



**HAL**  
open science

## Helium precipitation study in UO<sub>2</sub> by Transmission Electron Microscopy

A. Michel, G. Carlot, C. Sabathier-Devals, C. Onofri-Marroncle, M. Dumont,  
M. Cabie

► **To cite this version:**

A. Michel, G. Carlot, C. Sabathier-Devals, C. Onofri-Marroncle, M. Dumont, et al.. Helium precipitation study in UO<sub>2</sub> by Transmission Electron Microscopy. CIMTEC 2018 - 8th Forum on New Materials, Jun 2018, Perugia, Italy. cea-02338635

**HAL Id: cea-02338635**

**<https://cea.hal.science/cea-02338635v1>**

Submitted on 21 Feb 2020

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

DE LA RECHERCHE À L'INDUSTRIE



[www.cea.fr](http://www.cea.fr)

# CIMTEC 2018

## 8<sup>th</sup> Forum on New Materials

# Helium precipitation study in UO<sub>2</sub> by Transmission Electron Microscopy

A. Michel<sup>1,\*</sup>, G. Carlot<sup>1</sup>, C. Sabathier<sup>1</sup>, C. Onofri<sup>1</sup>, M. Dumont<sup>2</sup> and  
M. Cabie<sup>3</sup>

*1. CEA / DEN / DEC, Saint Paul Lez Durance, France*

*2. IM2NP – UMR CNRS 7334 – Aix-Marseille Université, Marseille, France*

*3. CP2M – Aix-Marseille Université, Marseille, France*

*\* [adrien.michel2@cea.fr](mailto:adrien.michel2@cea.fr)*

PERUGIA, ITALY – 14<sup>TH</sup> JUNE 2018

# INTRODUCTION – SPENT NUCLEAR FUEL



In-pile irradiation

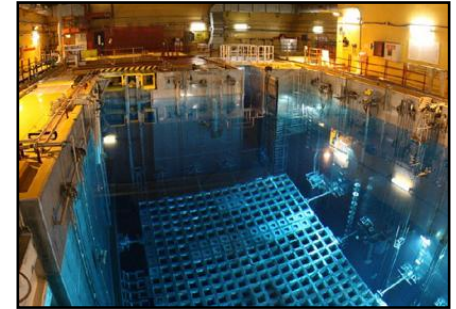


Fission products

Irradiation damages

Bubbles of fission gases

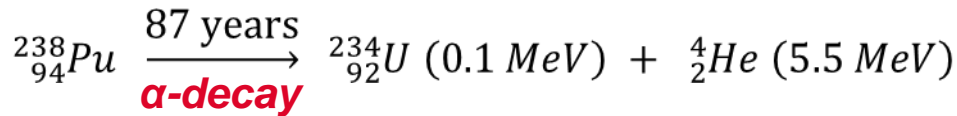
Microstructural changes



Management of spent fuel



## Direct disposal of spent nuclear fuel in deep geological layer

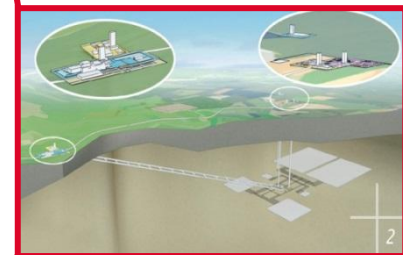


Microstructure alteration and/or pellet damaging

- Impact of helium on the microstructure evolution of the spent fuel
- To supply theoretical models of long time behaviour for spent fuel



Reference scenario :  
Reprocessing

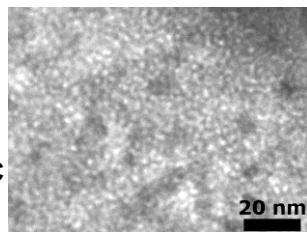


Optional scenario :  
Direct disposal

# LITERATURE ON HELIUM PRECIPITATION IN FUEL

	Authors	Fuel type	Implantation	Annealing conditions	Bubble size (nm)	Bubble density (b.m <sup>-3</sup> )
Implanted materials	Satonnay et al.	UO <sub>2</sub> single crystal	7 keV – 7.10 <sup>15</sup> at.cm <sup>-2</sup> - 1 at. %	600°C	~ 25	/
	Sabathier et al.	Polycrystalline UO <sub>2</sub>	7 keV - 7.10 <sup>15</sup> at.cm <sup>-2</sup> - 1 at. %	T° > 500°C	No bubbles	/
	Sabathier et al.	Polycrystalline UO <sub>2</sub>	500 keV – 1.10 <sup>16</sup> – 0.3 at. %	900°C 0min 1000°C 0min 1000°C 30min 1000°C 1h	~ 1 2.7 1.5 - 2 3	/ 4.10 <sup>23</sup> 1.10 <sup>24</sup> 1.10 <sup>24</sup>
Self-irradiated materials	Wiss et al.	(U <sub>0,9</sub> <sup>238</sup> Pu <sub>0,1</sub> )O <sub>2</sub>	/	> 600°C	1	/
	Talip et al.	(U <sub>0,9</sub> <sup>238</sup> Pu <sub>0,1</sub> )O <sub>2</sub>	/	1130°C 1h 1530°C 3h	1 - 2 5 - 6	/

TEM observation of helium bubbles in 500 keV He-implanted UO<sub>2</sub> (10<sup>16</sup> at.cm<sup>-2</sup>) and annealed 30 min at 1000°C



→ Only few results on helium precipitation in oxide fuels and scattered results

## Helium precipitation in UO<sub>2</sub> and (U,Pu)O<sub>2</sub> matrices

UO<sub>2</sub>

Separated effects study:

