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Modélisation de la dissolution de l'UO₂ en milieu nitrique, vers un modèle macroscopique des réacteurs de dissolution

F. Charlier, D. Canon, A. Gravinese, G. Borda, S. Lalleman, A. Magnaldo, E. Schaer

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Florence CHARLIER^{1*}, Delphine CANION¹, Anthony GRAVINESE¹, Gilles BORDA², Sophie LALLEMAN¹, Alastair MAGNALDO¹,
Éric SCHAER³

¹ CEA, Nuclear Energy Division, RadioChemistry & Processes Department, SERA, Laboratory of dissolution study, F-30207 Bagnols-sur-Cèze, France.

² CEA, Nuclear Energy Division, Technology of Fuel Cycle Department, SGCS, Laboratory of civil engineering and instrumentation, F-30207 Bagnols-sur-Cèze, France.

³ Laboratory of Reactions and Process Engineering, UMR 7274, CNRS, Université de Lorraine, 54001 Nancy, France

*Florence.Charlier@cea.fr

Introduction

Dissolution is a key step for the **recycling of uranium dioxide**, mainly present in spent nuclear fuel. **Reactions** are particularly **complex** in those cases as they are **triphasic** and **catalyzed by one of their products**.

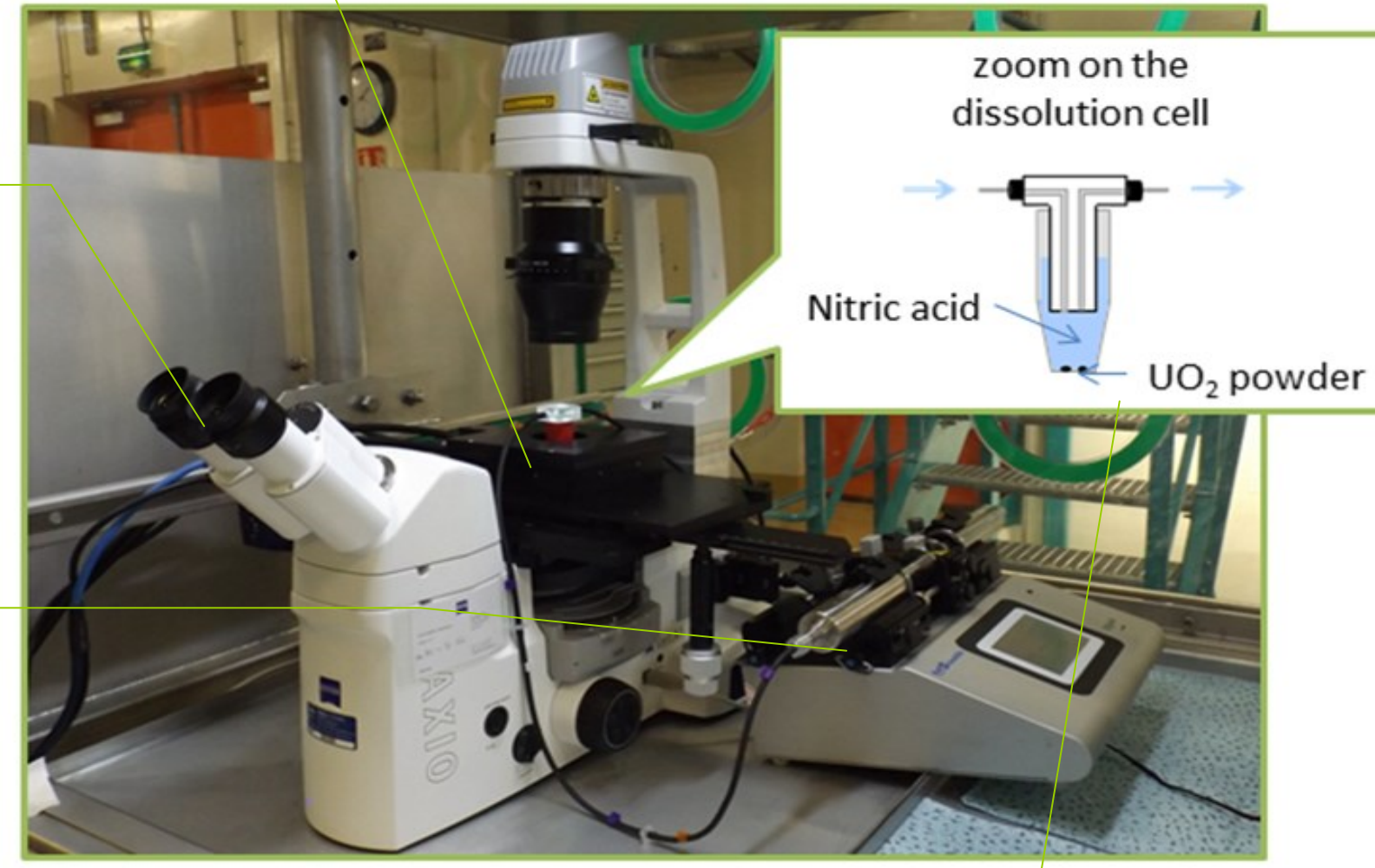
This work focuses on developing a **model** including all the characteristics of the dissolution and their effects on the kinetics.

