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AN AUTO-CATALYTIC MECHANISM TO MODEL THE FISSION GAS RELEASE BETWEEN 600°C AND 800°C ON UO_2 IRRADIATED FUEL.

YVES PONTILLON[1], GUILLAUME BRINDELLE[1, 2], GIANGUIDO BALDINOZZI[2], HÉLÈNE CAPDEVILA[1], LIONEL DESGRANGES[1]

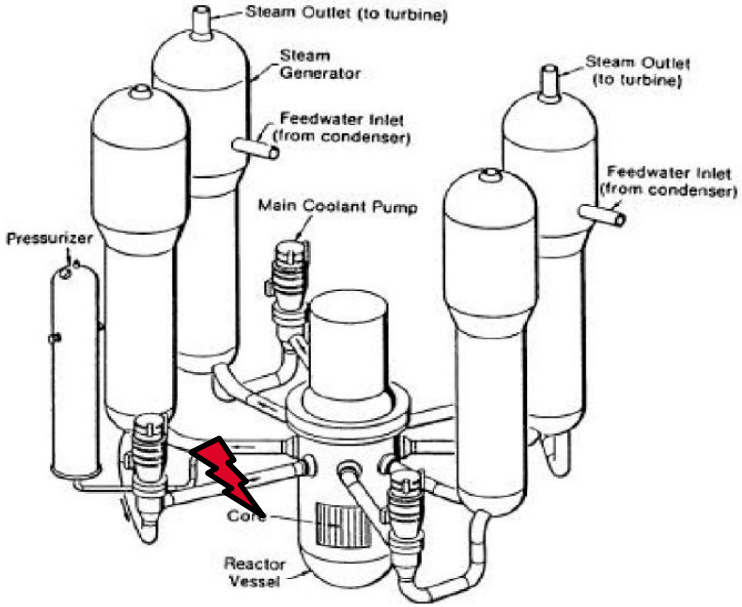
[1]CEA, DEN/DEC, F-13108 SAINT-PAUL-LEZ-DURANCE, FRANC