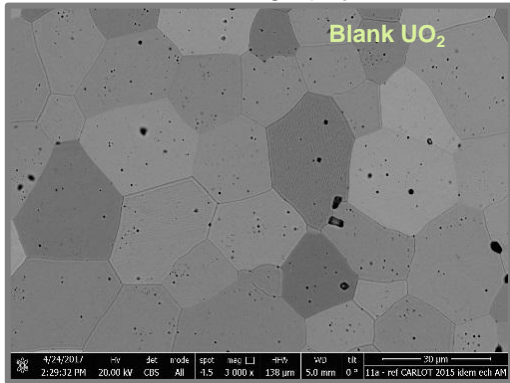
*Implantation - Irradiation – Temperature –  
Fine characterization : TEM*



Size and density characterization of helium bubbles populations in disposal conditions

# EXPERIMENTAL APPROACH

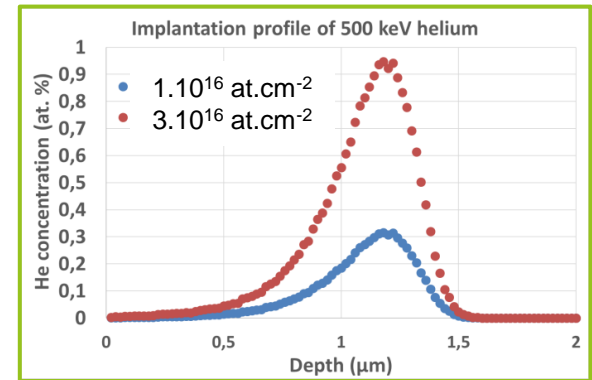
SEM micrography



Depleted UO<sub>2</sub> disks



SRIM calculations



**Helium implantation**  
**500 keV**  
 2 fluences  $1.10^{16}$  and  $3.10^{16}$  at.cm<sup>-2</sup>

In-situ annealing

Temperature (°C)	Time (h)
200	5,7
400	10,3
600	6,5

RT°  
 Ex-situ annealing

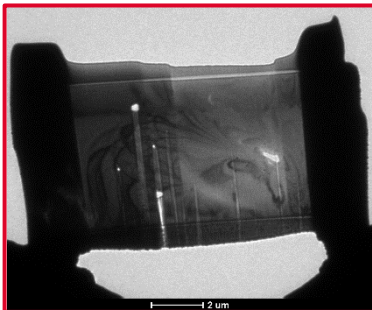
Temperature (°C)	Time (h)
600	1
600	6
800	1

FIB preparation

**Transmission Electron  
 Microscopy (TEM)**

FEI Tecnai G2  
 Spatial resolution : 0.27 nm  
  
 FEI TITAN  
 Spatial resolution : 0.1 nm

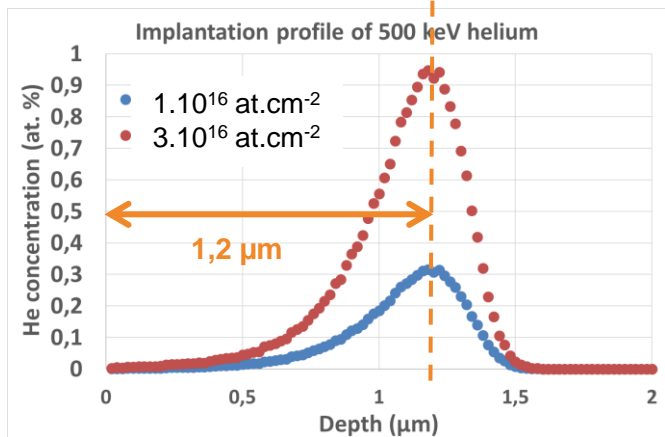
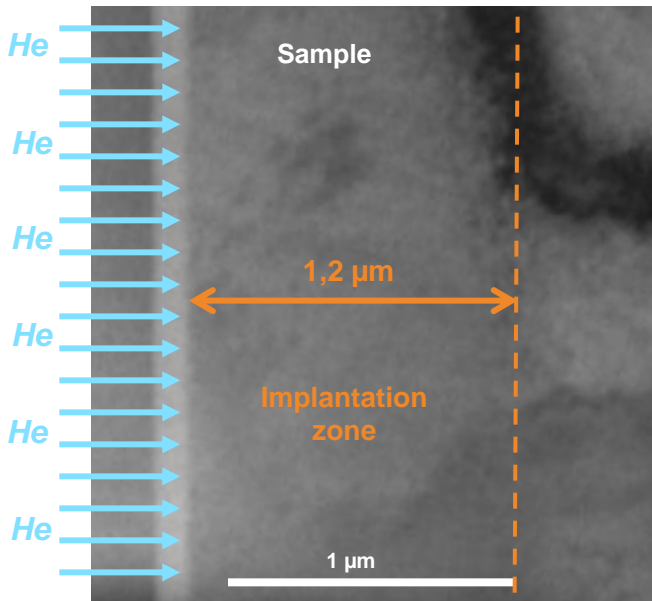
FIB cross section