Kinetic study without influence of heat transfer and mass transport

Peltier
→ no temperature Gradient.

Microscope
to follow the projected surface of the particles.

Syringue pump
→ renewal of nitric acid.
= No concentration gradient in the boundary layer.

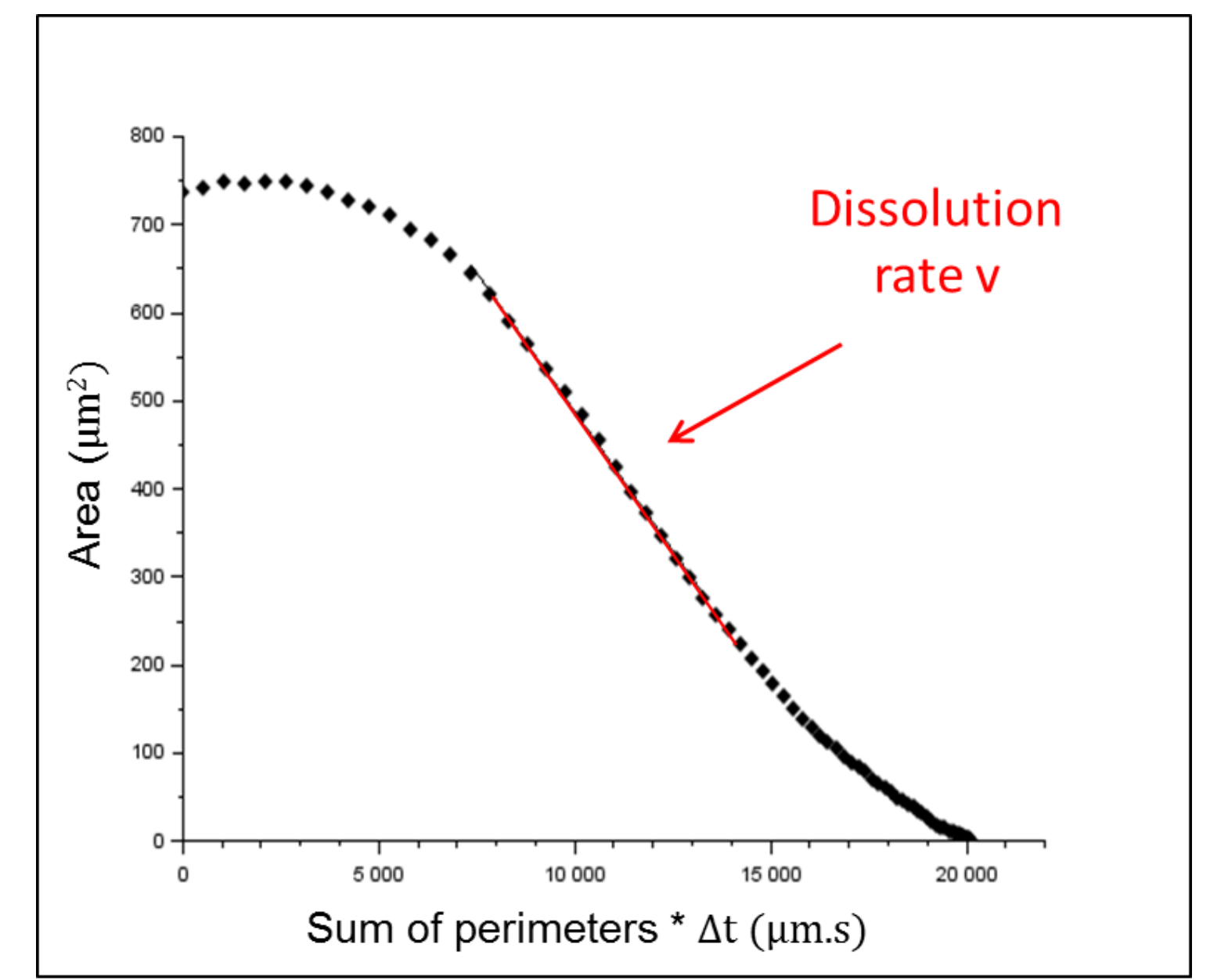
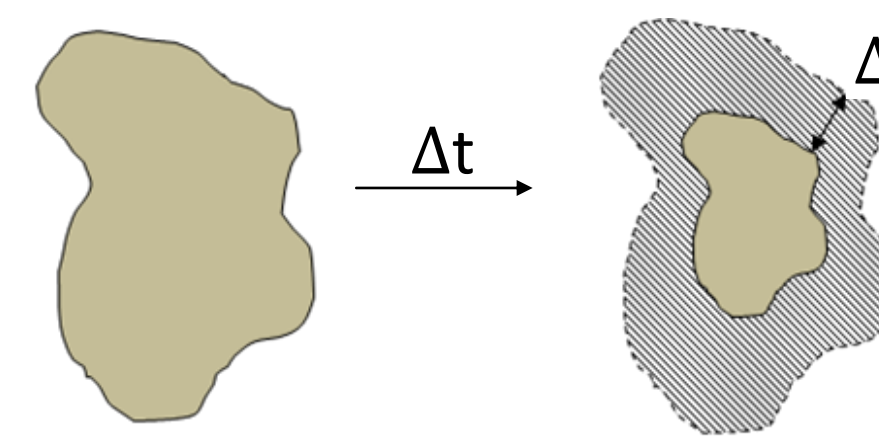


