

Modélisation de la dissolution de l'UO_2 en milieu nitrique, vers un modèle macroscopique des réacteurs de dissolution

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Dissolution of uranium dioxide in nitric medium, towards a macroscopic model of reactors.

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Introduction

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

This work focuses on developping 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

↓

Quantification of the influence of other phenomenas on the dissolution rate

↓

Model for the local kinetic rate

↓

Application of the kinetic model to ideal reactors

↓

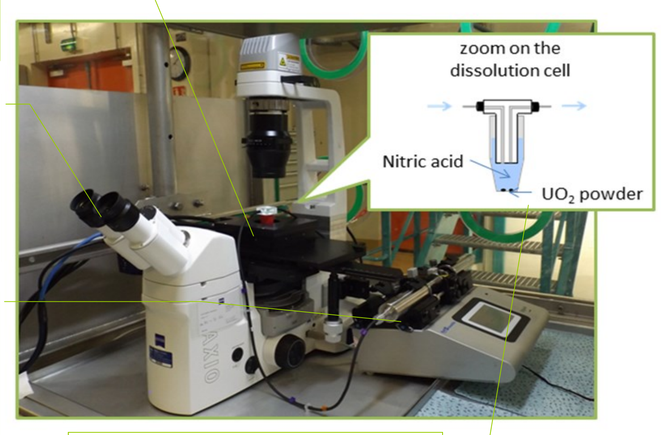
Verification of development ideas thanks to the model

Kinetic study

Peltier
→ no temperature Gradient.

Microscope
to follow the projected surface of the particles.

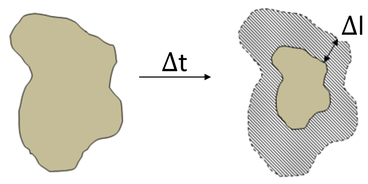
Syringue pump
→ renewal of nitric acid.
= No concentration gradient in the boundary layer.

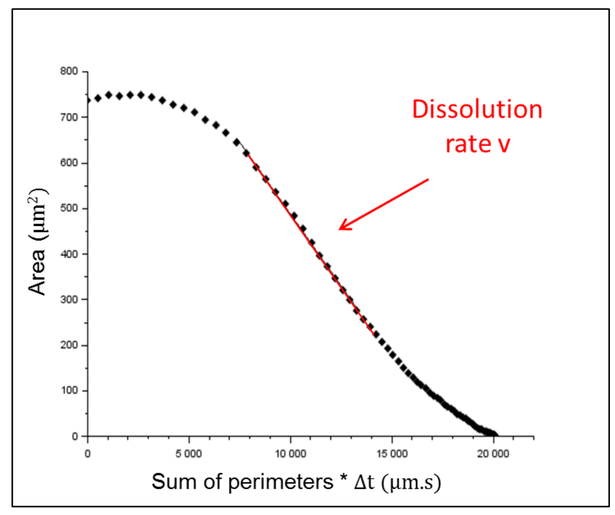


High liquid/solid ratio
No bulk accumulation

Area = perimeter x Δl

Δl = dissolution rate x Δt





Dissolution of an UO_2 particle
T = 60°C, $[HNO_3]$ = 5 mol.l^{-1} and $[HNO_3]/[HNO_3]_0$ = 0,98.

Other phenomenas that need to be included in the model

1. Mecanism and reaction rate for an autocatalytic reaction

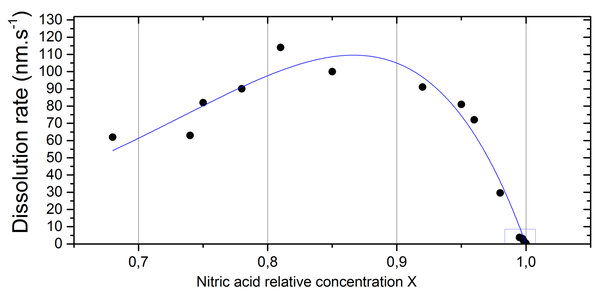
Non catalyzed reaction : $\nu_H H \rightarrow \nu_P P + \nu_Z Z$

catalyzed reaction : $\nu_H H + \nu'_Z Z \rightarrow \nu_P P + (\nu_Z + \nu'_Z) Z$

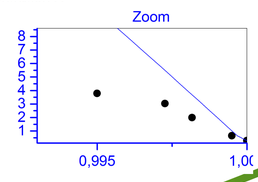
$$v = k_{nc} C_H^{n1} + k_c C_H^{n2} C_Z^{p}$$

[PHI - 14]