# TEM OBSERVATIONS

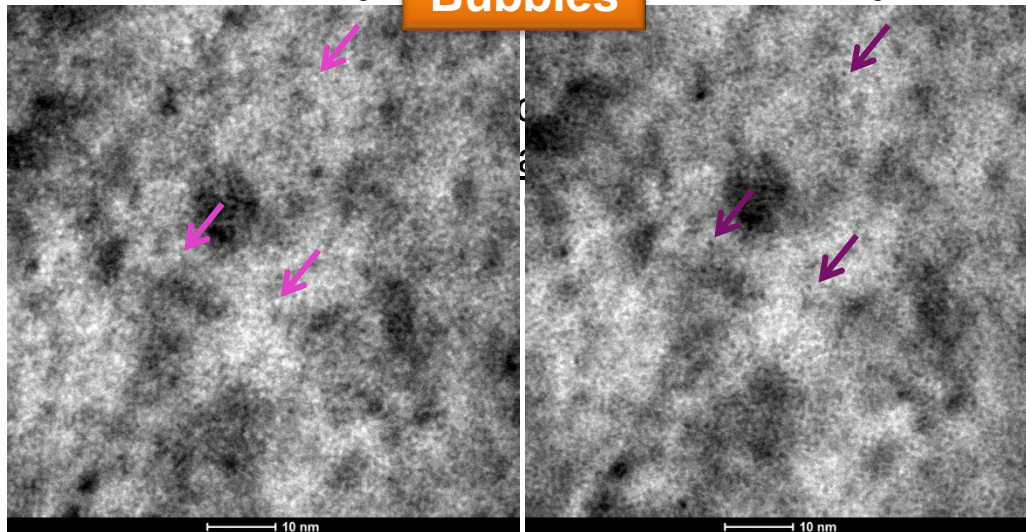
600°C and  $3.10^{16}$  at.cm<sup>-2</sup> implanted sample