[2]CNRS, SPMS, LRC CARMEN, CENTRALESUPELEC, F-91190 GIF-SUR-YVETTE, FRANCE

Release of Fission products during a LOCA accident

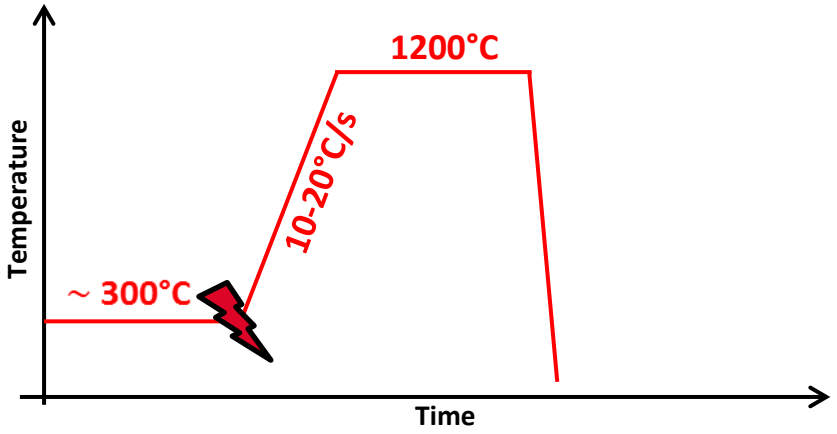
Loss Of Coolant Accident

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°C

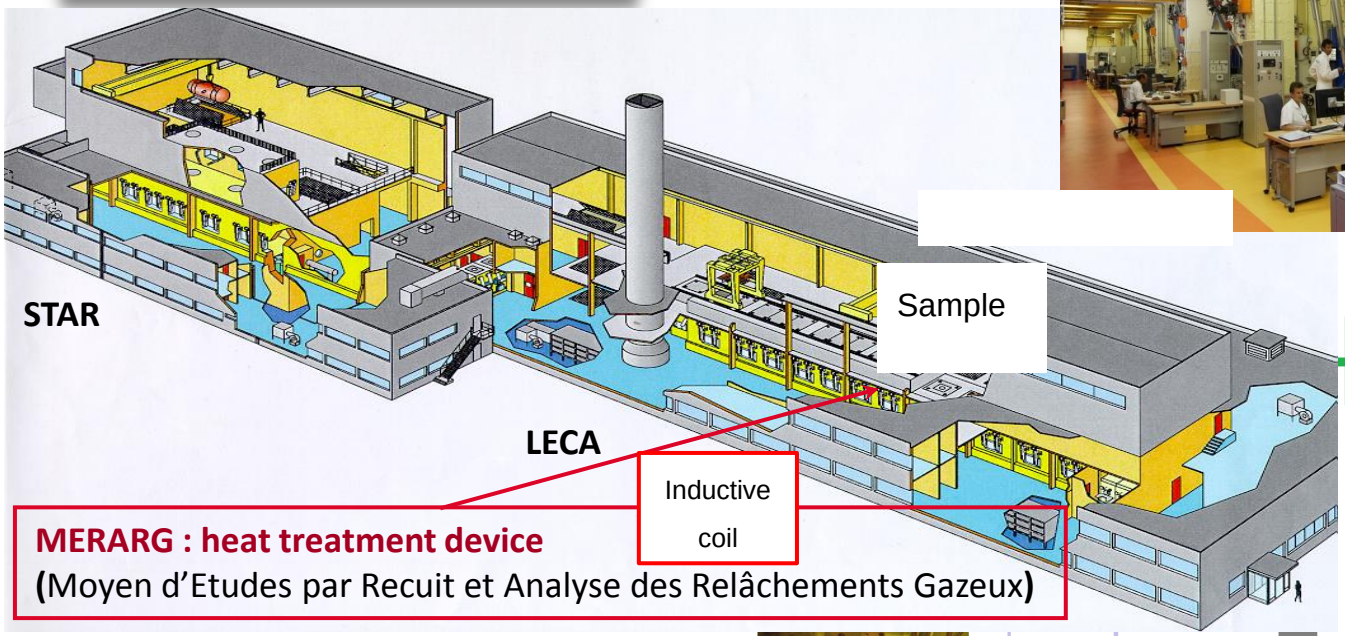
Fast cooling due to reflooding

NEA, OECD, Nuclear Fuel Behaviour in Loss-of-coolant Accident (LOCA) Conditions, State-of-the-art Report n°6846, 2009

Measurement of Fission Gas release at CEA Cadarache

LECA-STAR
Hot laboratory



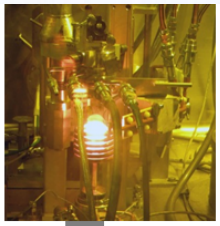


MERARG : heat treatment device
(Moyen d'Etudes par Recuit et Analyse des Relâchements Gazeux**)**

