



# Understanding the evolution of an interface during the dissolution of Nd-doped UO<sub>2</sub> by macro-/microscopic dual approach

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# Understanding the evolution of an interface during the dissolution of Nd-doped $\text{UO}_2$ by macro-/microscopic dual approach

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## Main objectives

Recycling of actinides is a key issue to consider in order to preserve natural fissile resources and to reduce the long-term radiotoxicity of high-level waste in deep geological disposal. Due to the high radioactivity of the spent nuclear fuel, the use of model compounds is often required to understand their behavior during reprocessing. Thus, this work was focused on the elaboration of a panel of Nd-doped  $\text{UO}_2$  samples and their sintering under reducing or inert atmosphere. Then, the dissolution kinetics and mechanisms associated were studied during dissolution experiments using a macro-/microscopic dual approach.

## Synthesis

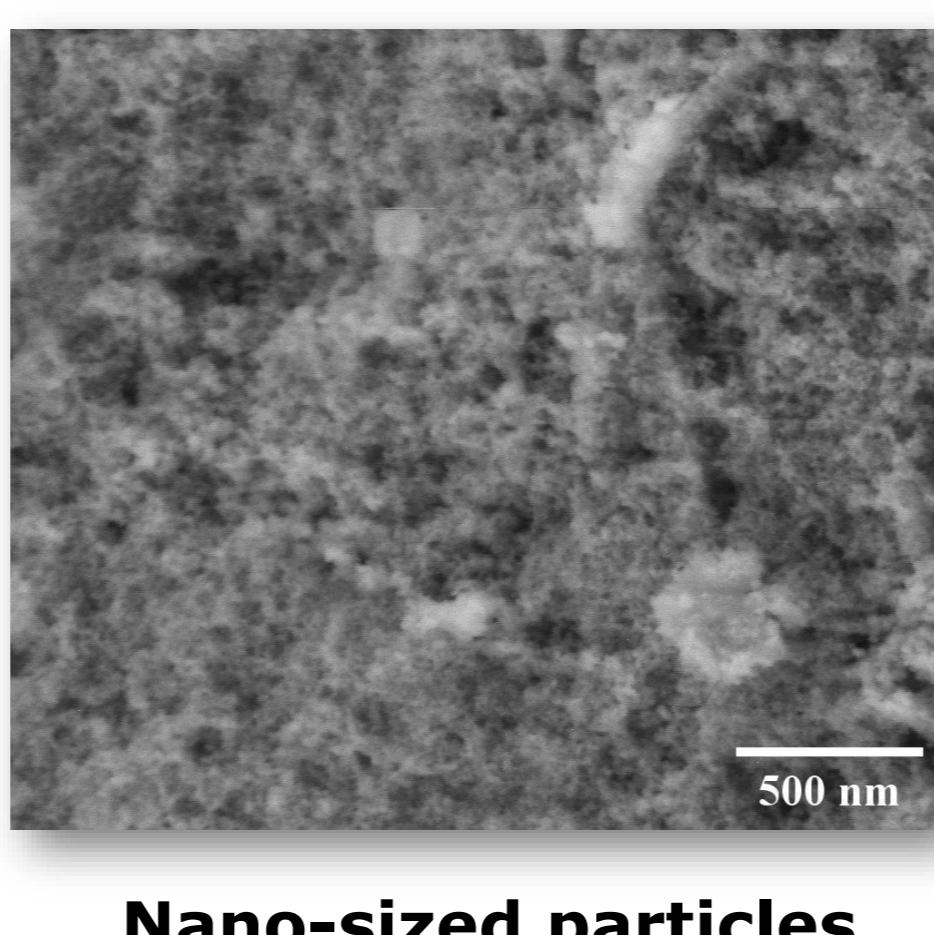
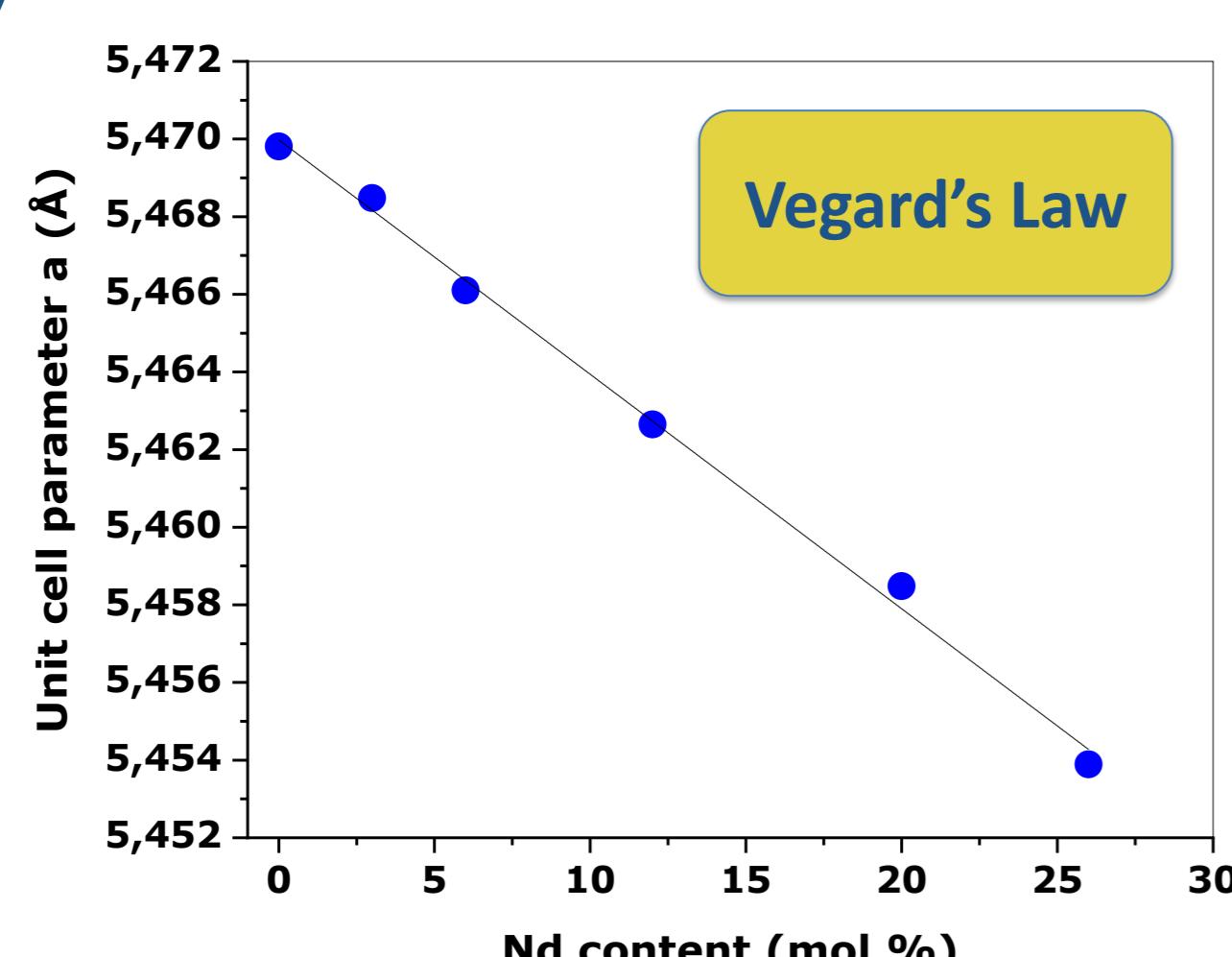
From the precursor to the targeted material

- Hydroxide route :
  - Quantitative coprecipitation of U(IV) and Nd(III)
  - Formation of homogenous  $\text{U}_{1-x}\text{Nd}_x\text{O}_2$  solid solutions