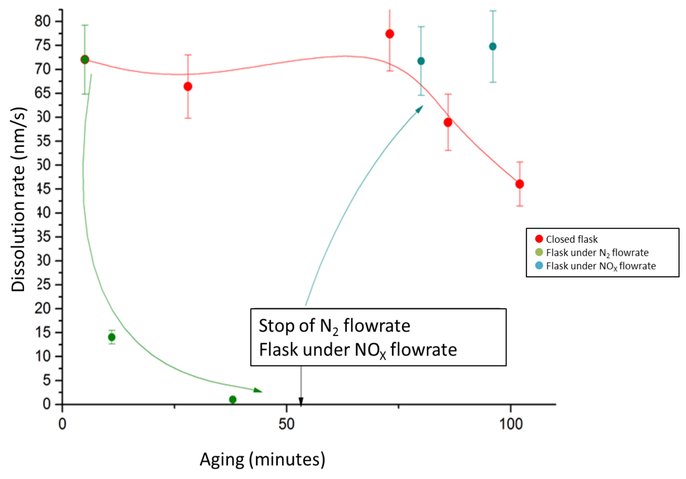
2. influence of the catalyst on the dissolution rate



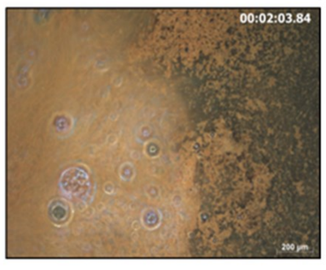
Model	catalyseur (User)		
Equation	v0*(X^n1+w*X^n2*(1-X)^p)		
Adj. R-Square	0,96165		
		Value	Standard Erro
Dissolution rate	w	7317	3094
	n1	3	--
	n2	6,54	0,92
	p	1,01	0,13



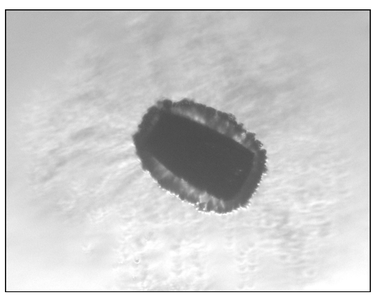
3. Gaz-liquid exchanges influence on dissolution rate



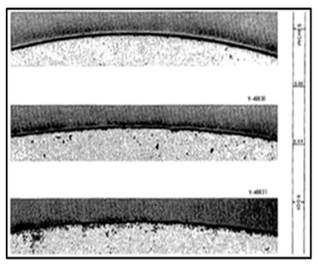
4. Bubbles influence on mass transfer



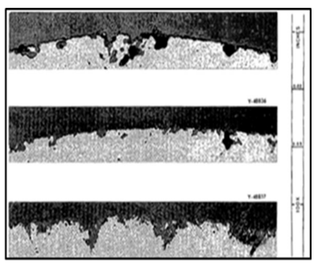
[PHI - 14]



5. Preferential attack of the solid : cracks

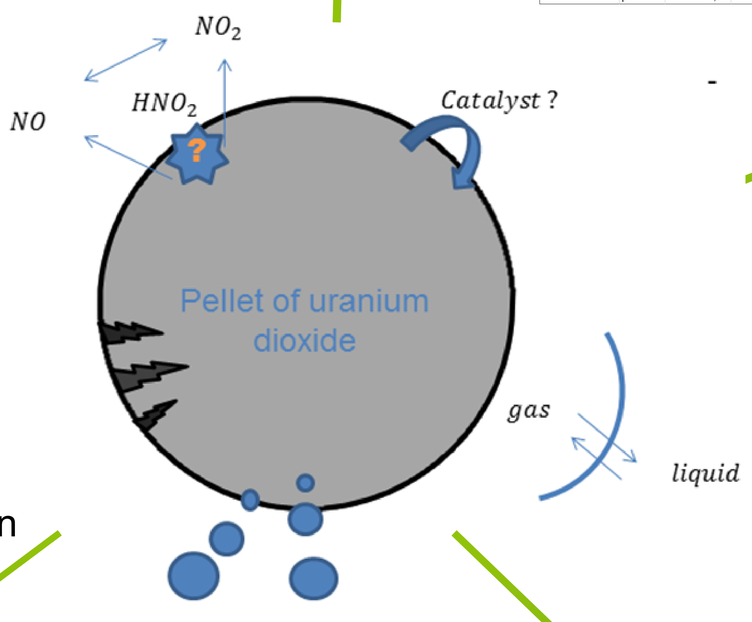


Pellet surface before dissolution



Pellet surface after dissolution

[URI - 65]



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 UO_2, PuO_2, and UO_2-PuO_2 pellets in inorganic acids », *Document technique ORNL*, Réf. ORNL-3695, avril 1965.