Carrier gas and released FPs

Gamma spectrometry
µ-GC





Carrier gas

Y. Pontillon, et al., J. Nucl. Mater. vol 385, issue 1, 2009, 137

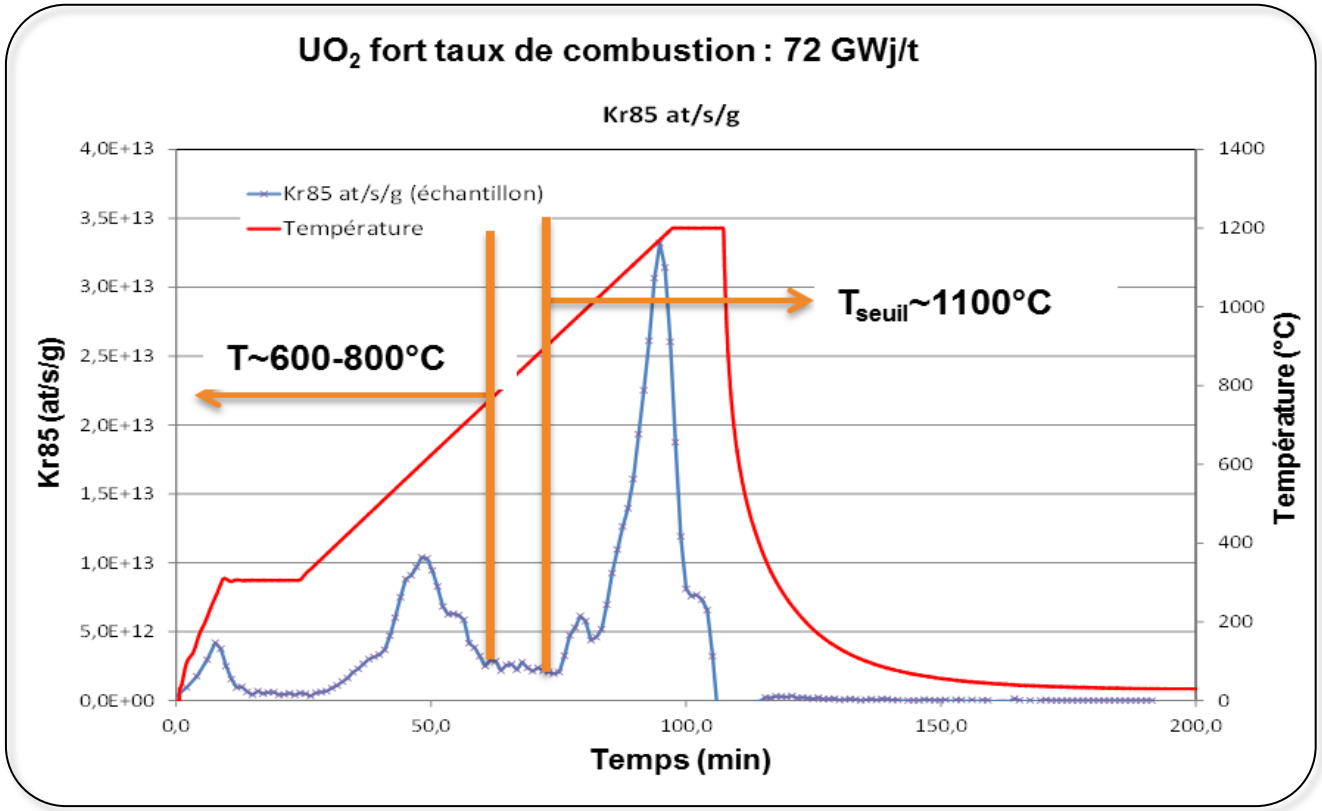
Fission Gas Release measured using MERARG

Temperature profile for LOCA :
- **first plateau at 300°C**
- **Ramp 20°C/s**
- **Plateau at 1200°C**