Room Temperature  
 $m_{\text{prepared}} \leq 1 \text{ g}$

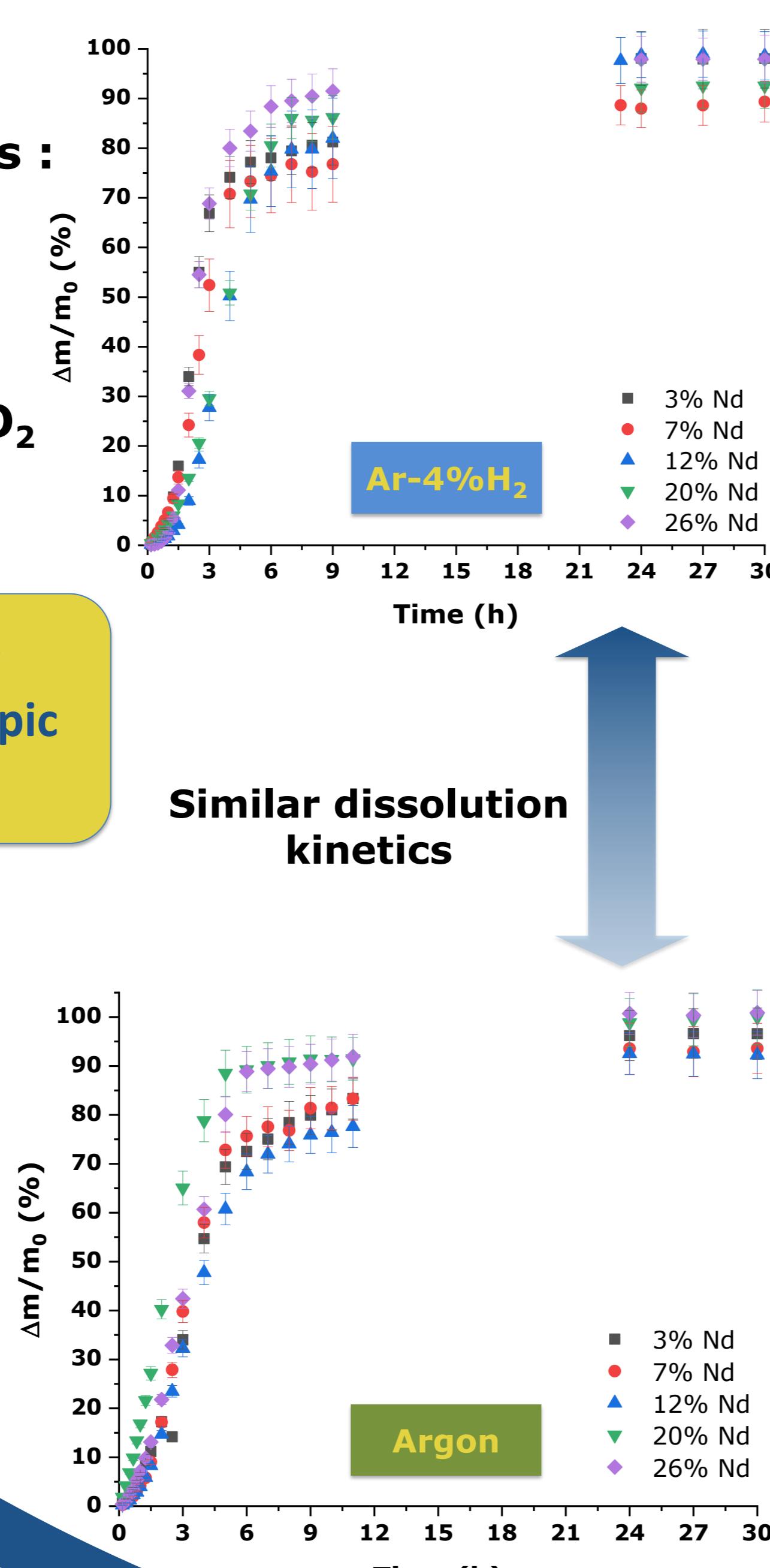
Recovery yield  
 $> 99.9 \%$

Stirring  
400% excess ammonia  
700°C  
Ar-4%H<sub>2</sub>



- Dissolution of  $\text{U}_{1-x}\text{Nd}_x\text{O}_{2-y}$  samples :
  - Congruent nature :  $R_L(\text{U}) \approx R_L(\text{Nd})$
  - Presence of Ln lowers the chemical durability : factor  $\approx 20$  for  $\text{U}_{0.8}\text{Nd}_{0.2}\text{O}_2$  vs.  $\text{UO}_2$

