



HAL
open science

Atomic scale insights on the microstructure evolution of urania under irradiation

A. Chartier, L. van Brutzel, Claire Onofri, C. Sabathier-Devals, O. Dorosh, J. Jagielski

► **To cite this version:**

A. Chartier, L. van Brutzel, Claire Onofri, C. Sabathier-Devals, O. Dorosh, et al.. Atomic scale insights on the microstructure evolution of urania under irradiation. Nuclear Materials (NuMat - 2016), Nov 2016, Montpellier, France. cea-02438727

HAL Id: cea-02438727

<https://cea.hal.science/cea-02438727v1>

Submitted on 28 Feb 2020

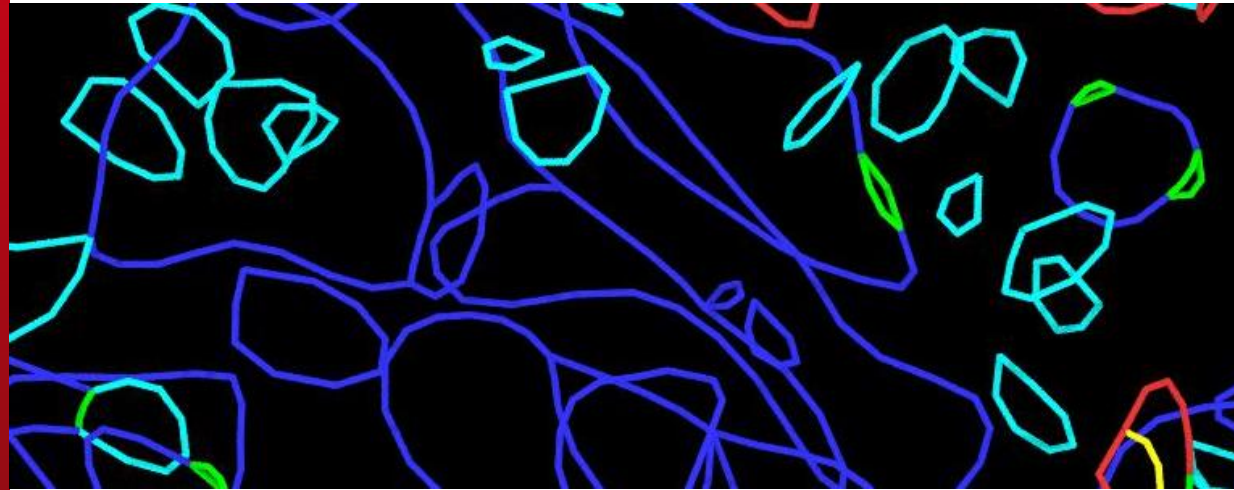
HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

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.

ATOMIC SCALE INSIGHTS ON THE
MICROSTRUCTURE EVOLUTION OF URANIA
UNDER IRRADIATION

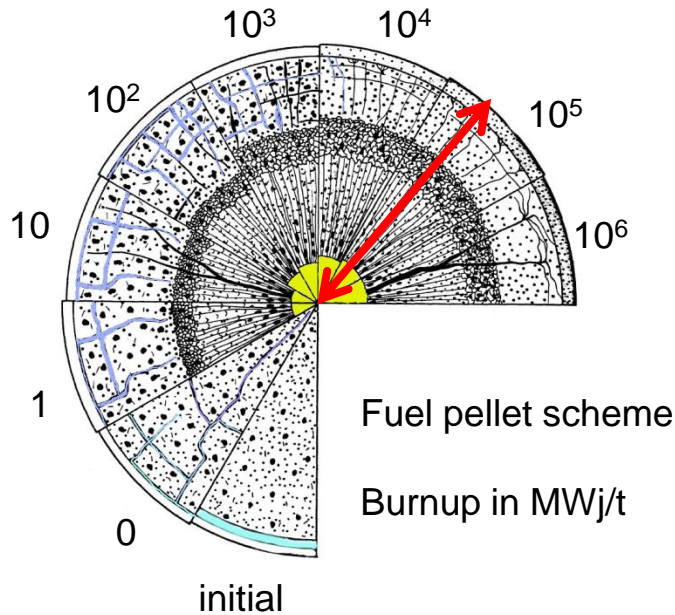
DE LA RECHERCHE À L'INDUSTRIE
cea den

université
PARIS-SACLAY



Alain Chartier, Laurent Van Brutzel, Claire Onofri, Catherine Sabathier,
Orest Dorosh and Jacek Jagielski