→ *Too fast in MERARG for mechanism decoupling* →

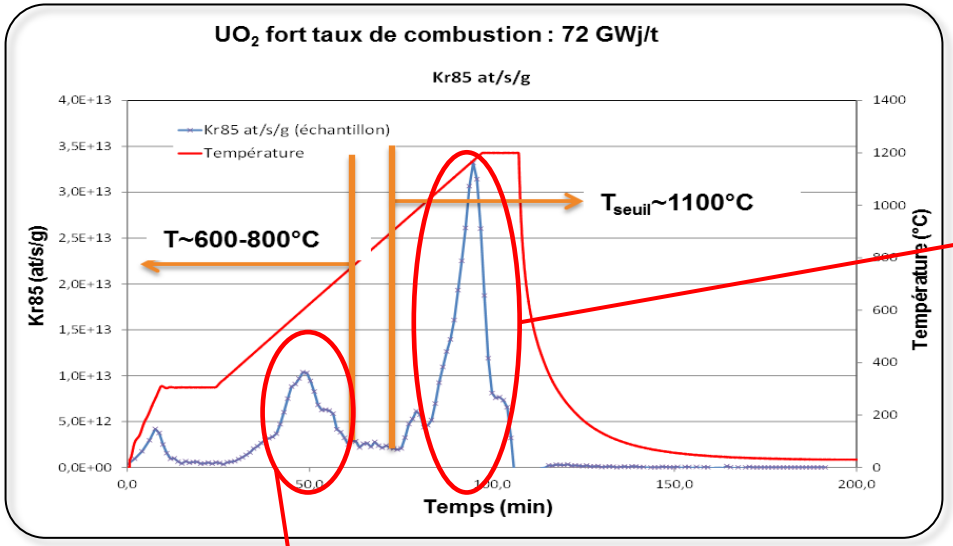
Understanding temperature profile:
- **Ramp 0,2°C/s**

