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Insight into the behavior of nuclear fuels through investigations coupling multi-scale modeling and separate effects experiments

G. Jomard

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DE LA RECHERCHE À L'INDUSTRIE



INSIGHT INTO THE BEHAVIOR OF NUCLEAR FUELS THROUGH INVESTIGATIONS COUPLING MULTI- SCALE MODELING AND SEPARATE EFFECTS EXPERIMENTS

NuMat 2018

The Nuclear Materials Conference
14–18 OCTOBER 2018 • SEATTLE, WA, USA

materialstoday
Connecting the materials community

Gérald Jomard *et al.*

CEA
Nuclear Energy Division
Fuel Research Department
Fuel behaviour study and simulation Group
Nuclear Fuels Multi-scale Modeling Lab



www.cea.fr

Fuel R&D « loop » at CEA/DEN

QUALIFICATION

DESIGN

PLEIADES

**MODELLING/
SIMULATION**

**BASIC
RESEARCH**

**CHARACTERIZATION
AFTER IRRADIATION**

Hot labs i.e. LECA

**MANUFACTURING/
CHARACTERIZATION**

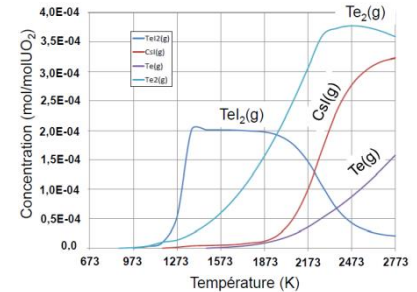
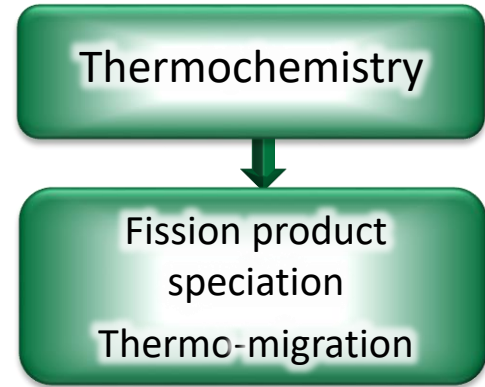
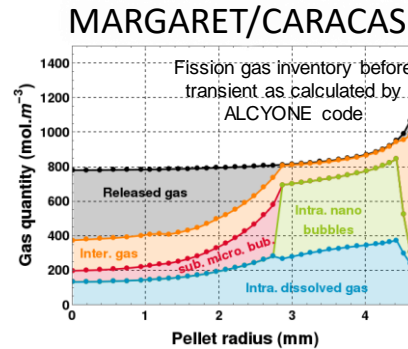
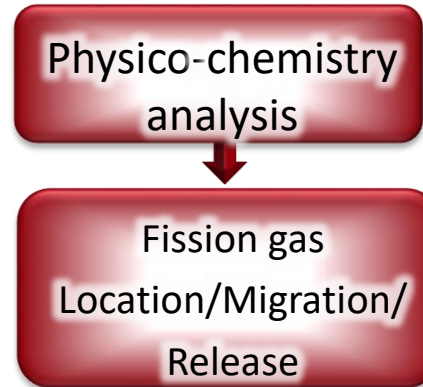
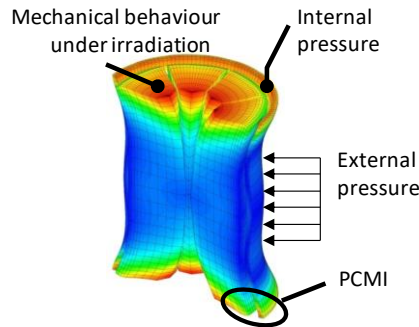
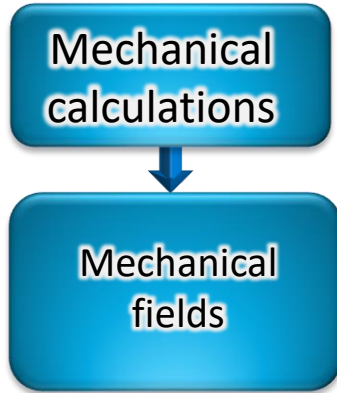
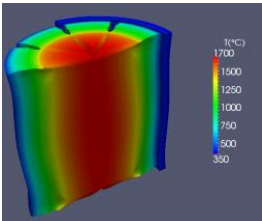
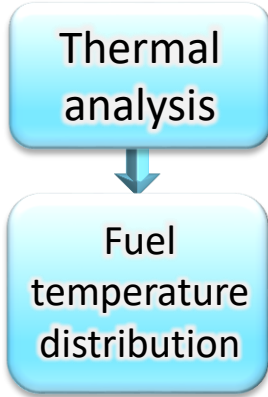
LCU, Hot labs ATALANTE