UO₂ nuclear fuel microstructure evolution



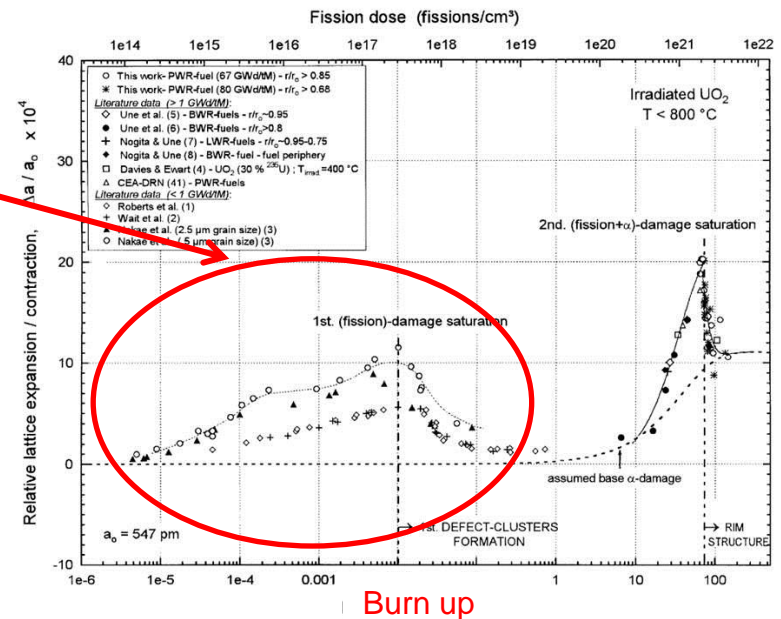
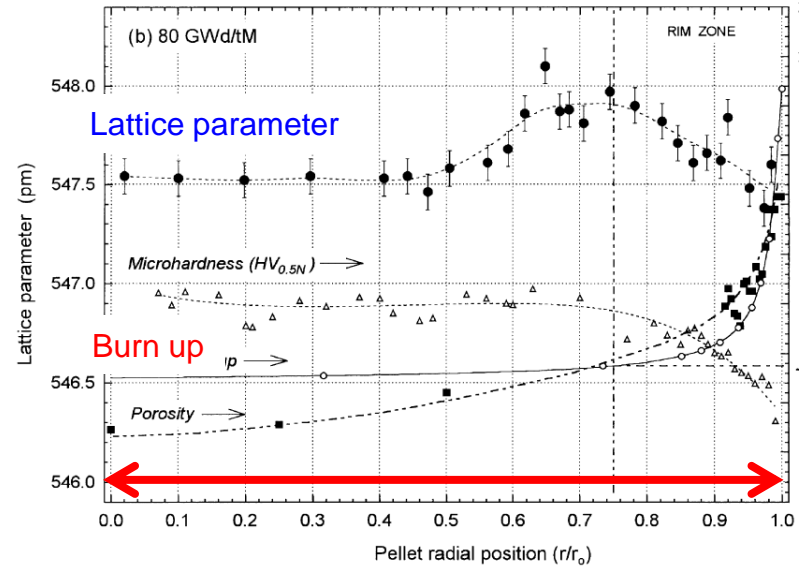
First dilatation-contraction peak:

1. point defects induce dilatation
2. Clustering and dislocations induce contraction

Rationale from:

- XRD mapped on rate equations
- TEM observations
- Ion irradiation experiments
- Mesoscopic modelling