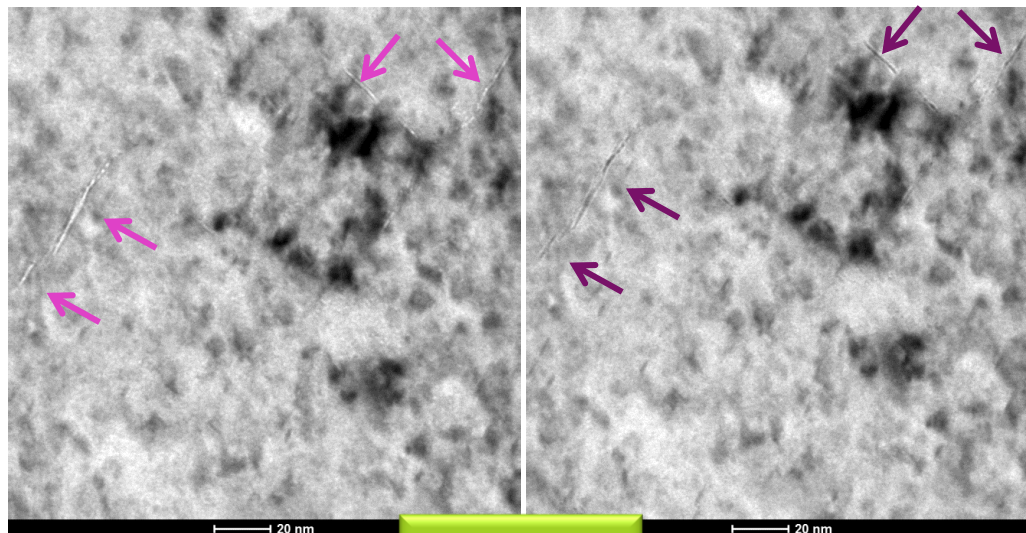
Under-focused image

## Bubbles

Over-focused image



Condition of observation : white/black contrasts for under/over focused

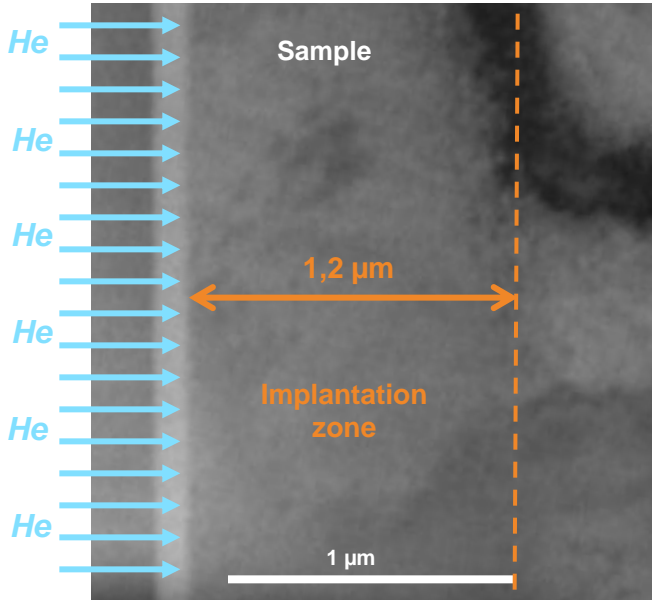


Under-focused image

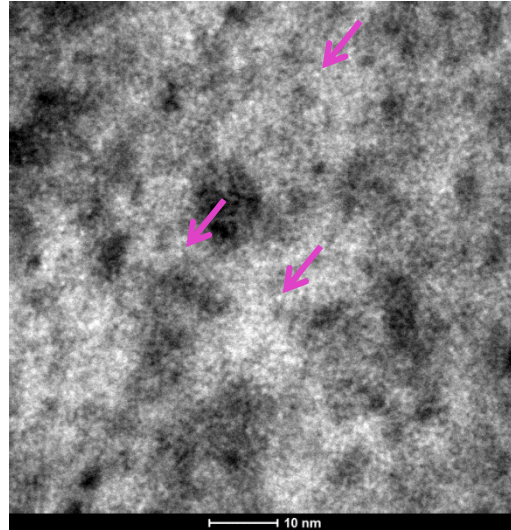
## Platelets

Over-focused image

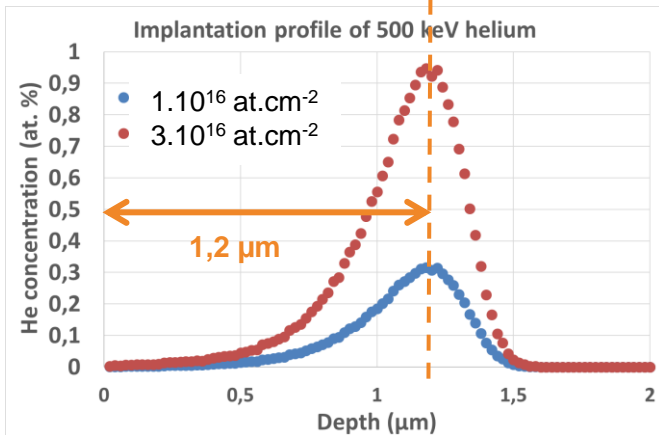
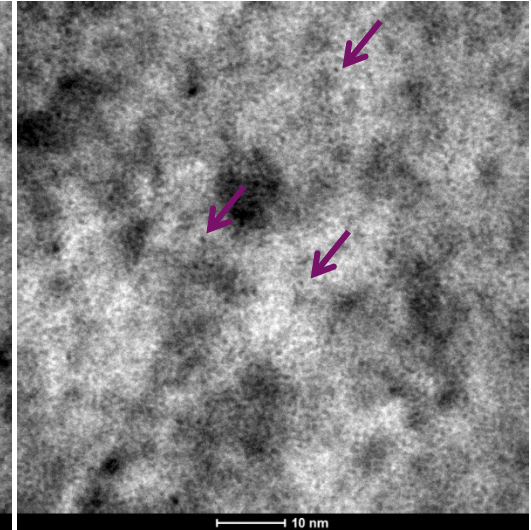
600°C and  $3 \cdot 10^{16}$  at.cm<sup>-2</sup> implanted sample



Under-focused image



Over-focused image



Annealing	Fluence (at.cm <sup>-2</sup> )	Temperature (°C)	Time (h)	Bubbles
In-situ	$1 \cdot 10^{16}$	200	4,4	No
		400	2,3	No
		<b>600</b>	1,8	<b>Yes</b>
	$3 \cdot 10^{16}$	200	5,7	No
		400	10,3	No
		<b>600</b>	6,5	<b>Yes</b>

Bubbles are observed in the implantation zone for 600°C implantations at  $1 \cdot 10^{16}$  and  $3 \cdot 10^{16}$  at.cm<sup>-2</sup>

Images do not permit to know if there are bubbles or not for the other conditions

Method : « simply sight count »



Counts of all bubbles that appear in both images (under- and –over focused)

Implantation conditions	Observation depth (μm)	He concentration (at.%)	Damages rate (dpa)	Bubble density (b.m <sup>-3</sup> ) <i>For 100 nm thick</i>	Diameter (nm)	Number of bubbles
1.10 <sup>16</sup> – 600°C	0.1	0.0023	0.02	(7.5 ± 6.0) .10 <sup>21</sup>	0.58 ± 0.58	4
1.10 <sup>16</sup> – 600°C	0.8	0.072	0.12	(2.6 ± 1.5) .10 <sup>22</sup>	0.91 ± 0.53	14
3.10 <sup>16</sup> – 600°C	0.1	0.0069	0.06	(1.5 ± 1.0) .10 <sup>22</sup>	0.79 ± 0.57	8
3.10 <sup>16</sup> – 600°C	0.1	0.0069	0.06	(1.1 ± 0.8) .10 <sup>22</sup>	0.85 ± 0.63	6
3.10 <sup>16</sup> – 600°C	1.4	0.23	0.12	(4.5 ± 2.3) .10 <sup>22</sup>	0.89 ± 0.47	24
3.10 <sup>16</sup> – 600°C	1.2	0.92	0.81	(3.0 ± 1.9) .10 <sup>22</sup>	0.86 ± 0.57	9
3.10 <sup>16</sup> – 600°C	1.2	0.92	0.81	(2.4 ± 1.4) .10 <sup>22</sup>	0.85 ± 0.52	13

Bubbles density of about  $10^{22}$  b.m<sup>-3</sup>

Maximum size of 1 nm

→ No evolution of the diameter and of the bubble density as a function of the depth or of the helium concentration

Very low statistics

Important quantity of defects → Addition of parasite contrast → Insufficient image quality

**Need to improve the observations**

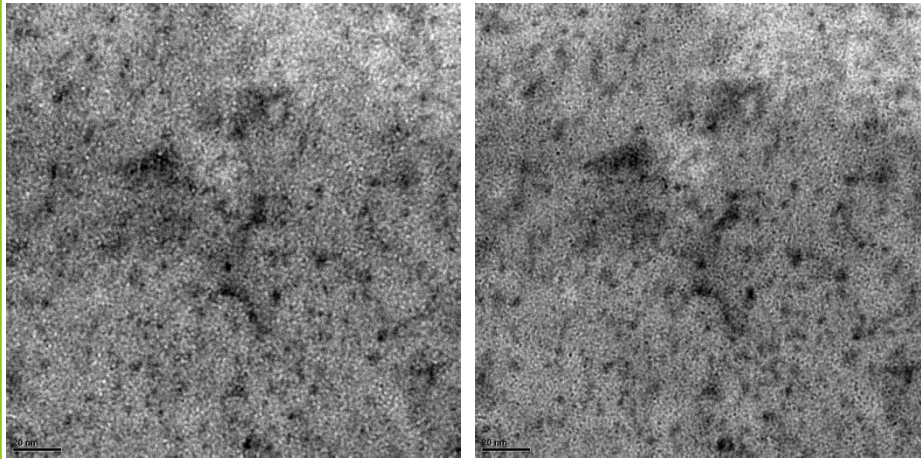
→ Complicated image analysis AND high measurement uncertainties