IRRADIATION TEST

Integral irradiation
Separate effects or analytical tests
(Ion irradiation/implantation;
thermal treatment after irradiation; MTR irradiation)

Simulation of nuclear fuels behaviour at the macro scale

Input from neutronics, thermo-hydraulics



C. Introini
NuMat 03.25

J-C. Dumas
NuMat 03.02

Progress in physical description to improve Fuel Performance Codes predictivity



Need for justified simplifications to enable use in industrial code

⇒ Supported by basic research

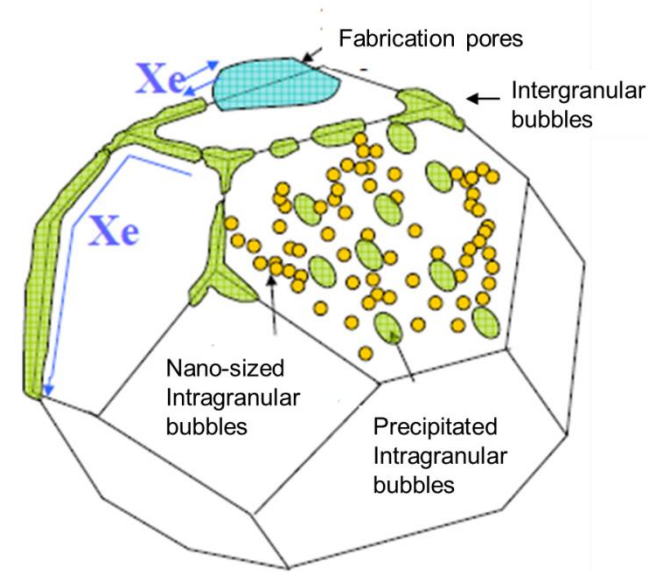
Simulation of FG behavior at the grain scale

■ For guaranteeing fuel rod integrity, in-pile FG behavior is of major concern

→ Most up-to-date model (MARGARET) has been validated on a huge PIE database

Many observations are correctly described:

- Fission gas release in nominal conditions as well as in power ramp
- Fission gas repartition : intra/inter-granular, dissolved/trapped in bubbles
- Fission gas swelling



■ But...

→ some empiricism need to be verified (e.g. **bubble size distribution**)