High liquid/solid ratio
No bulk accumulation

Kinetic study

$$\text{Area} = \text{perimeter} \times \Delta l$$

$$\Delta l = \text{dissolution rate} \times \Delta t$$

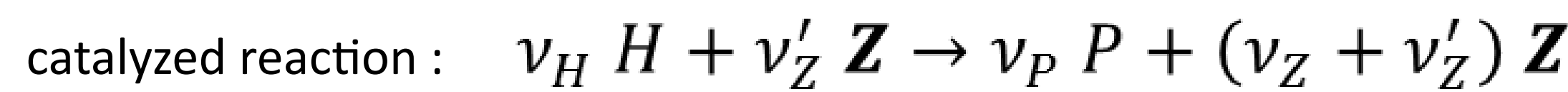


Dissolution of an UO₂ particle
T = 60°C, [HNO₃] = 5 mol.l⁻¹ and [HNO₂]/[HNO₃]₀ = 0.98.

Quantification of the influence of other phenomenas on the dissolution rate

Other phenomenas that need to be included in the model

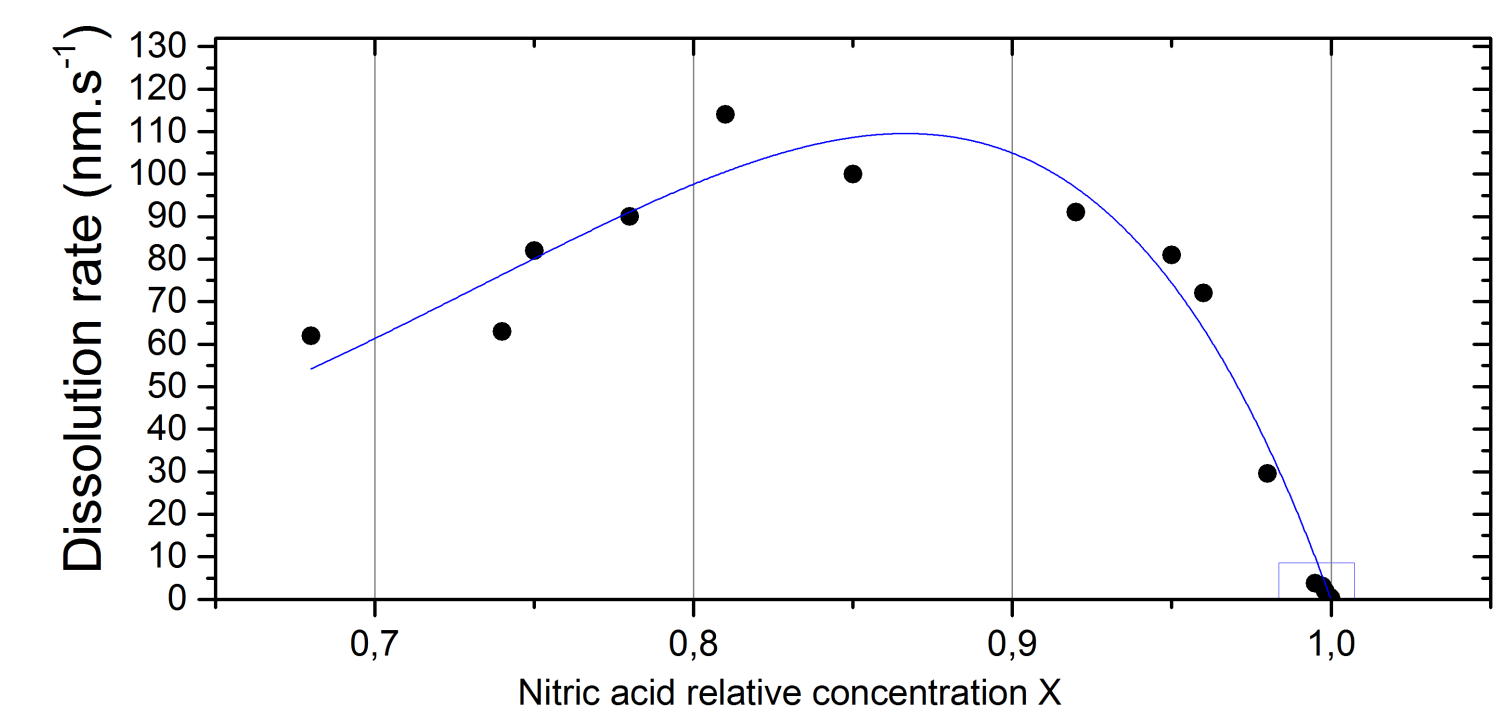
1. Mecanism and reaction rate for an autocatalytic reaction



