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AN AUTO-CATALYTIC MECHANISM TO MODEL THE FISSION GAS RELEASE BETWEEN 600°C AND 800°C ON UO₂ IRRADIATED FUEL.

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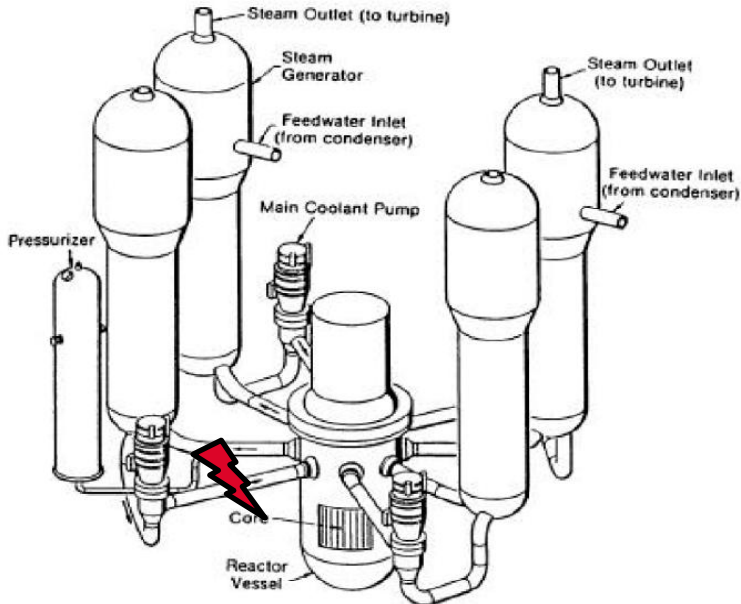
²CNRS, SPMS, LRC CARMEN, CENTRALESUPELEC, F-91190 GIF-SUR-

