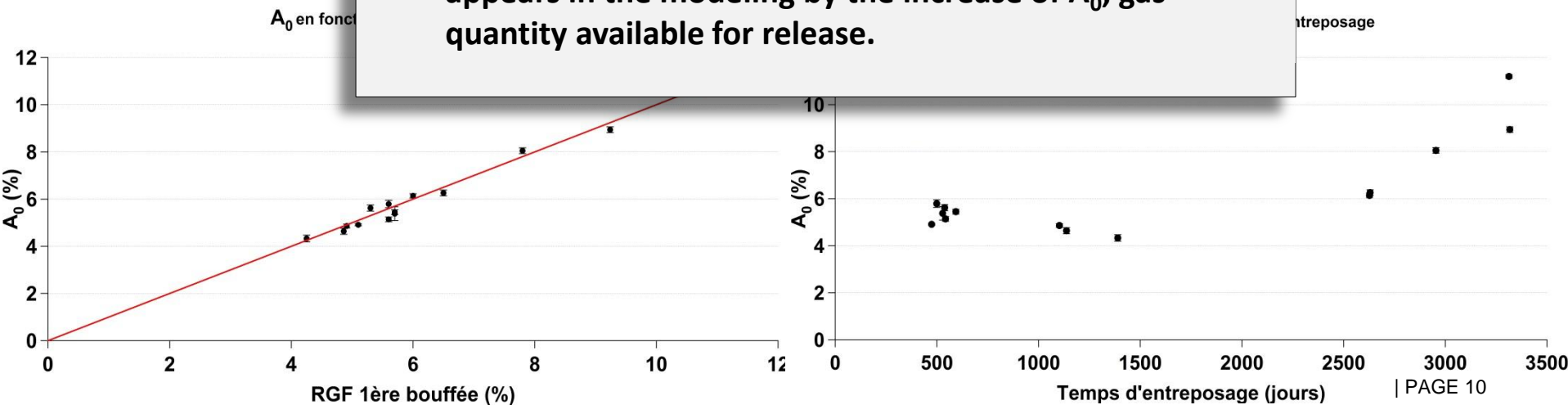
$k_{1\text{moy}} = 2,7 \pm 0,11 \cdot 10^{-4} \text{ min}^{-1}$

$k_{2\text{moy}} = 2,97 \pm 0,07 \text{ min}^{-1}$



Conclusion, storage time induces:

- An increase of gas release at 600-800°C burst that appears in the modeling by the increase of  $A_0$ , gas quantity available for release.



## Conclusion

The quantity of gas release during the burst at 600-800°C depends on storage time.

The release mechanism could be due to the annealing of self-irradiation defects that were produced during storage. A kinetic model is proposed to describe this release mechanism.

The storage time is a first order parameter for gas release experiment that needs to be taken into account.

# Thank You for your attention

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