$$v = k_{nc} C_H^{n1} + k_c C_H^{n2} C_Z^p$$

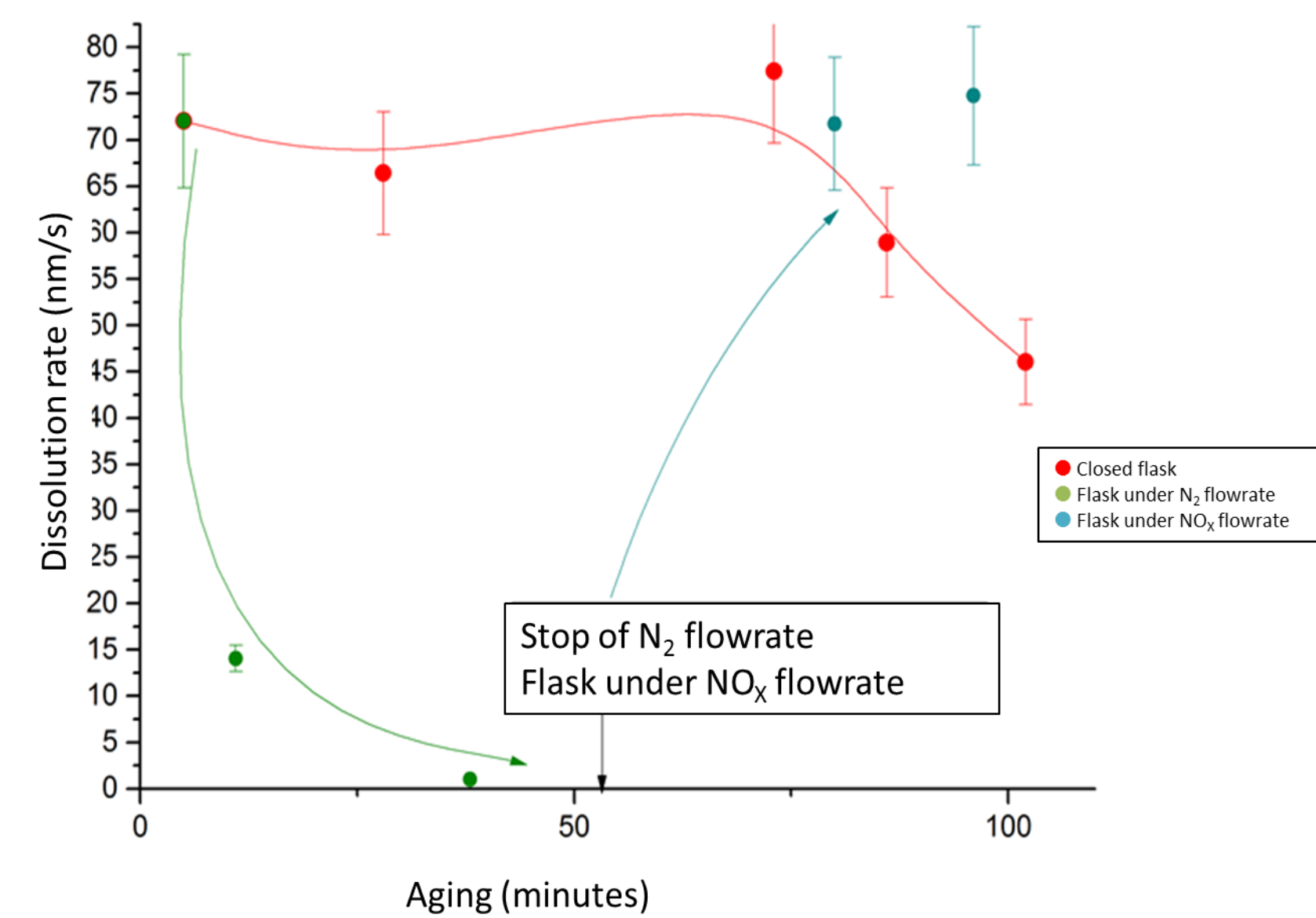
[PHI - 14]

2. influence of the