→



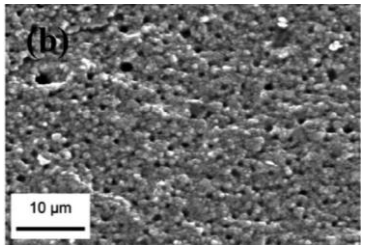
Y. Pontillon et al. Top Fuel 2004, 1025

Bursts of Fission Gas Release



Comes from High Burn-up Structure

J. Noirot , 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 ??

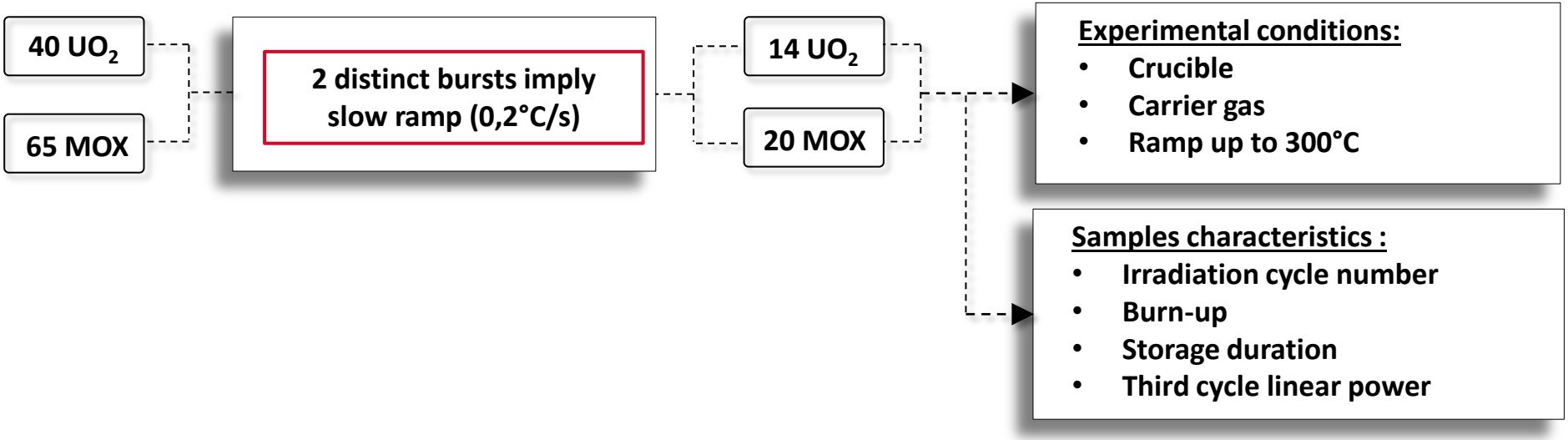
Y. Pontillon et al. Top Fuel 2004, 1025

Parameters of MERARG data set

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

<u>Which ones are relevant?</u>

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



14 UO_2

~~20 MOX~~

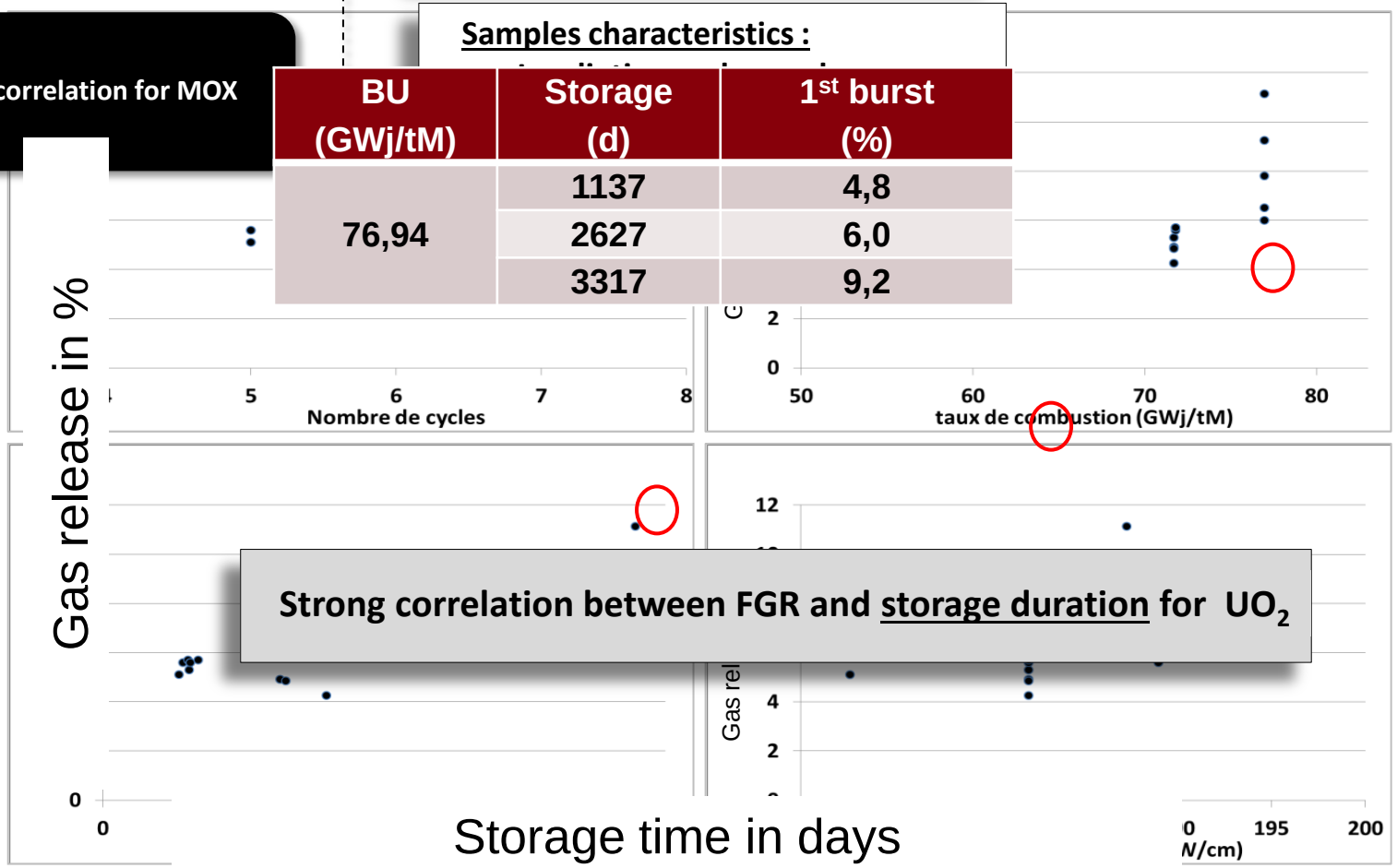
Experimental conditions:
- Crucible
- Carrier gas
- Ramp up to 300°C