YVETTE, FRANCE

Release of Fission products during a LOCA accident

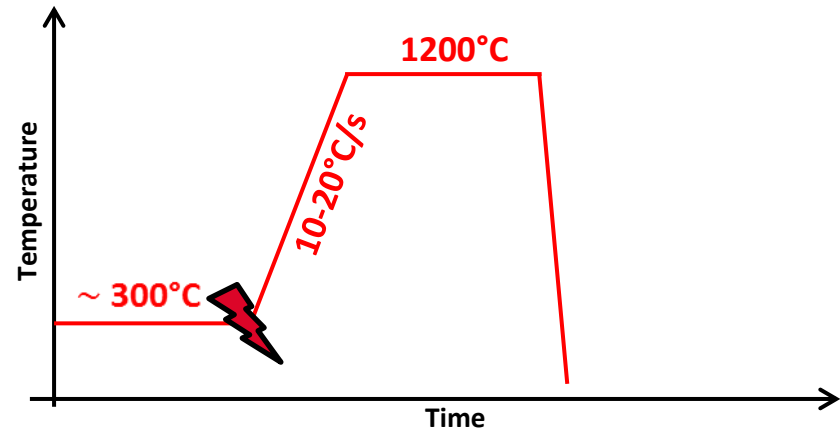
Loss Of Coolant Accident

Breach in primary circuit



Safety assessment requires the inventory of released Fission Products

Average temperature during a LOCA



Fast heating

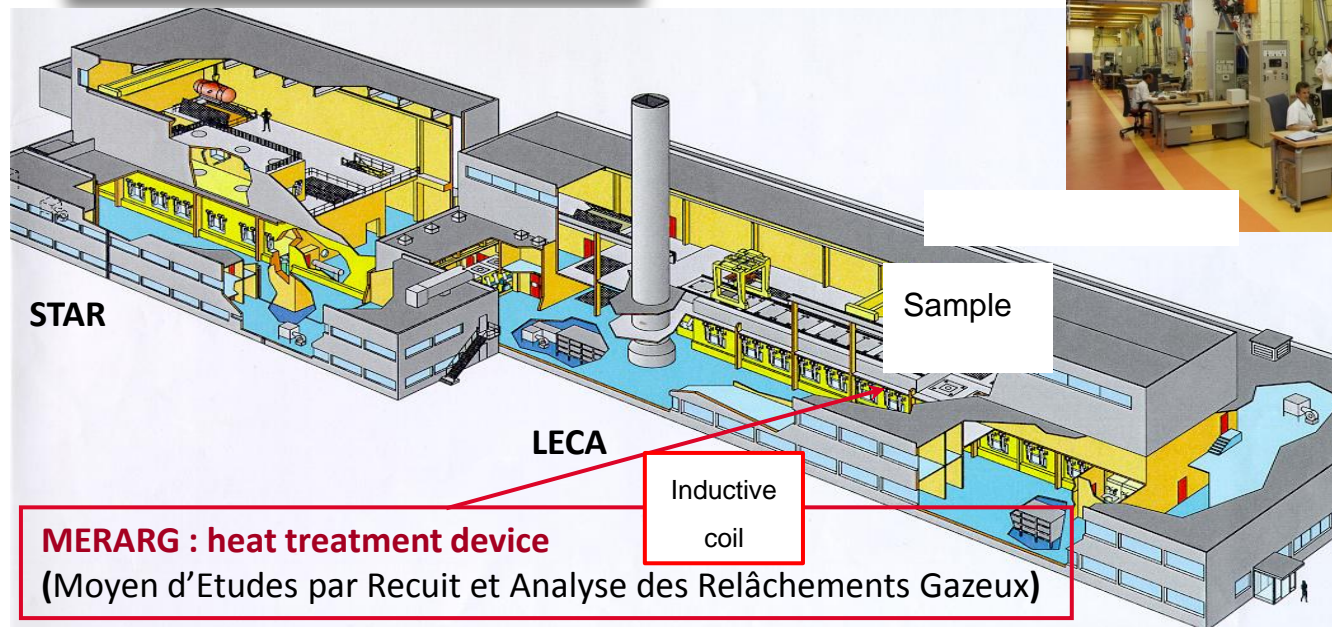
Plateau at $T_{max} = 1200^{\circ}\text{C}$

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
 μ -GC



Carrier gas

Fission Gas Release measured using MERARG

Temperature profile for LOCA :

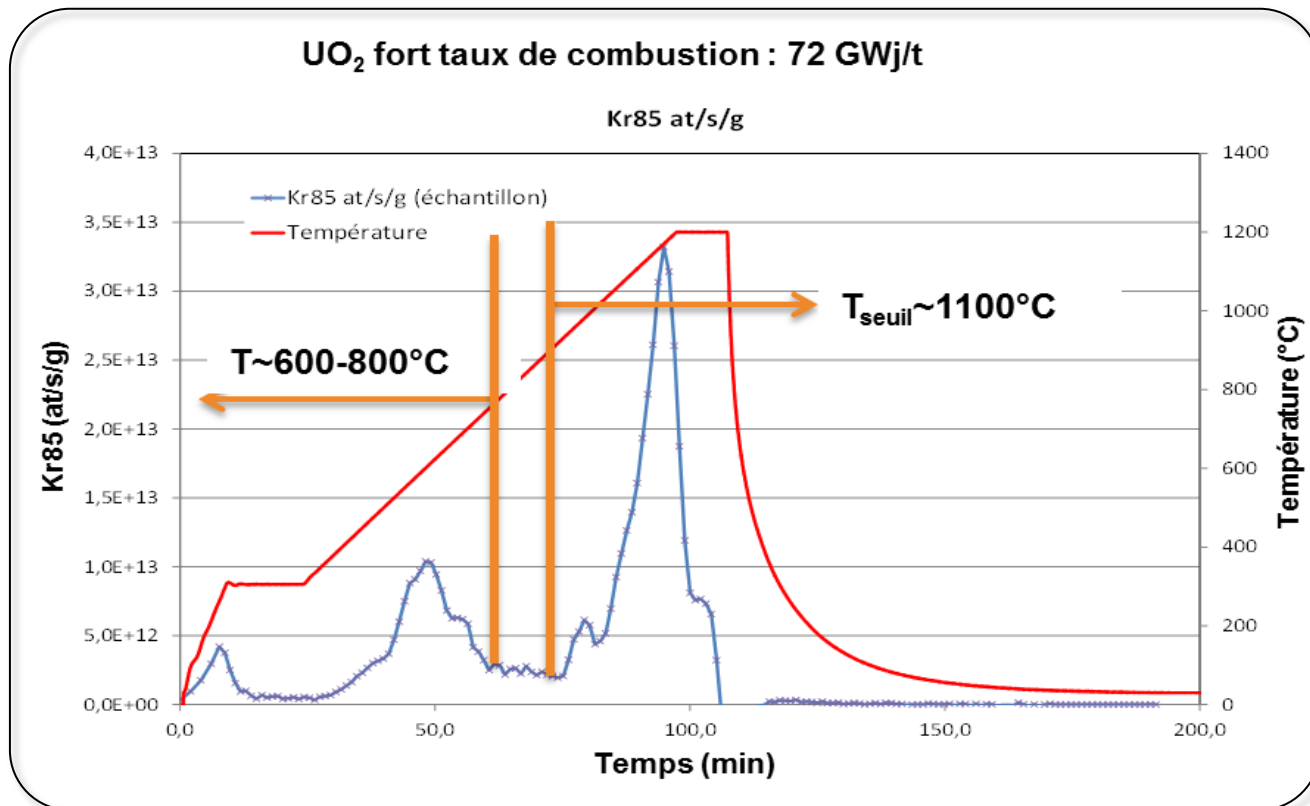
- first plateau at 300°C
- Ramp 20°C/s
- Plateau at 1200°C