catalyst on the dissolution rate

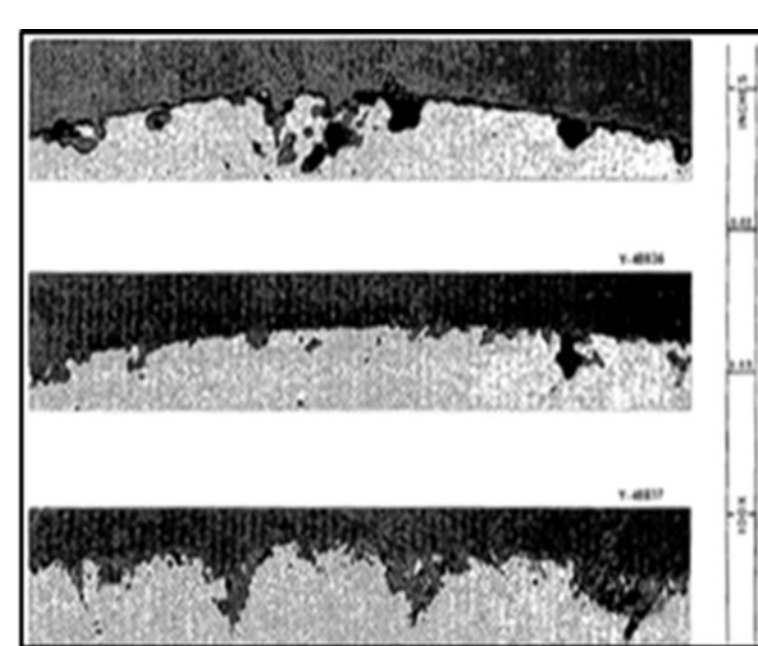
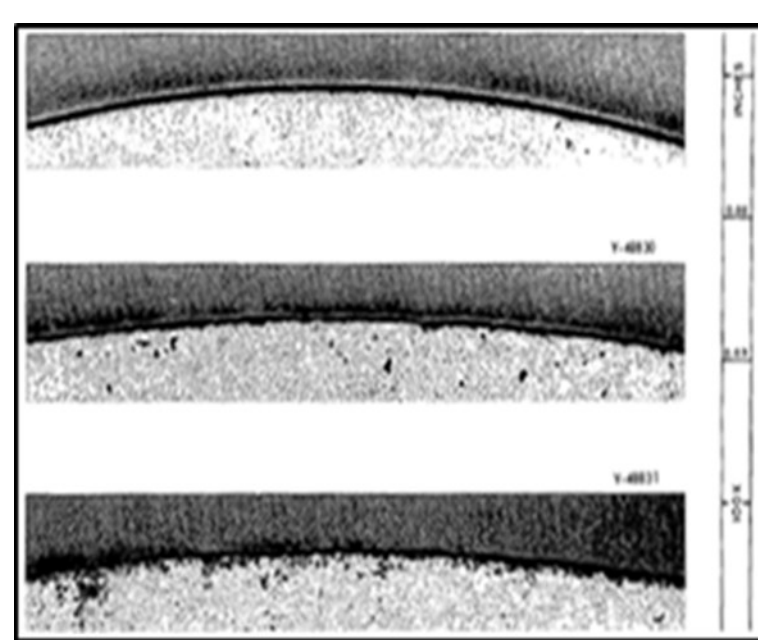