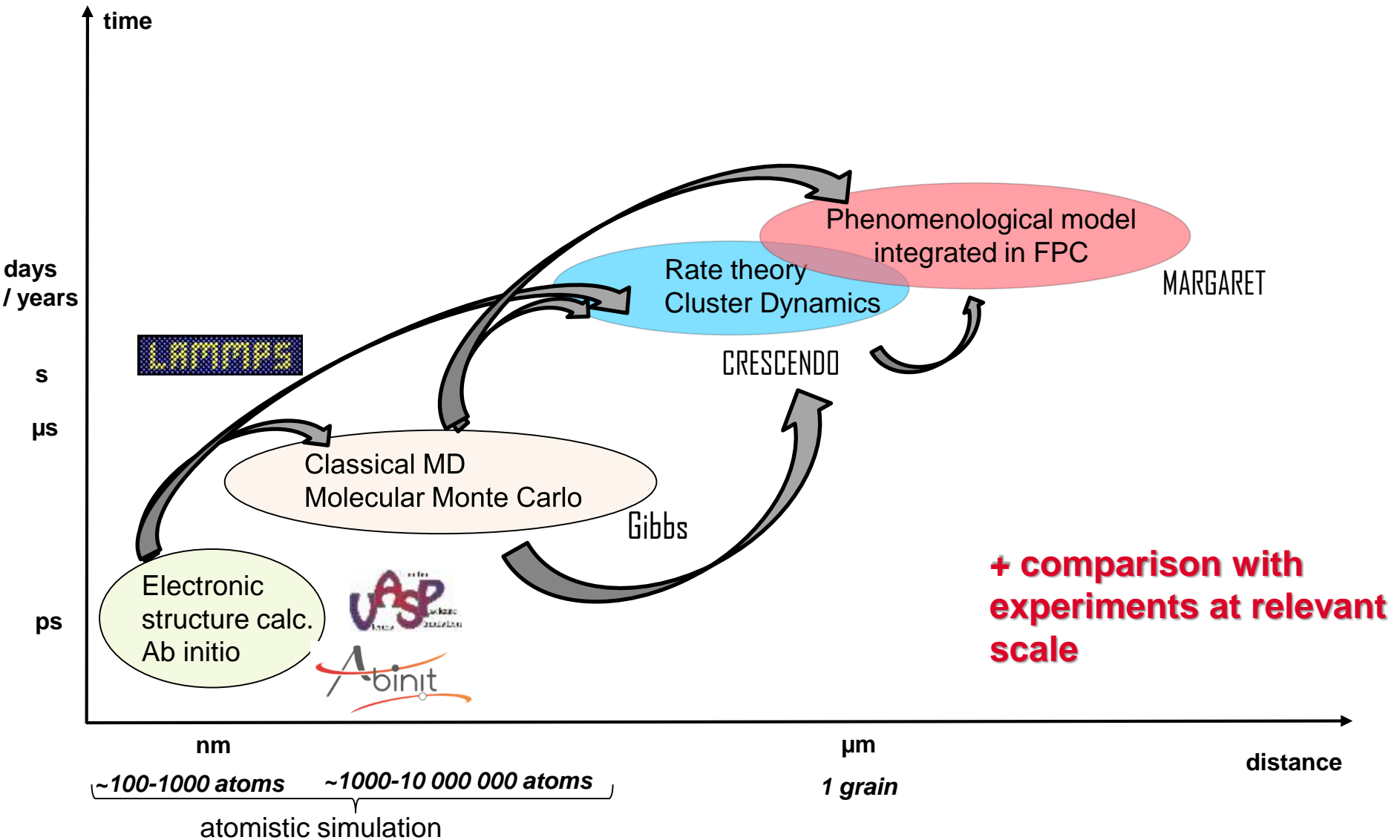
→ **unable to describe thermal annealing of spent fuel**

→ more physics will allow more predictivity (what happens outside the database regime?)



Coupling between multi-scale modeling approach and separate effects experiments to challenge the model

Multiscale modeling of FG and defect behavior at the grain scale



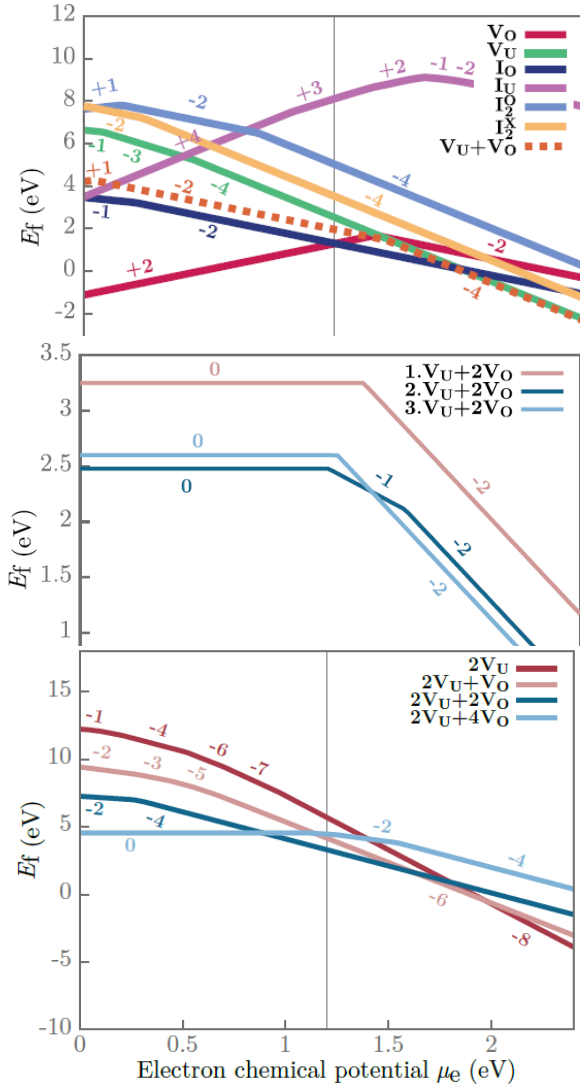
Study of Point defects and Fission Gases in UO_2