**Need to improve the image analysis method**



**Observations with a TEM FEI TITAN****« Simply sight count » method :**

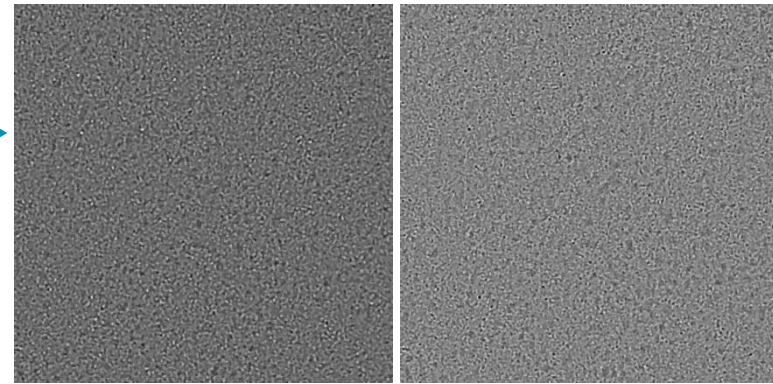
72 counted bubbles so  $(5.3 \pm 2.2) \cdot 10^{22} \text{ b.m}^{-3}$  for density and  $1.43 \pm 0.90 \text{ nm}$  for the mean diameter



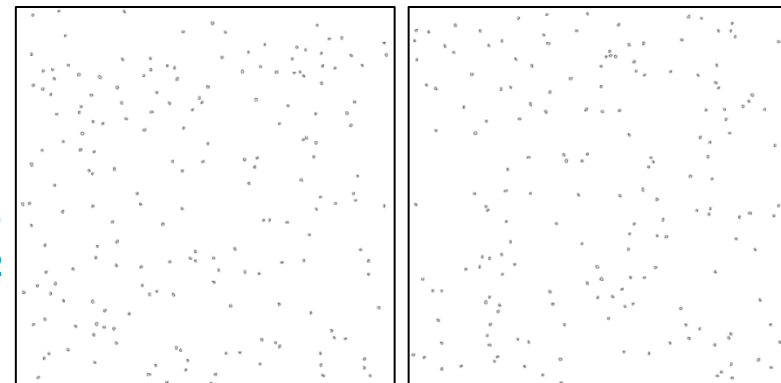
$3 \cdot 10^{16} \text{ at.cm}^{-2}$  –  $600^\circ\text{C}$  implanted sample after an  $1\text{h}/400^\circ\text{C}$  annealing on FIB cross section

**Development of a new method using ImageJ software :**

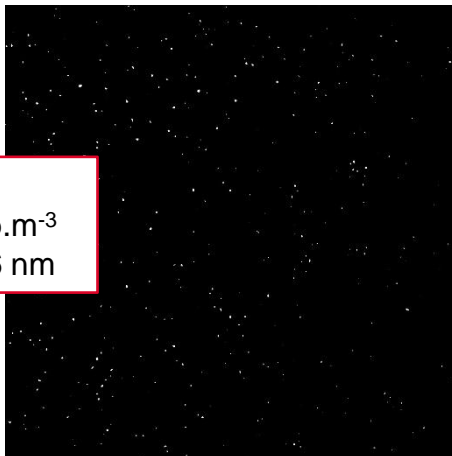
Correction of magnification as a function of focus  
Treatment and alignment of images



Thresholding of images and particle analysis



←  
Covering of 2  
images



130 counted objects  
Density :  $(9.3 \pm 3.6) \cdot 10^{22} \text{ b.m}^{-3}$   
Mean diameter :  $1.4 \pm 0.86 \text{ nm}$

- Comparison of both methods :

	Bubbles number	Density (b.m <sup>-3</sup> )	Mean diameter (nm)
Simply sight count method	72	$(5.3 \pm 2.2) \cdot 10^{22}$	$1.43 \pm 0.90$
Method using ImageJ	130	$(9.3 \pm 3.6) \cdot 10^{22}$	$1.40 \pm 0.86$

→ Difference on number of counted objects between the 2 methods but not on the mean diameter

### Advantages

- No dependence of the user on the results
- Take into account the difference of alignment of the images
- Treatment and thresholding steps enable to free itself from defects problems
- Count rate more important

### Optimization

- Implement automatic fonctions to detect the particles
- Use of macros to automate the method