But no direct insight of **the early stages** ← our goal



Relative lattice parameter

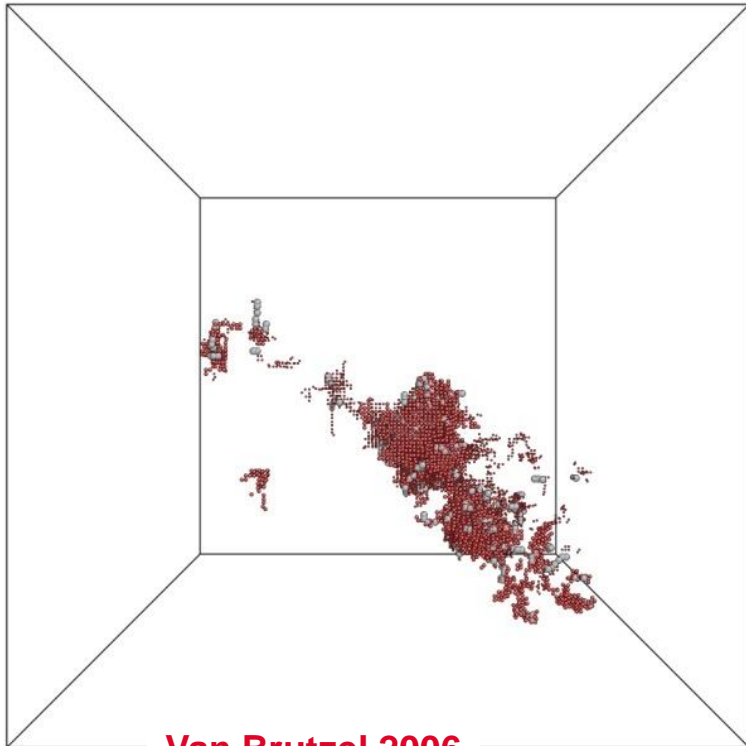
Molecular dynamics

Code: LAMMPS

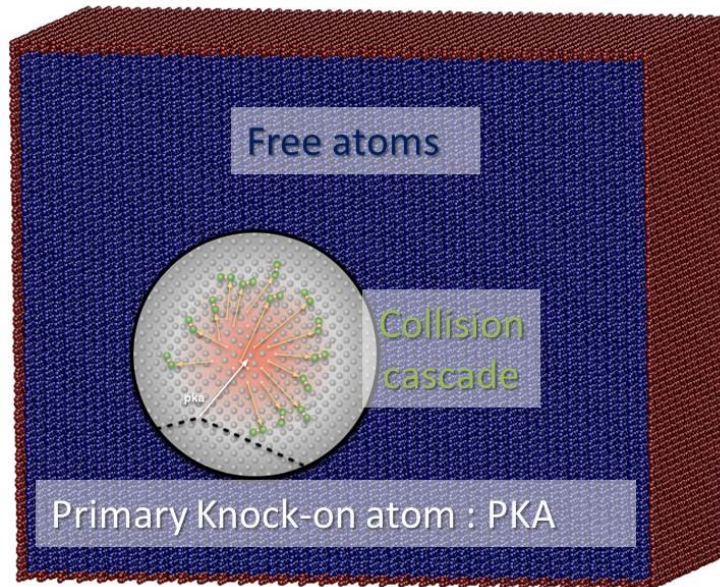
Empirical potentials: Morelon

Displacement cascades:

Primary Knock-on Atom: U

 $E_{\text{PKA}} = 80 \text{ keV}$ 

Thermostat : frozen atoms

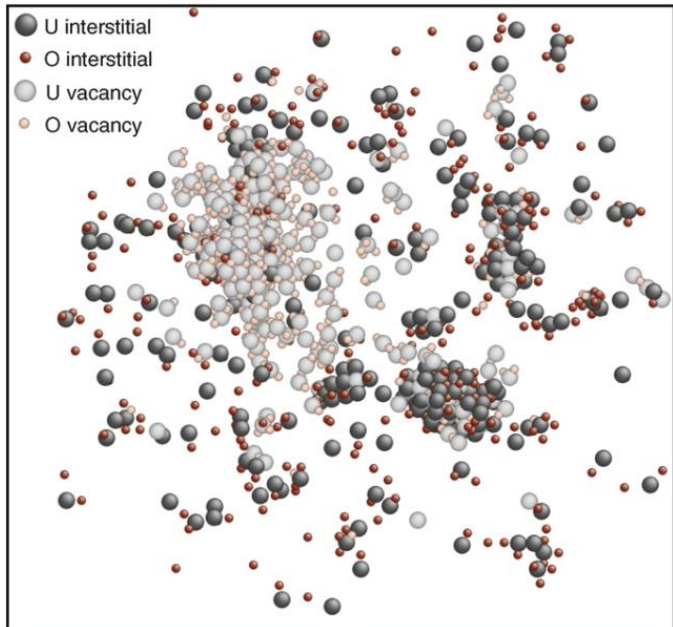


- no direct impact amorphisation
- small defects (interstitials / vacancies)