Formation
Incorporation
Migration

Formation of point defects in UO₂

Defect stability from ab initio calculations

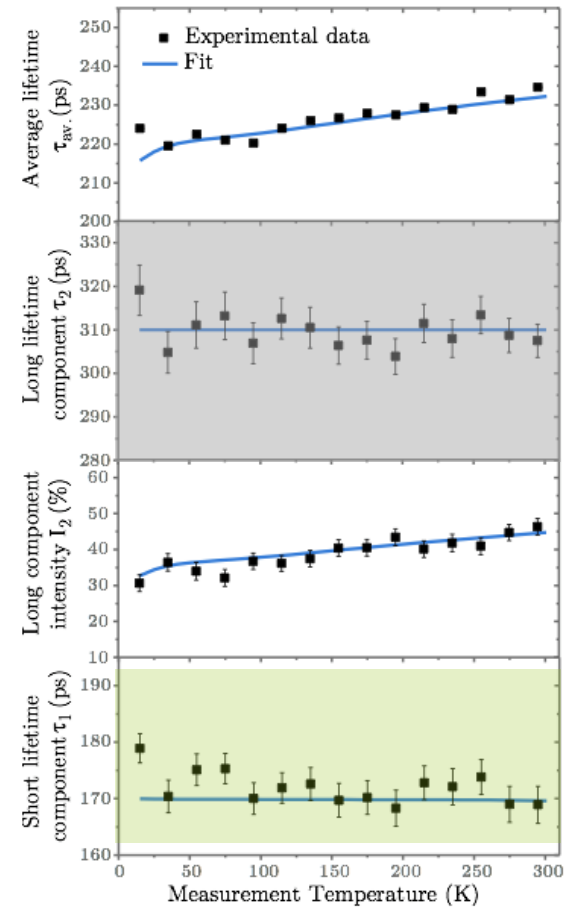
$$\Delta E_{\text{form}}(X^q) = E_{\text{tot}}(X^q) - n_U \mu_U^{\text{UO}_2} - n_O \mu_O^{\text{UO}_2} + q\mu_e + \Delta E_{\text{corr}}$$



Application of Positron Annihilation Lifetime Spectroscopy

Associated positron lifetimes

	τ (ps)
Lattice	167
V _O	199
V _U	304
V _U ⁴⁺	293
V _U +V _O	306
V _U +V _O ²⁻	301
V _U +2V _O	313
V _U +V _U	318
V _U +V _U ⁸⁻	289
2V _U +2V _O	339
2V _U +2V _O ⁴⁻	319
2V _U +4V _O	329
2V _U +4V _O ²⁻	365