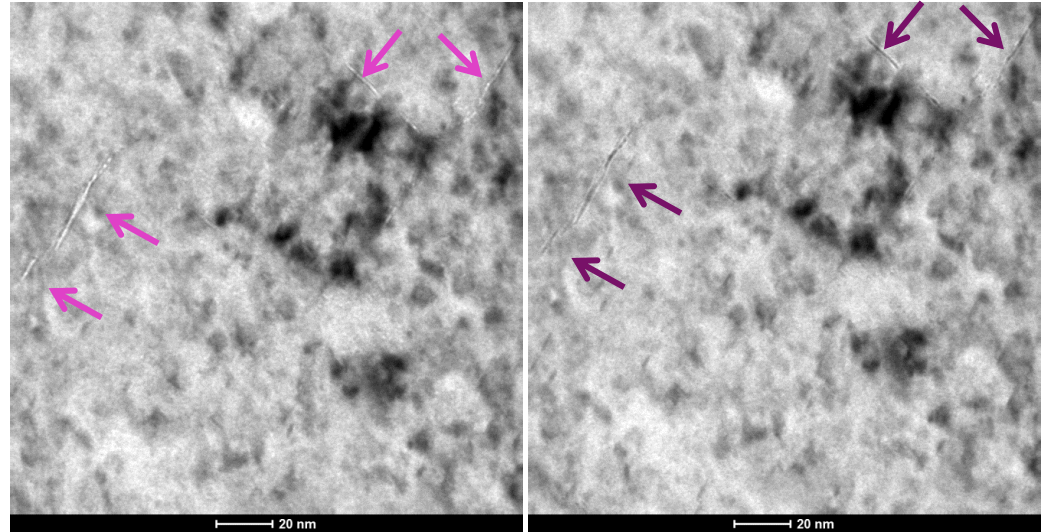
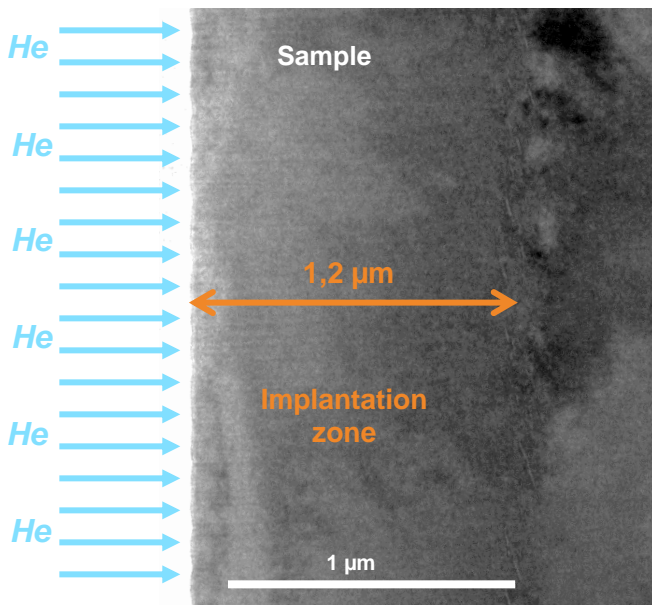
⇒ Observations of the samples with the more powerful TEM

⇒ Optimization step of the ImageJ method

⇒ Use the ImageJ method to characterize helium bubbles improving values and measurements uncertainties

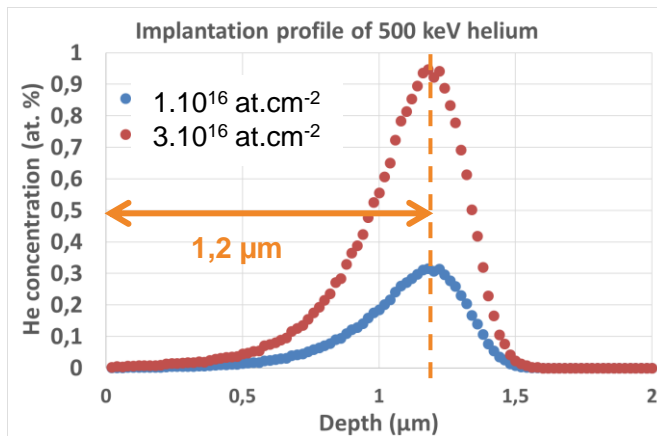
On 200 nm centered at the depth of the implantation peak