Too fast in MERARG for mechanism decoupling

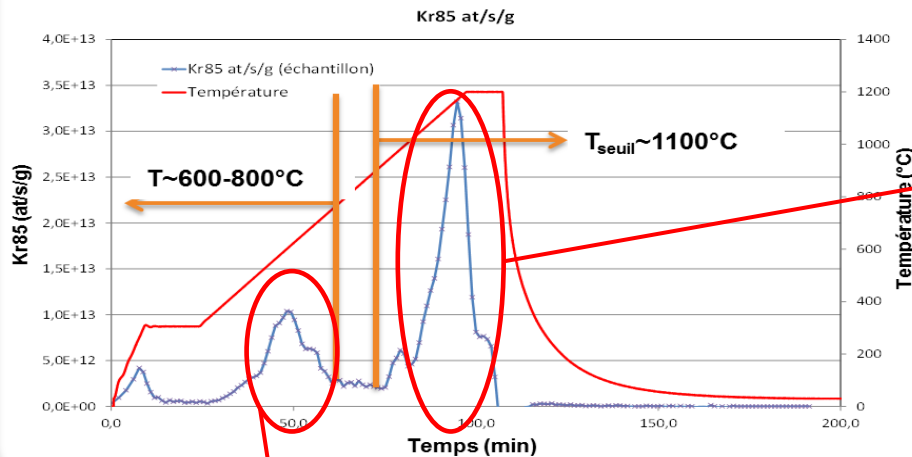


Understanding temperature profile:
• Ramp 0,2°C/s



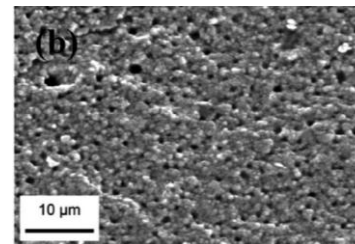
Bursts of Fission Gas Release

UO₂ fort taux de combustion : 72 GWj/t



Comes from High Burn-up Structure

J. Noirod , 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.

40 UO₂

65 MOX

2 distinct bursts imply
slow ramp (0,2°C/s)

14 UO₂

20 MOX

Experimental conditions:

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

Samples characteristics :

- Irradiation cycle number
- Burn-up
- Storage duration
- Third cycle linear power

14 UO₂

~~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 st burst (%)
76,94	1137	4,8
	2627	6,0
	3317	9,2

no correlation for MOX

Gas release in %

:

5

Nombre de cycles

7

8

0

2

50

taux de combustion (GWj/tM)

60

70

80

12

Strong correlation between FGR and storage duration for UO₂

Gas rel

4

2

0

0

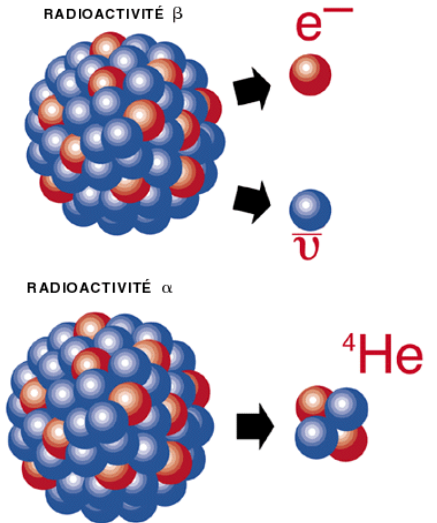
Storage time in days

0 195 200
N/cm)

What happen during storage duration ?