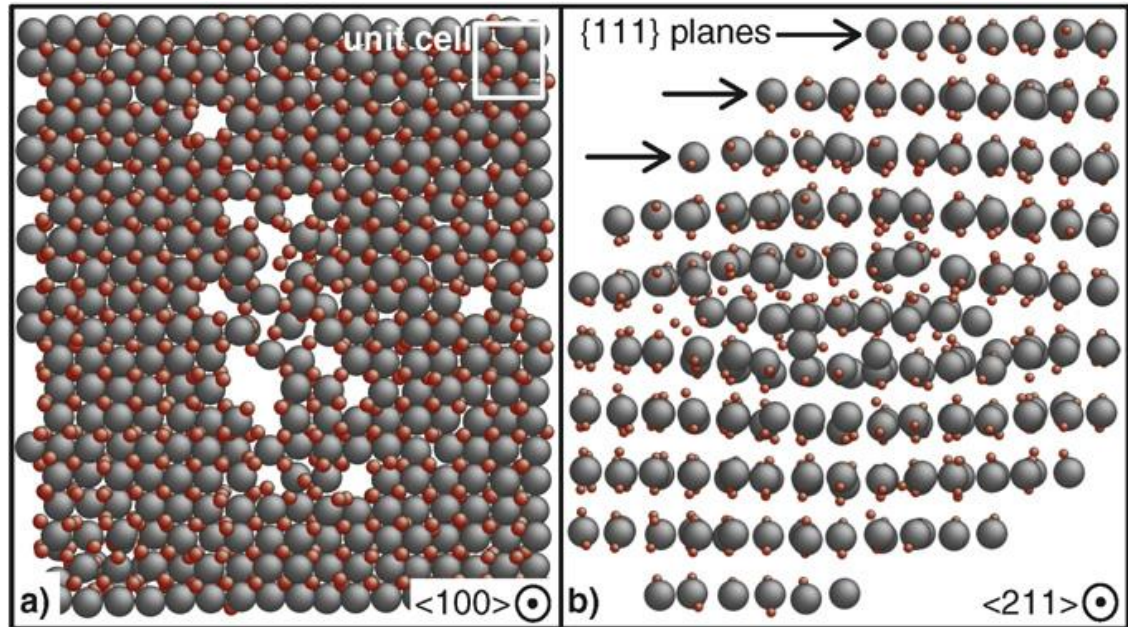
Primary damage in UO_2

Cascades overlap: $E_{\text{PKA}} = 10 \text{ keV}$ and 36 cascades

Martin 2010



Small defects



Cluster of vacancies

Dislocations

Dose effect: defects accumulation

Creation of Frenkel pairs every 2 ps

Dose rate: $1.6 \cdot 10^9$ dpa/s

Dose: $\rightarrow 10$ dpa

80 - 300 - 900 K - with constant pressure and temperature

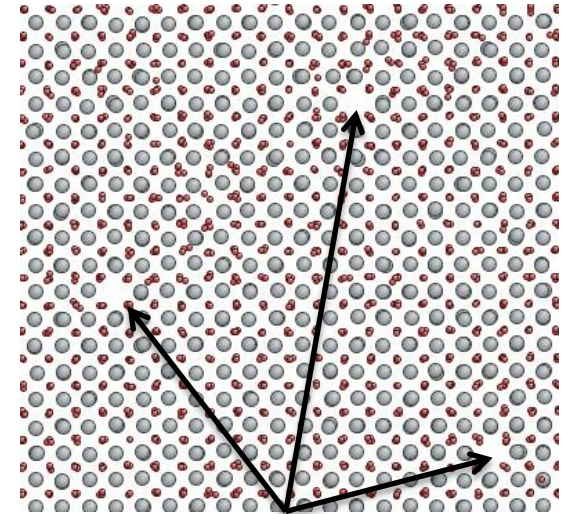
Analysis:

Dislocations with DXA algorithm

Defects with Voronoï

Lattice expansion - contraction from XRD

HRTEM and SAED



Frenkel pairs

Experiments versus calculations:

Dose rate : 10^{-3} dpa/s $\rightarrow 34 \text{ \AA}$ between two displaced Uranium after 1 second

$D(\text{U at } 900\text{K}) = 10^{-31} \text{ m}^2/\text{s} \rightarrow 1.18 \cdot 10^{14} \text{ s}$ for travelling 34 \AA