No impact of the sintering atmosphere at the macroscopic scale  
Experimental parameters :  
2M HNO<sub>3</sub>  
T = 60°C  
Static conditions



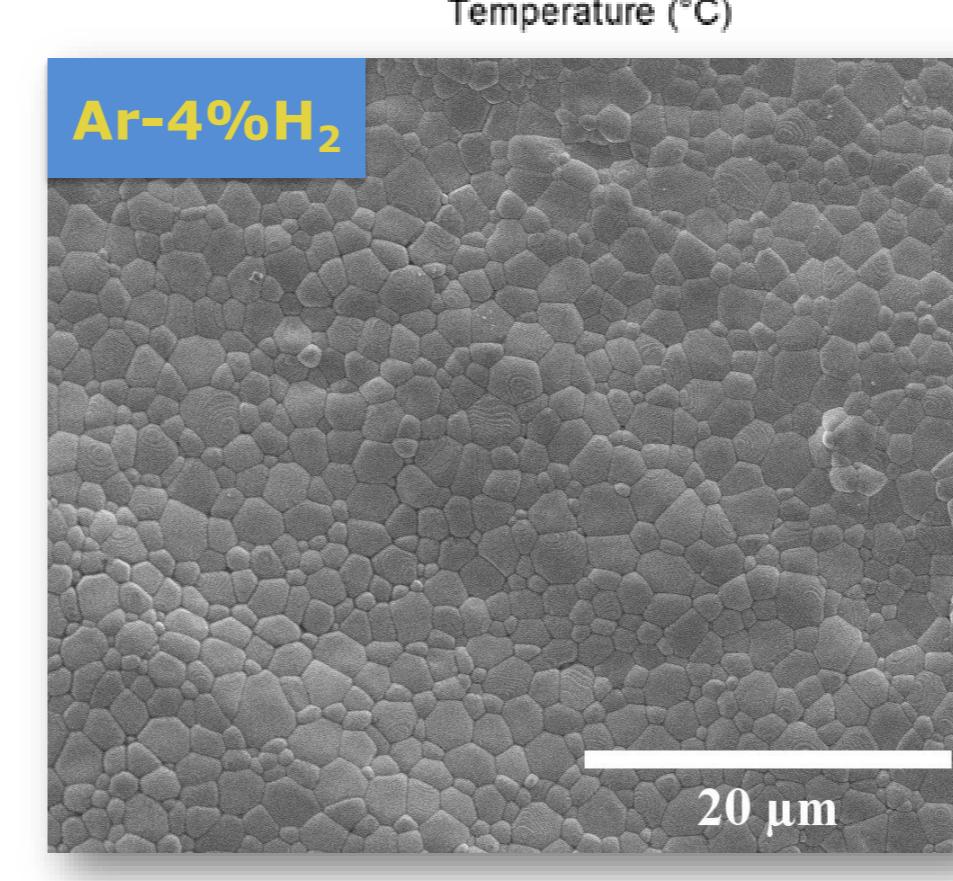
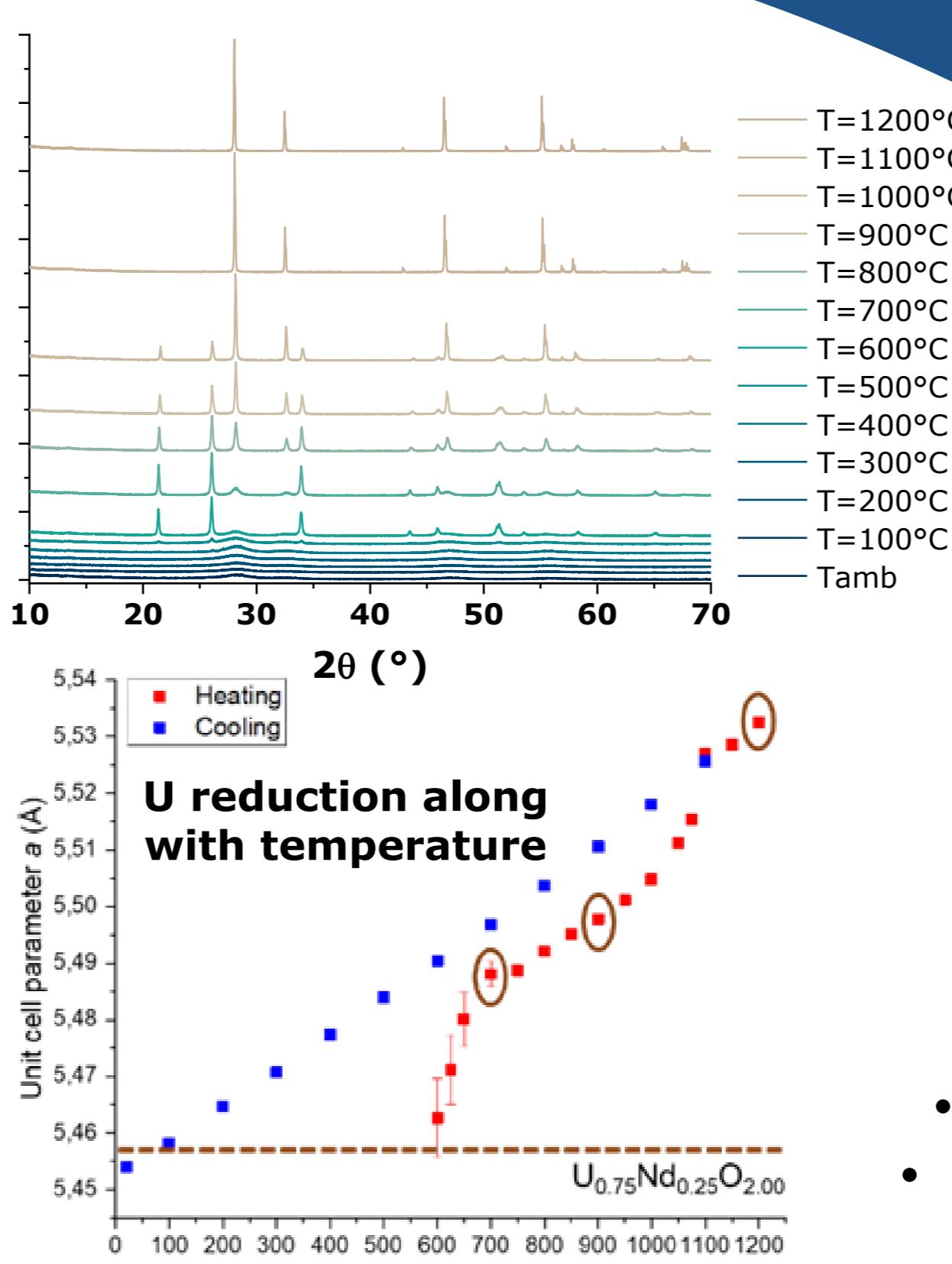
## Dissolution experiments