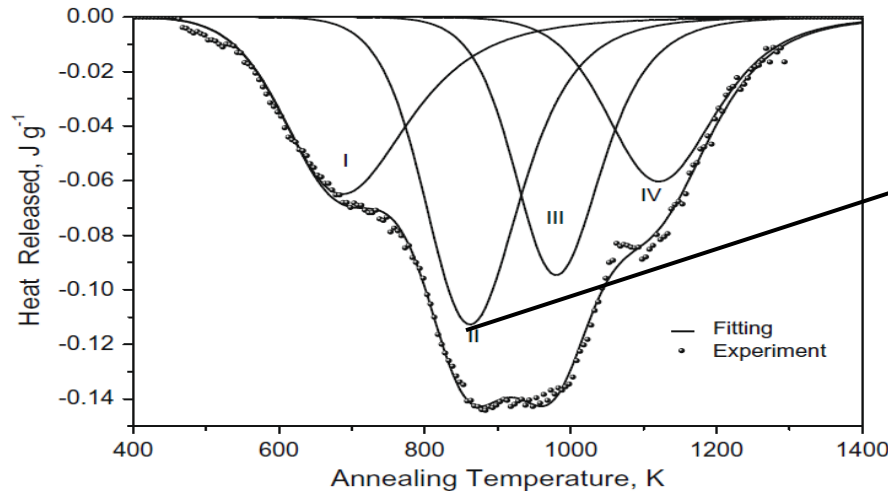
Storage at low temperature :

- 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

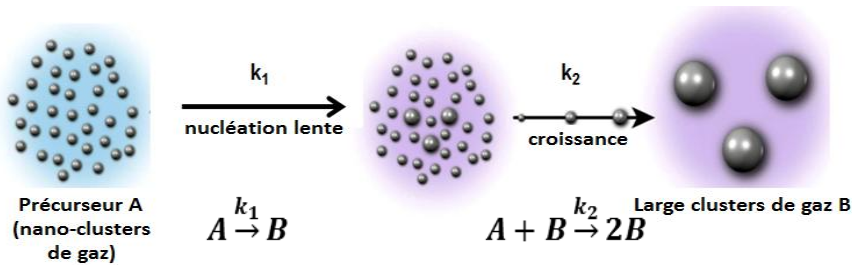
Simplified mechanism with 2 steps (Finke-Watzky type)