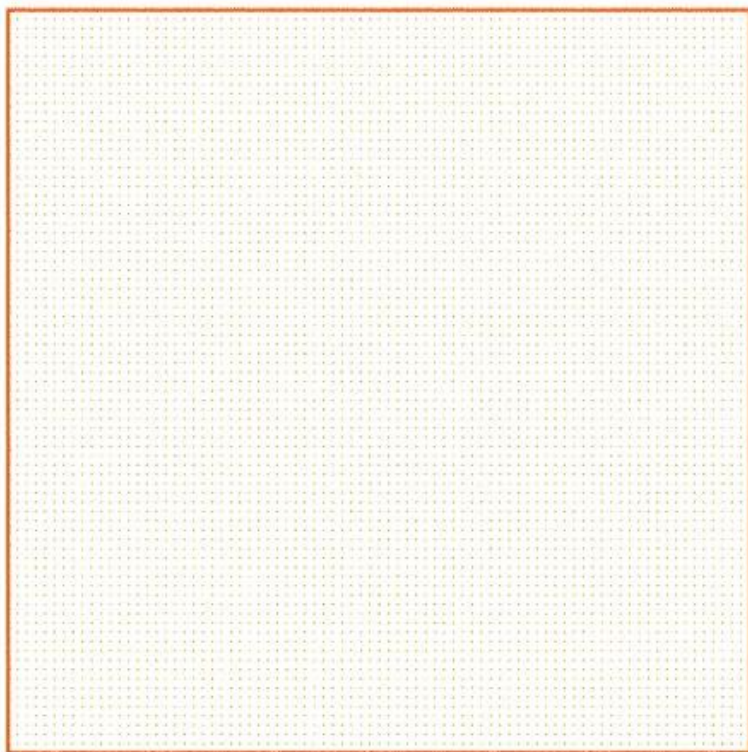
\rightarrow **recombination regime !**

Dislocation rescaling (Rest 2004) with $\rho_d \propto K_0^{1/6} \rightarrow \rho_{\text{norm}} \approx 9.3 \times 10^{-3} \rho_{\text{calc}}$.

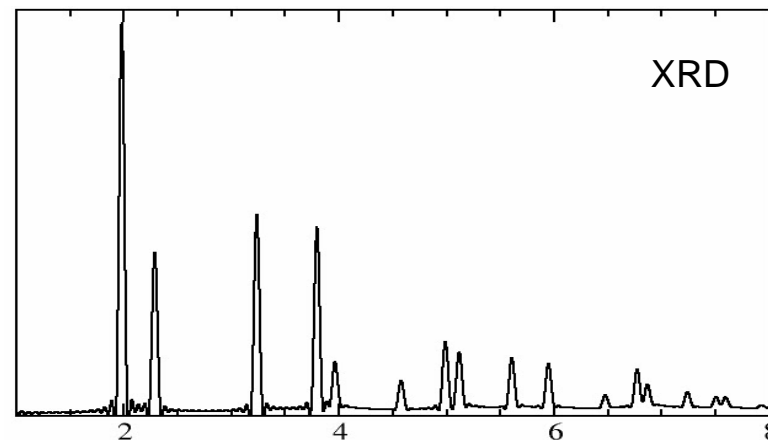
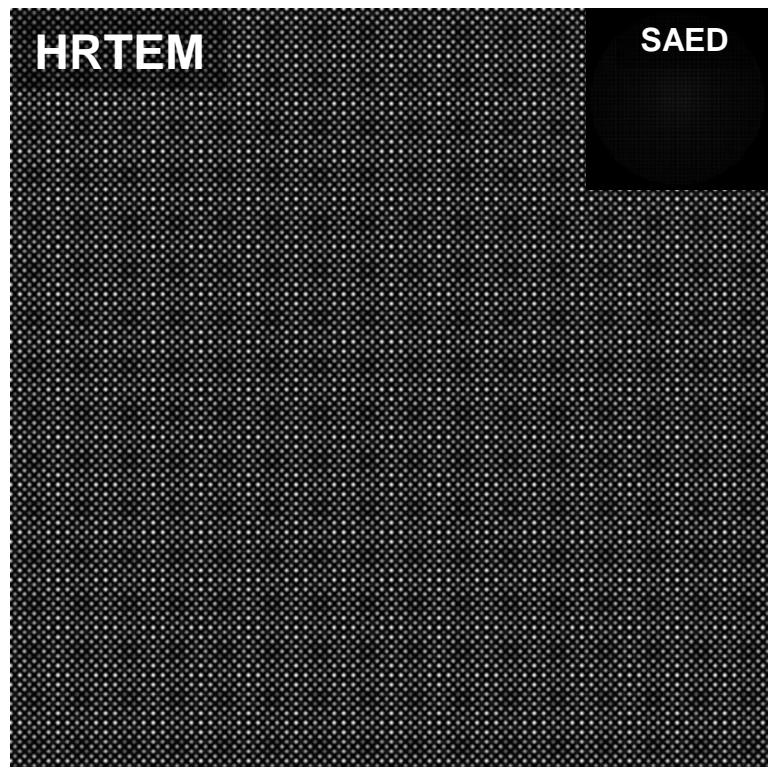
Dose effect from MD - Analysis from curvature criterion, HRTEM, SAED and XRD calculations