No influence of experimental conditions

no correlation for MOX

Samples characteristics :

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

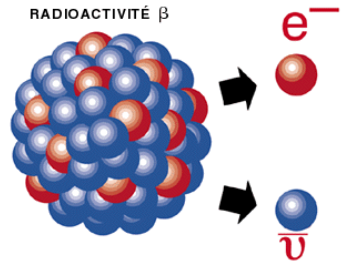


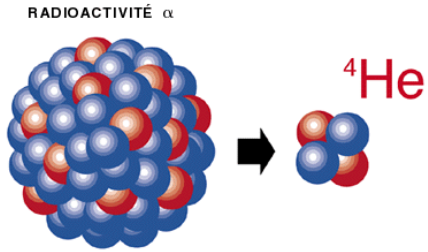
Strong correlation between FGR and storage duration for UO_2

What happen during storage duration ?

Storage at low temperature :

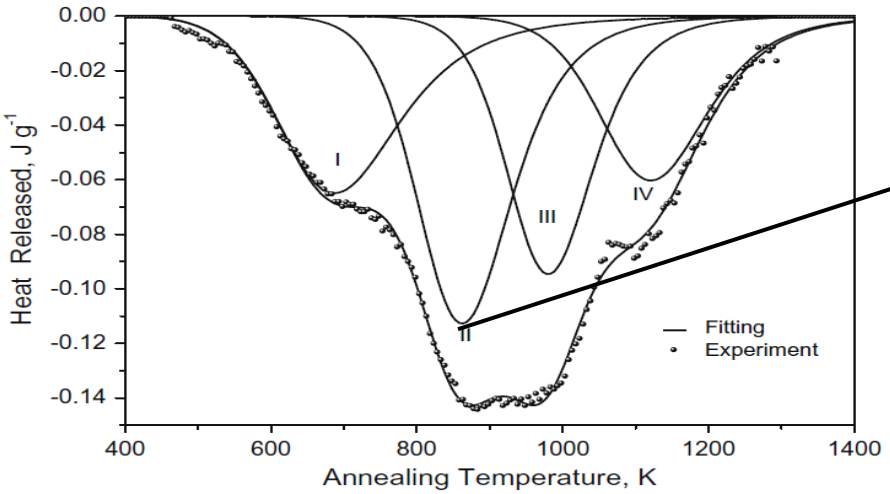
- No thermally activated diffusion
- Creation of defects due to self-irradiation
- Defects that can be annealed by heat treatment





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 ?

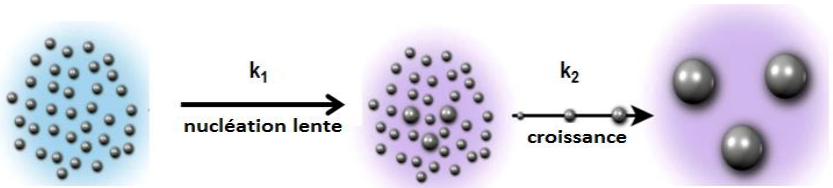
[1] H. Matzke, *Radiation-Damage in Nuclear-Materials*, Mar. 1992.
[2] H. Labrim & al. Aug. 2007.