600°C and  $3.10^{16}$  at.cm<sup>-2</sup> implanted sample



Under-focused image

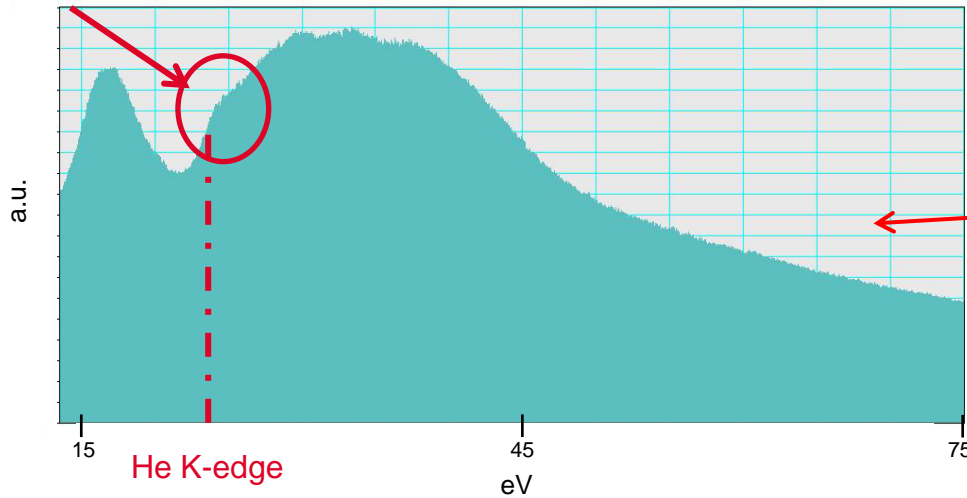
Over-focused image



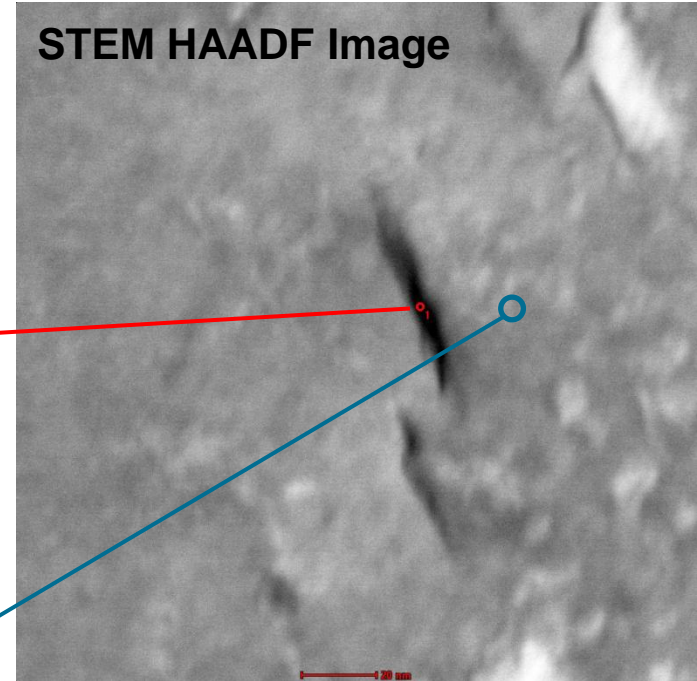
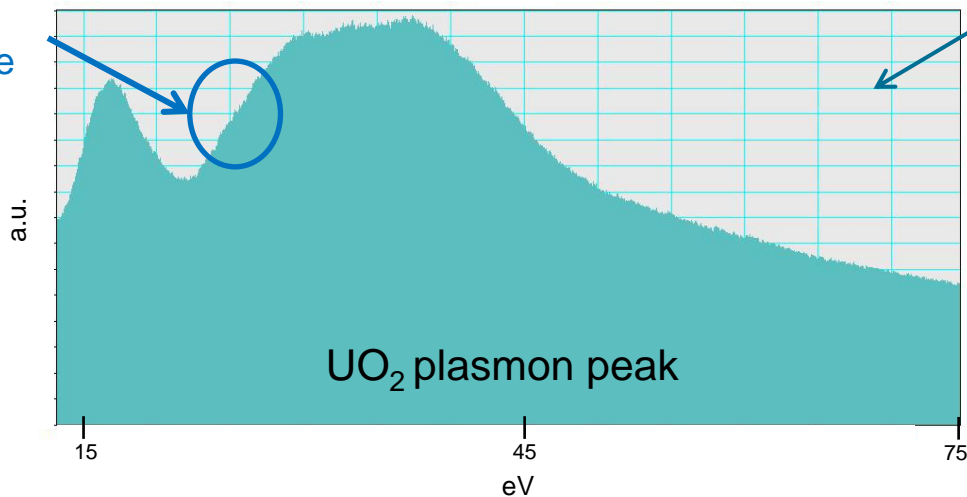
Annealing	Fluence (at.cm <sup>-2</sup> )	Temperature (°C)	Time (h)	Platelets
In-situ	1.10 <sup>16</sup>	200 / 400 / 600	/	No
	<b>3.10<sup>16</sup></b>	200	5,7	No
		400	10,3	No
		<b>600</b>	<b>6,5</b>	<b>Yes</b>
Ex-situ	1.10 <sup>16</sup>	800	1	No
	<b>3.10<sup>16</sup></b>	600	1	No
		<b>600</b>	<b>6</b>	<b>Yes</b>
		<b>800</b>	<b>1</b>	<b>Yes</b>

## EELS spectrum

Increase observed at the level of the He K-edge



No increase



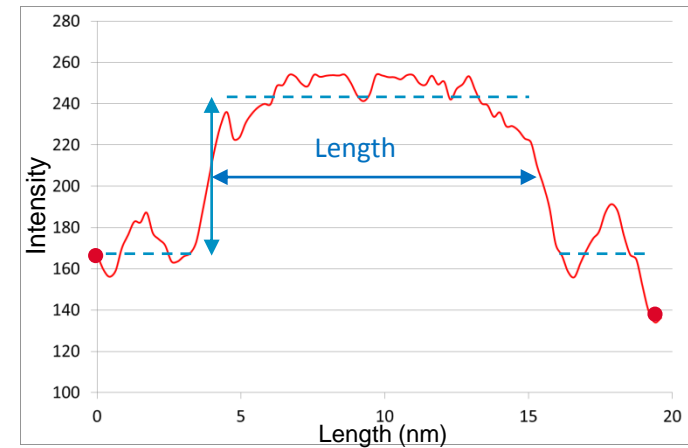
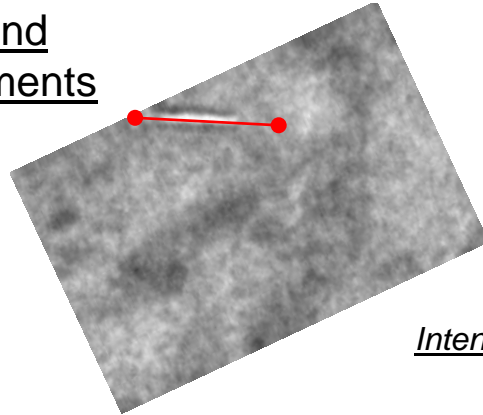
For a diluted gas, the K threshold is equal to 21.218 eV [1]

It can be shifted to the high value (blue shift) when He density increases [2]

[1] Kuhn, Atomic spectra (1962) 132  
[2] Fréchar et al. JNM 393 (2009) 102



## Step 1 : Length and thickness measurements



Implantation conditions (°C – at.cm <sup>-2</sup> )	Bulk annealing	Platelet length		Platelet thickness		Number of platelets	Surface density of platelets (platelets.m <sup>-2</sup> ) <i>On the layer of 200 nm thick</i>
		Minimum length (nm)	Maximum length (nm)	Minimum thickness (nm)	Maximum thickness (nm)		
600 – 3.10 <sup>16</sup>	No	17 ± 3	76 ± 15	0.4 ± 0.1	1.1 ± 0.5	12	(9.3 ± 2.7) .10 <sup>13</sup>
600 – 3.10 <sup>16</sup>	No	5 ± 1	49 ± 8	0.6 ± 0.1	3.9 ± 0.8	25	(8.4 ± 1.7) .10 <sup>13</sup>
RT – 3.10 <sup>16</sup>	800°C / 1 h	16 ± 3	77 ± 15	0.5 ± 0.1	2.1 ± 0.4	17	(3.7 ± 0.9) .10 <sup>13</sup>
RT – 3.10 <sup>16</sup>	600°C / 6 h	6 ± 1	93 ± 18	0.4 ± 0.1	0.9 ± 0.2	11	(1.4 ± 0.4) .10 <sup>14</sup>