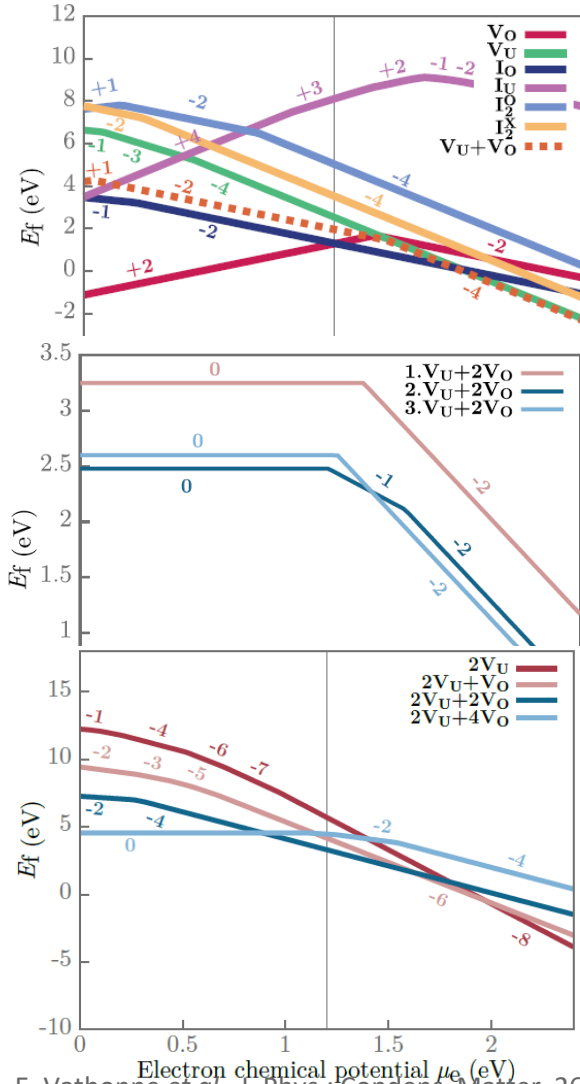
UO₂, 1700°C/24h/ArH₂
O/U=2.005±0.005

Irradiated with α 45 MeV, $3 \times 10^{16} \text{cm}^{-2}$

Near stoichiometric UO₂, V_O²⁺ and I_O²⁻ most stable defects

Formation of point defects in UO₂

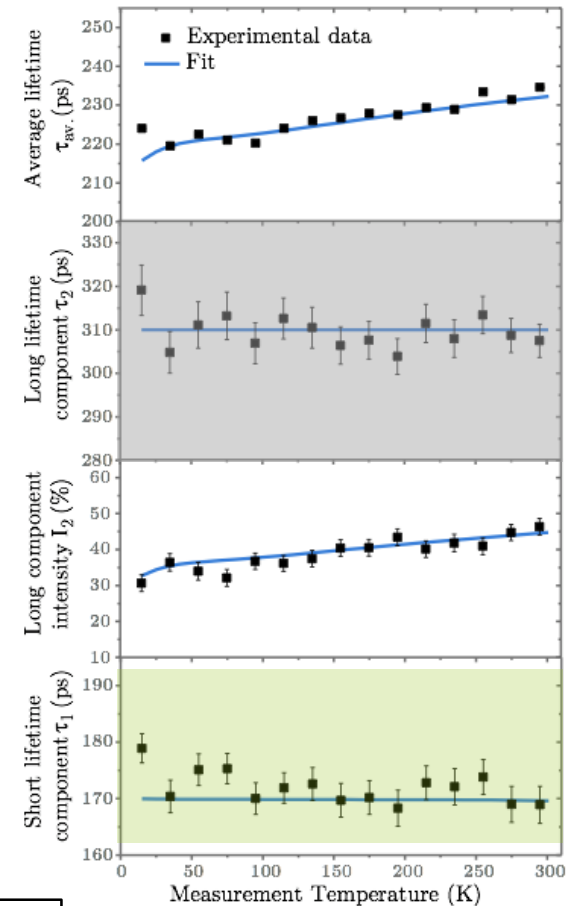
Defects stability from ab initio calculations $\Delta E_{\text{form}}(X^q) = E_{\text{tot}}(X^q) - n_U \mu_U^{\text{UO}_2} - n_O \mu_O^{\text{UO}_2} + q \mu_e + \Delta E_{\text{corr}}$



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Lattice	167
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2V _U +2V _O	339
2V _U +2V _O ⁴⁻	319
2V _U +4V _O	329
2V _U +4V _O ²⁻	365

V_U+2V_O (Schottky defect) predominant
Negative vacancies – probably mix of
V_U⁴⁻, V_U+V_O²⁻, and 2V_U+2V_O⁴⁻