Dose = 0.0000 dpU

- Perfect dislocation
- Frank dislocation
- Shockley dislocation
- Undefined

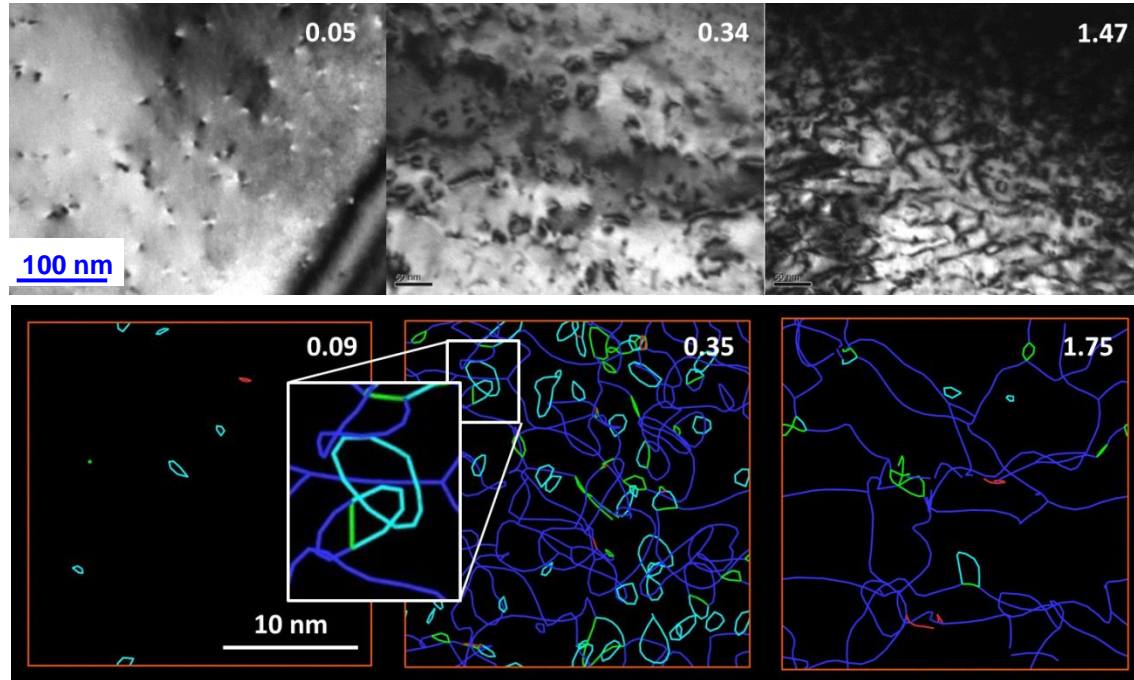
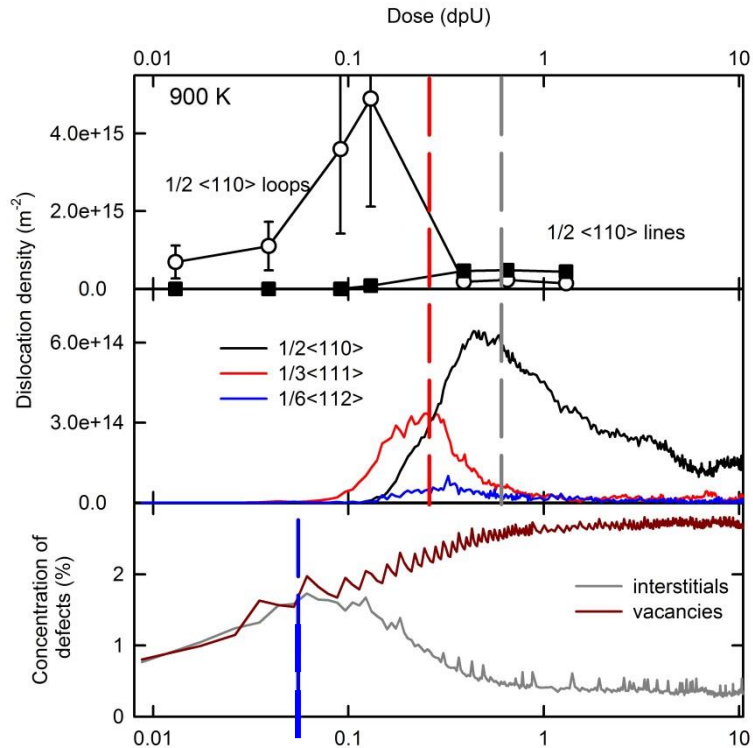