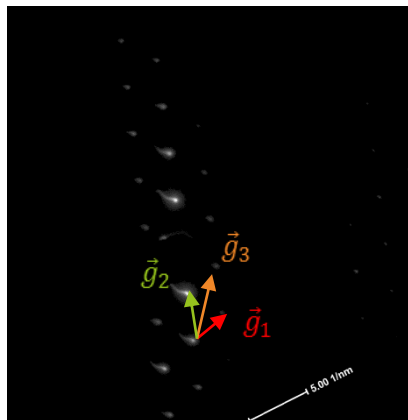
Length included between a few nanometers and around a hundred of nanometers

Thickness of about a few nanometers

Density of about 10<sup>13</sup>/10<sup>14</sup> platelets.m<sup>-2</sup>



## Step 2 : Diffraction patterns indexation



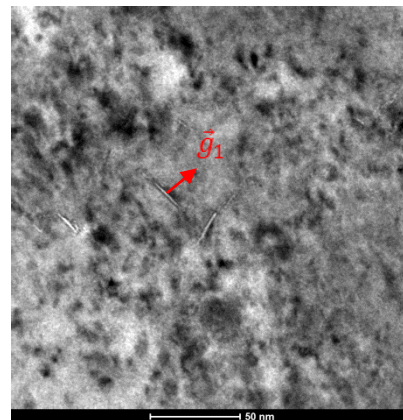
Interreticular distances measurements :

$$\vec{g}_1 = \langle 1 \ 1 \ 1 \rangle$$

$$\vec{g}_2 = \langle 0 \ 0 \ 2 \rangle$$

$$\vec{g}_3 = \langle 1 \ 1 \ 3 \rangle$$

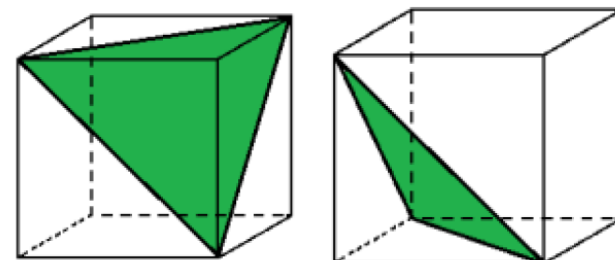
## Step 3 : Search of habit planes



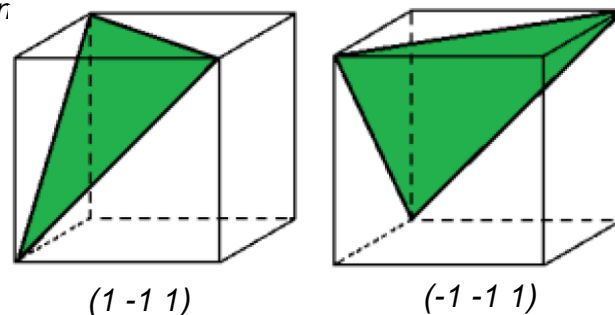
Platelets that are perpendicular to a diffraction vector  $\vec{g}$  lie in the habit plan that corresponds to  $\vec{g}$ . Here, for example, the platelet lie in the plane  $\{1 \ 1 \ 1\}$

Implantation conditions (°C – at.cm <sup>-2</sup> )	Annealing	Platelets number	Habit plane
600 – 3.10 <sup>16</sup>	No	25	{1 1 1}
			{1 1 1}
RT – 3.10 <sup>16</sup>	800°C / 1 h	17	{1 1 1}
RT – 3.10 <sup>16</sup>	600°C / 6 h	11	{1 1 1}

→ It seems that platelets preferentially appear in {1 1 1} type habit plans.



4 possible variants of the {1 1 1} habit plan in UO<sub>2</sub> :



# CONCLUSION ON HELIUM PRECIPITATION IN $\text{UO}_2$

## Bubbles

- Presence of helium bubbles for 600°C implantations for two fluences  $1.10^{16}$  and  $3.10^{16}$  at.cm<sup>-2</sup> : maximum size of 1 nm and a density of about  $10^{22}$  b.m<sup>-3</sup>
- Difficulties to highlight the influence of temperature and helium concentration on their features

Development optimized method of TEM images



To characterize helium bubbles populations with improving values and uncertainties

SAXS measurements



To obtain complementary characterizations on helium bubbles features

## Platelets

- Presence of helium platelets for an implantation in temperature or a post-implantation annealing at 600°C or 800°C for the higher fluence  $3.10^{16}$  at.cm<sup>-2</sup>
- Effect of annealing time on the presence or not of platelets.
- Helium platelets preferentially form in {1 1 1} type habit plans

Complementary TEM observations



To determine precisely habit planes with different variants



To study the intergranular behaviour of platelets

## He precipitation in $\text{UO}_2$

Influence of fission gases bubbles

Influence of irradiation damages

## Outlook

## He precipitation in $(\text{U,Pu})\text{O}_2$

TEM characterization on MOX containing radiogenic He

Thank you for your attention !

---

Contact :  
[adrien.michel2@cea.fr](mailto:adrien.michel2@cea.fr)