Formation of nano-clusters in steels

Evolution of nano-particles in colloidal solutions²

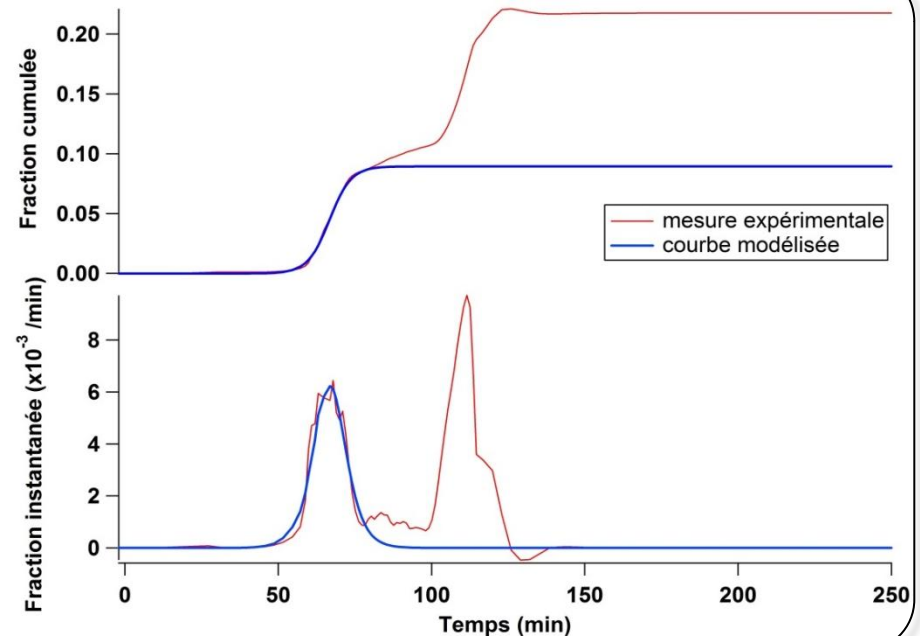
Modeling for auto-catalytic kinetics

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



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

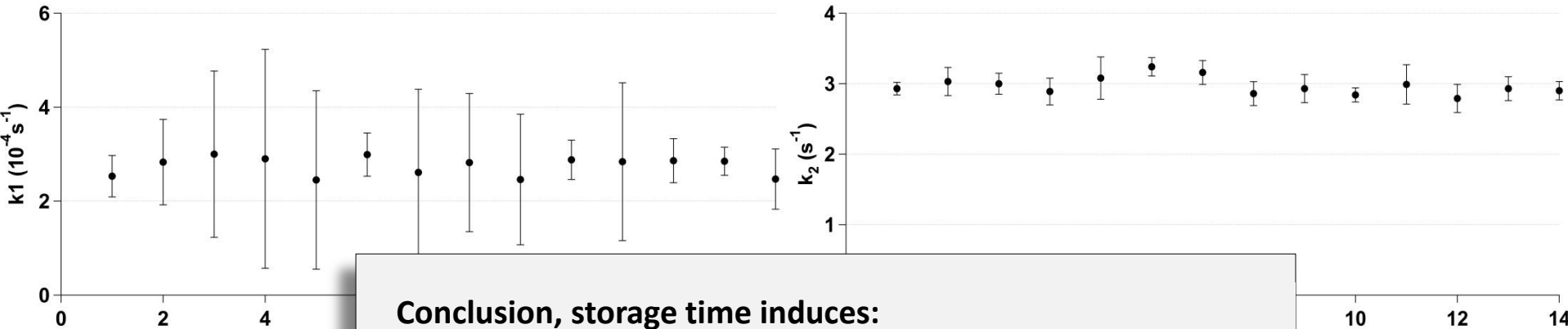
¹M. A. Watzky & al., *Am. Chem. Soc.*, vol. 130, no. 36, pp. 11959–11969, Sep. 2008.

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

Comparison modeling/measurements

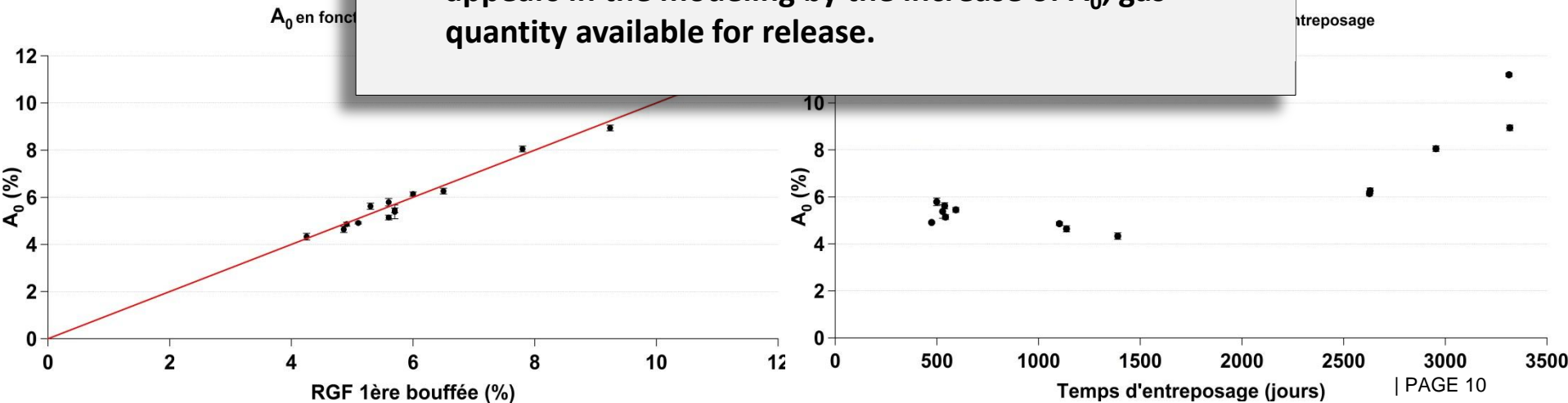
$k_{1\text{moy}} = 2,7 \pm 0,11 \cdot 10^{-4} \text{ min}^{-1}$

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



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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