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-particules in colloidal solutions [2]**

Modeling for auto-catalytic kinetics

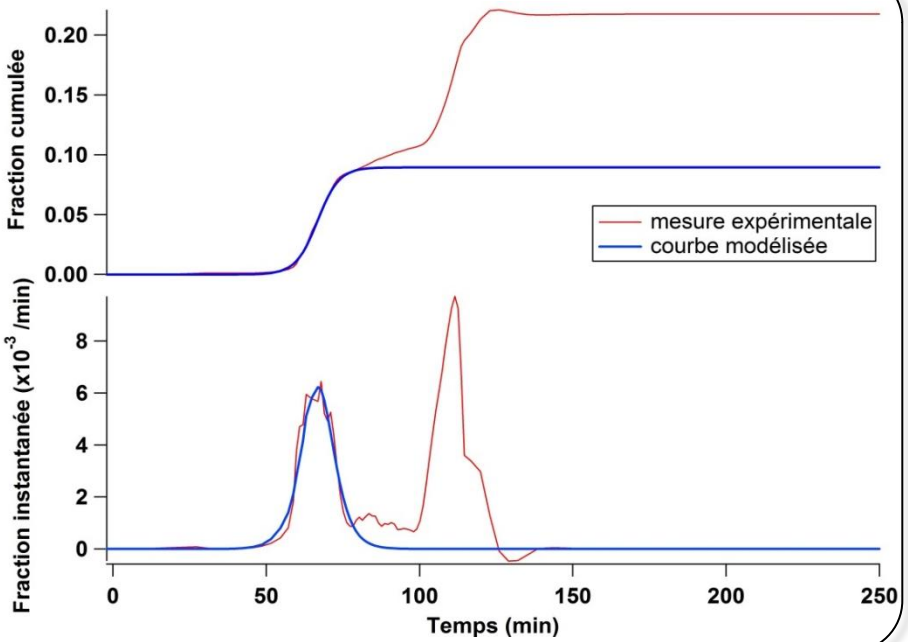
Brindelle, Numat2016



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

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

A_0 : available gas capacity for release

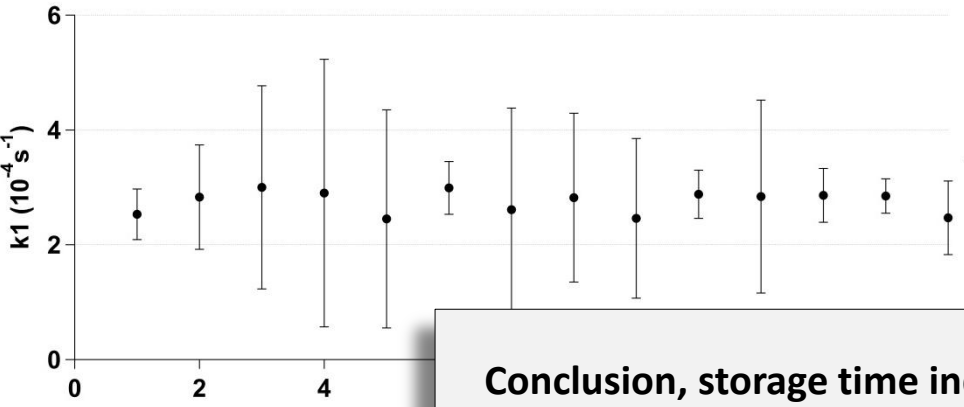


$$cumulated fraction = \frac{\text{released gas quantity}}{\text{initial gas quantity}}$$