A. Chartier - CEA Saclay



- Visualization of dislocations - MD and HRTEM
- Nanodomains - MD and HRTEM
- Dislocation density - MD
- Swelling from size of box - XRD
- no amorphization - SAED and XRD

Irradiation induced dislocations in UO₂



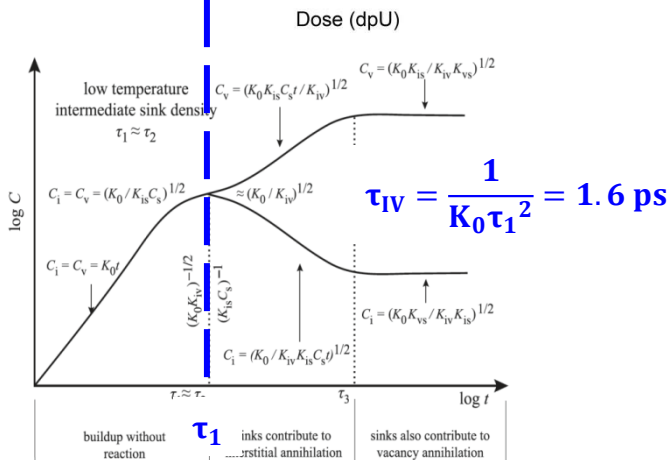
Exp: *not seen* ↔ *smaller than 2 nm* ½ <110> loops → ½ <110> lines

MD: point defects → ⅓ <111> loops → ½ <110> loops → ½ <110> lines
 Frank loops perfect loops

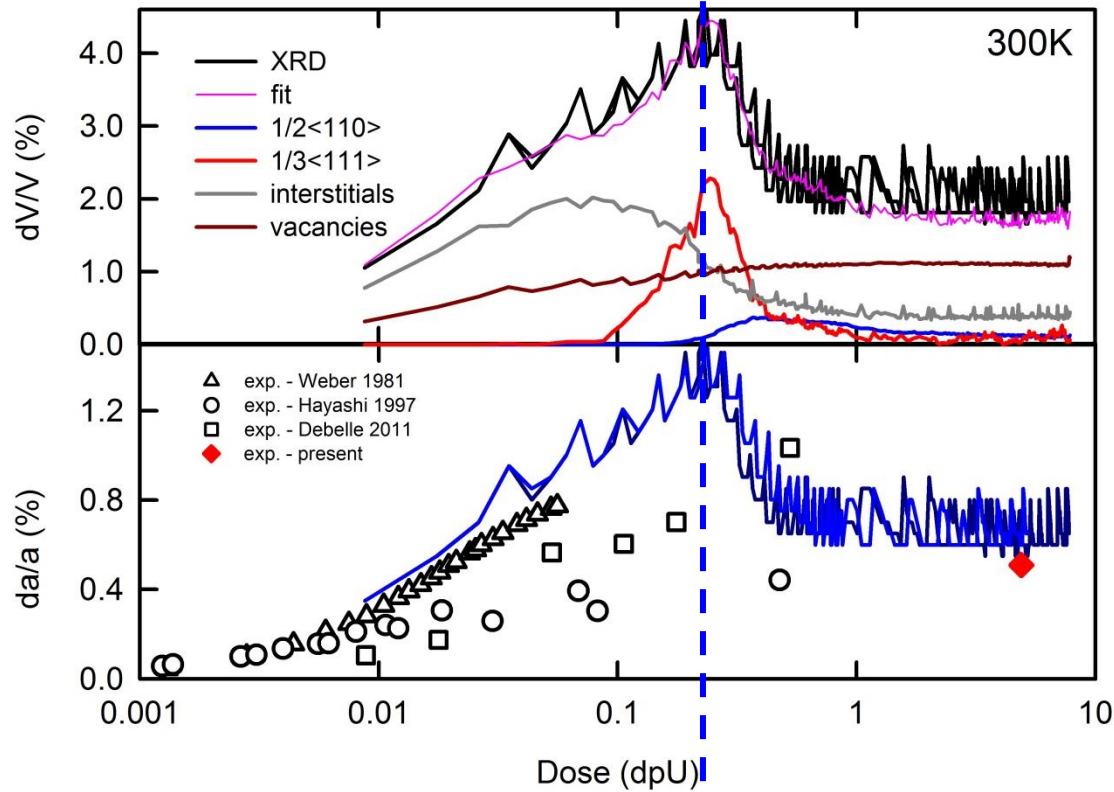
dislocation density: MD = 4 10¹⁴ m⁻² and Exp. = 6 10¹⁴ m⁻² (rescaled)

