EUROPEAN SYNCHROTRON RADIATION FACILITY
INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

	Experiment title: Investigation of grain size evolution and solid solution formation of a mixture of UO2 and PuO2 oxides during sintering by high resolution micro-tomography and micro-xanes	**Experiment number**: ME488
Beamline: ID22	**Date of experiment**: from: 07 september 02 to: 13 september 02	**Date of report**:
Shifts: Total: 15	**Local contact(s)**: A. Somogyi, A. Simionovici, P. Bleuet	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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Report:

The report gives the slides which will be presented in the 17th ICXOM Conference

μ-XANES and High resolution X-ray μ-tomography as tools to study nuclear fuel ceramic sintering process

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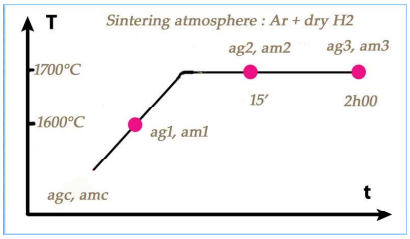
INPG : J.M. Chaix, C.P. Carry

Nuclear Fuel Sintering study

- sintering stage of Mixed Oxide Fuel (UO_2-PuO_2) manufacturing process :
  - Influence on grain size (Fission Gas release)
  - Homogeneity of Pu localisation (Hot Spots)
- A two-scale model has been developped (first scale : fuel pellet, second scale : smaller than grain size), here use of **μ-XANES and High resolution X-ray μ-tomography as tools for validation**

Sample preparation

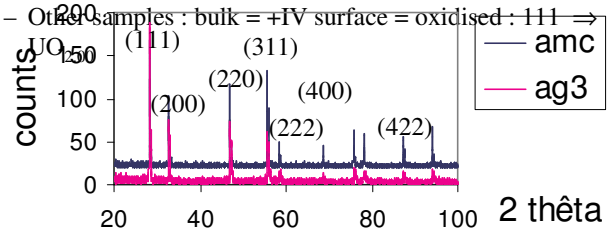
- UO_2 samples (size ≈ 40μm)



- green mixing of UO_2(95%) and PuO_2(5%): « ESRF2 »

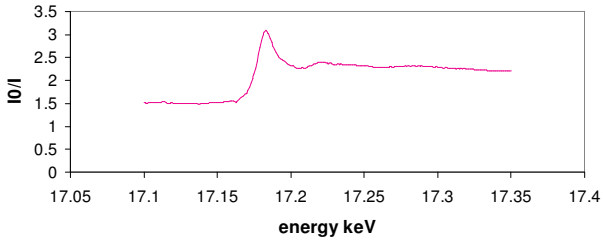
Oxidation states of U (1/3)

- XRD on non-oriented powders Cu $K_{\alpha1}$=1.540598, $K_{\alpha2}$=1.544426:
  - amc and agc : UO_2+U_4O_9 +U_3O_7: 111 location gives $UO_{2.13}$
  - Other samples : bulk = +IV surface = oxidised : 111 ⇒ $UO_{2.33}$



Oxidation states of U (2/3)

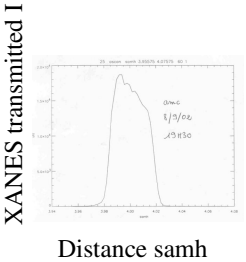
- Xanes spectra :



Oxidation states of U (3/3)

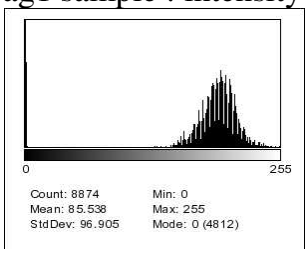
- XANES absorption edge :

sample	mode	Energy (keV)	Shift(eV)	Expected oxidation state
ag2	T	17.175	0	+IV
	F	17.169	-6	+4 ≤ x ≤ +4.26
amc	T	17.177	+2	+4.26
	F	17.173	-2	+4.26
ESRF2	T	17.174	-1	+IV
	F	17.172	-3	+4.26
agc	T	17.176	+1	+4.26
	F	17.172	-3	+4.26
U_3O_7	T	17.178	+3	+4.66
	F	17.176	+1	+4.66
U_3O_8	T	17.179	+4	+5.33
	F	17.175	0	+5.33



µ-tomography

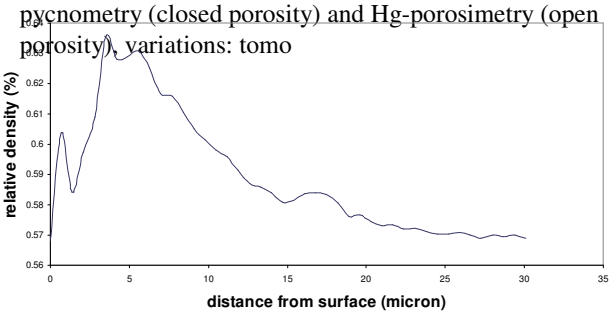
- Exemple of ag1 sample : intensity analysis



$$\rho_{slice} = \frac{\sum_{n=1}^{255} n \,.\, nb\_pixel(n)}{sample\_pixels\_in\_slice}$$

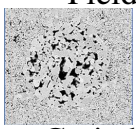
3D – half grain density curves

- Absolute mean value of relative density obtained from He-pycnometry (closed porosity) and Hg-porosimetry (open porosity), variations: tomo

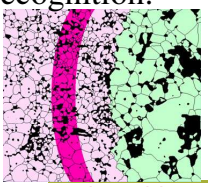


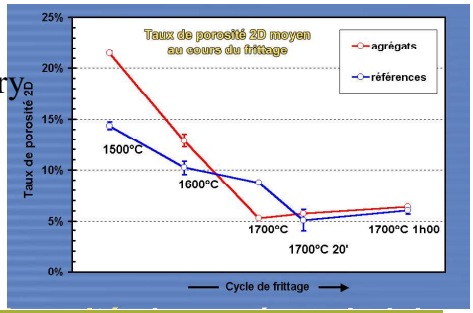
Comparison with 2D measurements : FEG SEM

- Field Electron Gun - SEM and image analysis:



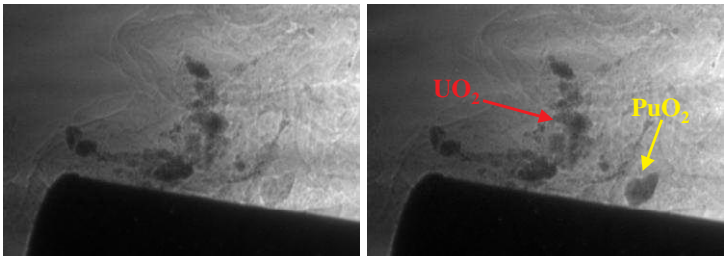
After Grain boundary recognition:





U and Pu tomographies

- ESRF2 sample (2D slices without reconstruction)



Between U and Pu thresholds

Above Pu threshold

Conclusion

- Next step: comparison between models (software results) and 3D-high resolution tomography density
- Limitations: due to resolution
- perspectives: tomographies below and above Pu edge to study U and Pu repartition within single grains , and around U-edge to study U repartition according to its oxidation states