Macroscopic description of the chemical durability

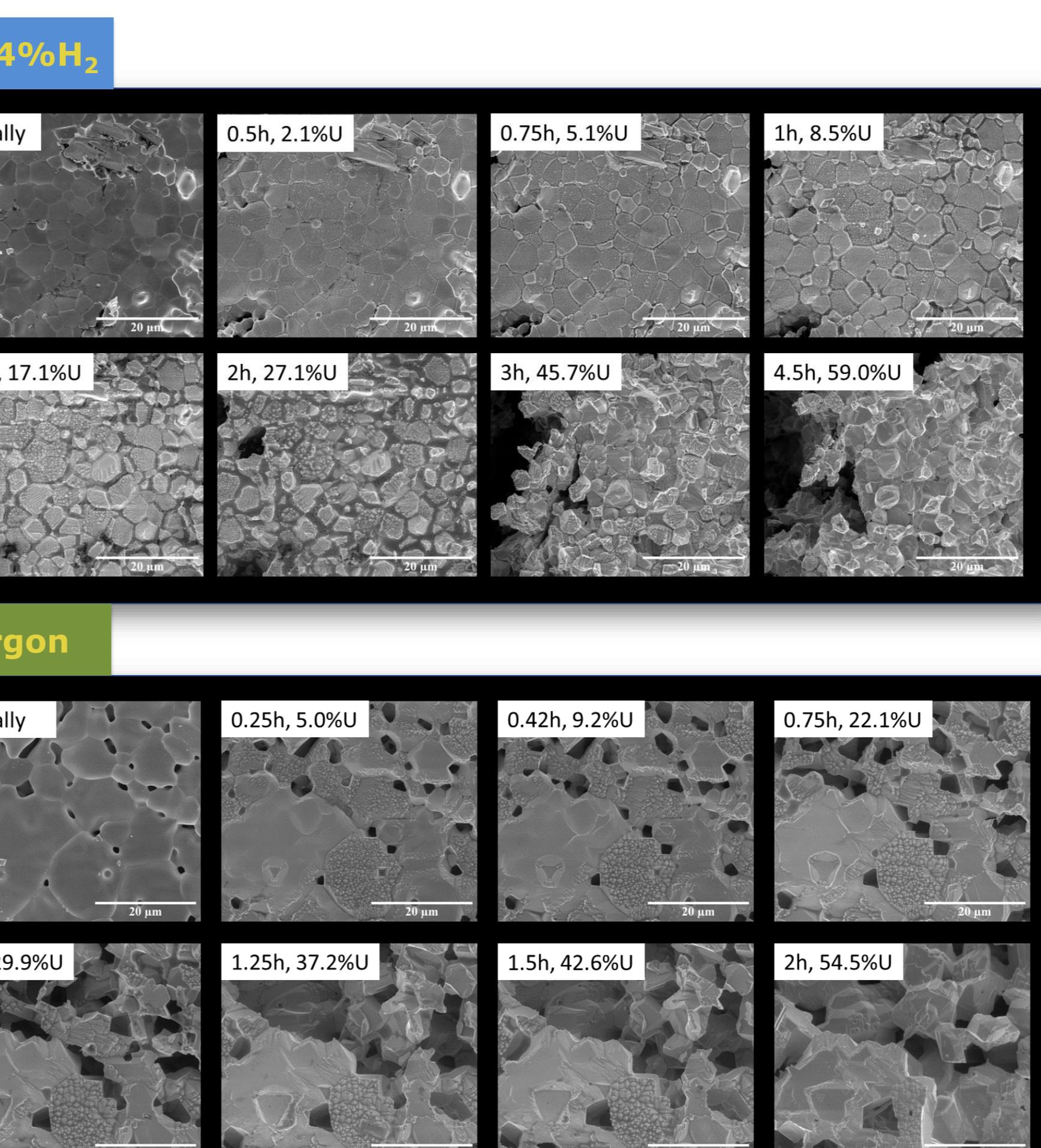
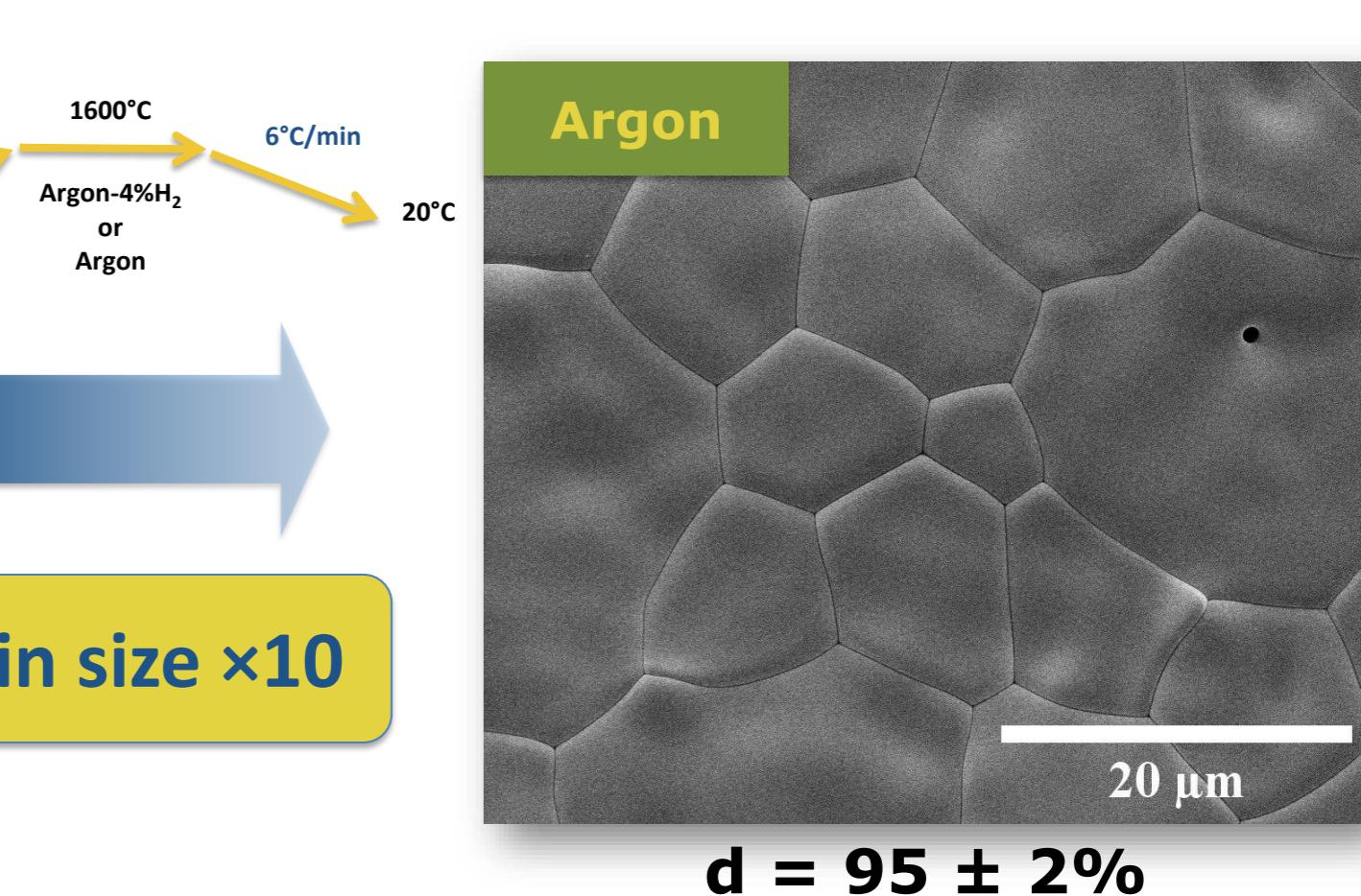
## Sintering

From the powder to the dense pellet

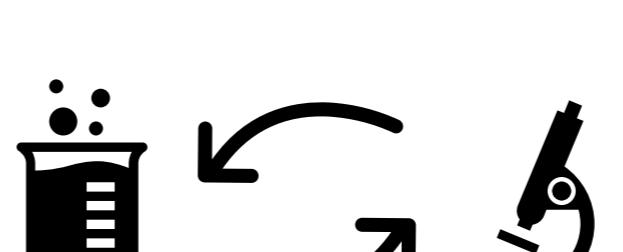
HT-XRD of  $\text{U}_{0.8}\text{Nd}_{0.2}\text{O}_{2-y}$  sample under argon atmosphere



- Impact of the sintering atmosphere :
  - Various microstructures
  - U speciation : U(IV)/U(V)
  - O/M stoichiometry : oxygen vacancies



Operando monitoring of the Solid/Liquid interface evolution during dissolution by ESEM



## Interface monitoring

Microscopic description of the evolving interface

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