Nota:

- point defects behave as expected from rate equations
- 1.6 ps corresponds to spontaneous recombination regime (Van Brutzel 2008)



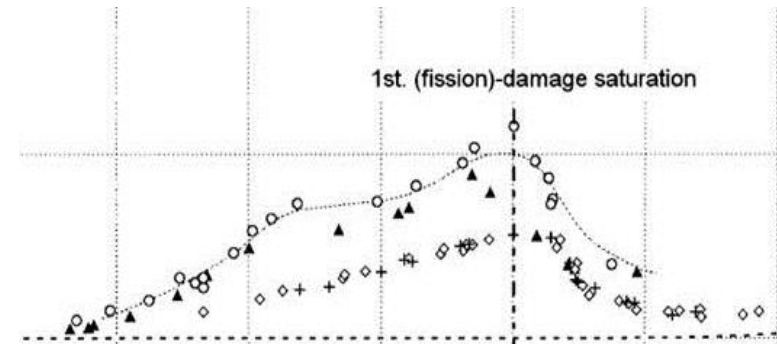
Lattice evolution with irradiation dose



$$\frac{\Delta V}{V} = V_I^f \times \rho_{line} + \frac{\overline{R_{loop}} \times b}{2} \times \rho_{loop} + \frac{V_{rel}^I}{\Omega} \times C_I + \frac{V_{rel}^V}{\Omega} \times C_V$$

with $V_I^f = 5.6 \text{ \AA}^2$; $\overline{R_{loop}} \sim 3 \text{ nm}$; $V_{rel}^I = V_{rel}^V = 1. \Omega$

Lattice expansion - contraction comparable to measurements on nuclear fuel



Expansion:

- interstitials and vacancies
- $1/3\langle 111 \rangle$ Frank loops

Contraction:

- transformation of Frank loops into perfect loops
- vacancies at steady state

Mechanism of irradiation induced dislocations in UO_2 :

- Molecular dynamics: point defects $\rightarrow \frac{1}{3}\langle 111 \rangle$ loops $\rightarrow \frac{1}{2}\langle 110 \rangle$ loops $\rightarrow \frac{1}{2}\langle 110 \rangle$ lines
- Experimental observations: *not seen* \leftrightarrow *smaller than 2 nm* $\frac{1}{2}\langle 110 \rangle$ loops $\rightarrow \frac{1}{2}\langle 110 \rangle$ lines

Expansion - contraction peak:

- Expansion: point defects and then Frank loops
- Contraction: transformation of Frank loops in unfaulted dislocations.

\rightarrow re-interpretation of accepted picture

Recombination regime arguments:

- Comparison between dose rates and characteristic time for diffusion \Rightarrow recombination regime
- Link between MD and exp. dislocation density with renormalisation formula \Rightarrow recombination regime

Thank you for listening

Questions ?

Commissariat à l'énergie atomique et aux énergies alternatives
DEN/DANS/DPC/SCCME/LM2T bât. 450S
Centre de Saclay | 91191 Gif-sur-Yvette Cedex
T. +33 (0)1 69 08 31 68 | F. +33 (0)1 69 08 92 21

Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

Direction de l'énergie Nucléaire
Département de Physico-Chimie
Service de Corrosion et du
Comportement des Matériaux dans
leur Environnement