Model	catalyseur (User)	
Equation	$w^0 X^{n1} + w^1 X^{n2} (1-X)^p$	
Adj. R-Square	0.96165	
	Value	Standard Error
w	7317	3094
n1	3	-
n2	6.54	0.92
p	1.01	0.13

3. Gaz-liquid exchanges influence on dissolution rate



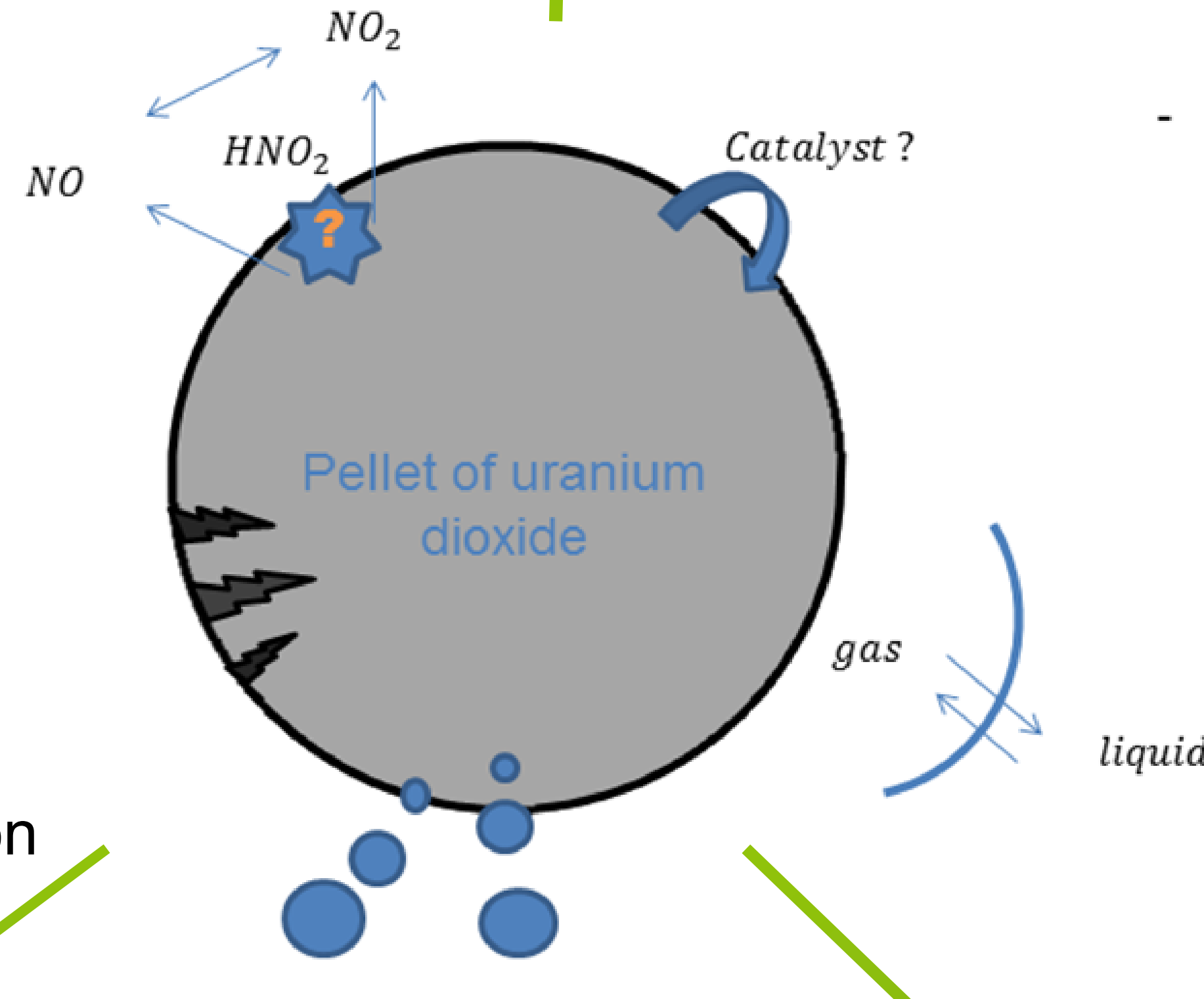
5. Preferential attack of the solid : cracks