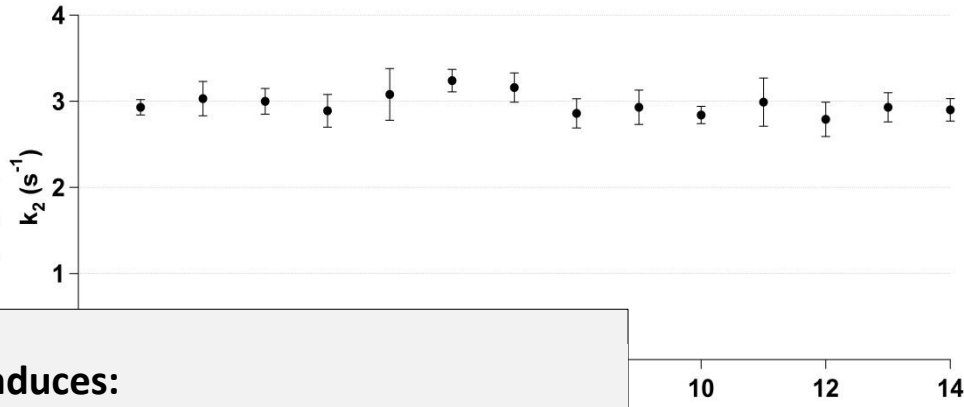
[1] M. A. Watzky & al., *Am. Chem. Soc.*, vol. 130, no. 36, pp. 11959–11969, Sep. 2008.
[2] D. V. Talapin & al., *J. Phys. Chem. B*, vol. 105, no. 49, pp. 12278–12285, Dec. 2001.

Comparison modeling/measurements

k_{1moy}= 2,7 ± 0,11 .10^{-4} min^{-1}

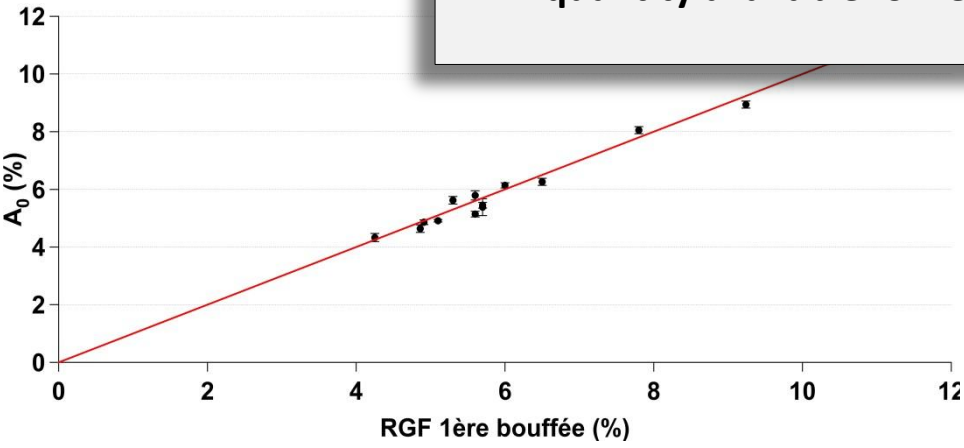


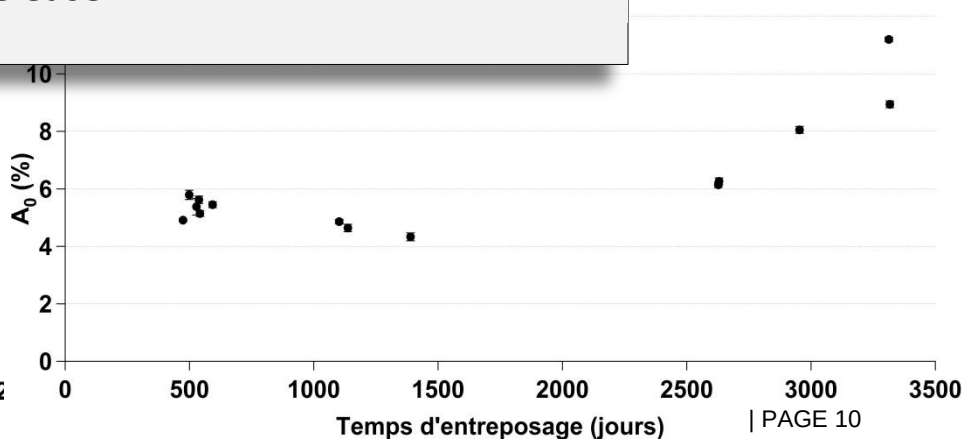
k_{2moy}= 2,97 ± 0,07 min^{-1}



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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