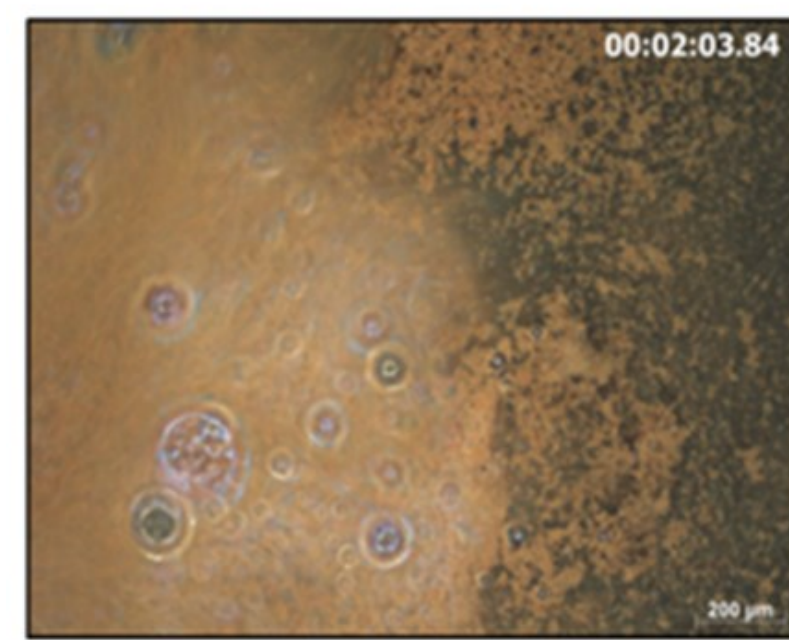
[URI - 65]

Pellet surface before dissolution

Pellet surface after dissolution



4. Bubbles influence on mass transfer



[PHI - 14]

Model for the local kinetic rate

Application of the kinetic model to ideal reactors

Verification of development ideas thanks to the model

Caption:

Well advanced

Ongoing

Upcoming

Conclusions and perspectives

Such a **multiscale** model for the dissolution kinetic will enable to :

- **optimize** actual dissolvers,
- **develop innovative reactors** for recycling of metals or spent nuclear fuels.

[PHI - 14] P. MARC, *Etude de réactions hétérogènes autocatalytiques. Application à la dissolution du dioxyde d'uranium*. Thèse de l'université de Lorraine, 17 décembre 2014.

[URI - 65] A. L. URIARTE & R. H. RAINEY, « Dissolution of high-density UO₂, PuO₂, and UO₂-PuO₂ pellets in inorganic acids », *Document technique ORNL*, Réf. ORNL-3695, avril 1965.