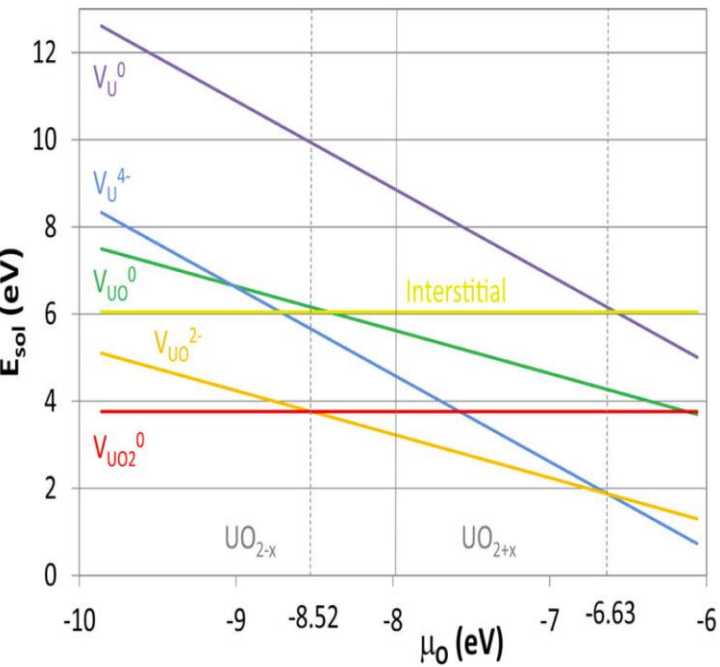
E. Vathonne *et al.*, *J. Phys.: Condens. Matter.* 26, 349601 (2014)

Wiktor *et al.*, *Phys. Rev. B* 90, 184101 (2014)

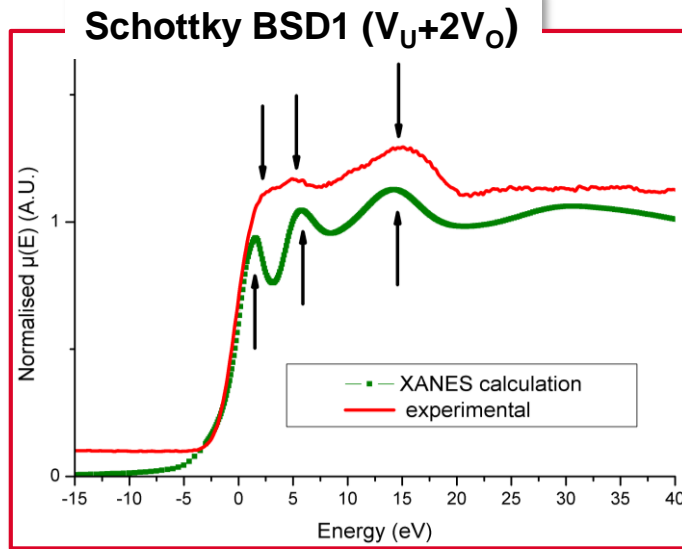
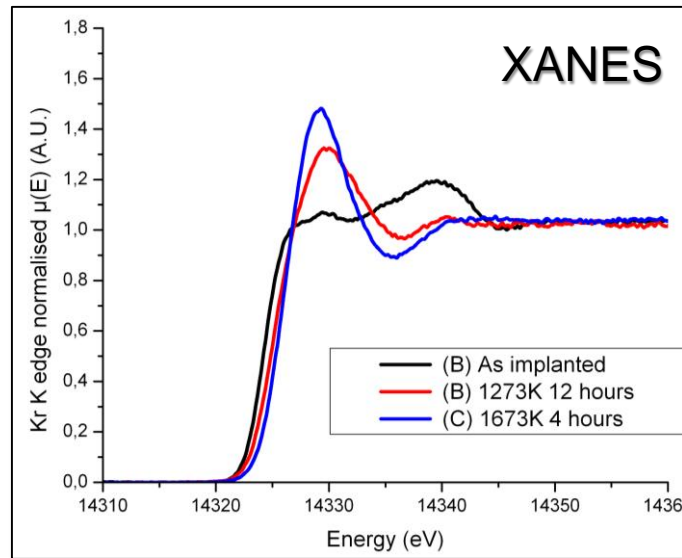
Incorporation of Fission Gases in UO_2

Kr E_{solution} from ab initio calculations

$$\Delta E_{\text{sol}}(Kr, V_X, q) = E_{\text{tot}}(Kr, V_X, q) - n_U \mu_U - n_O \mu_O + n_{Kr} \mu_{Kr} + q(E_{VBM} + \mu_e + \Delta V) + \Delta E_{\text{el}}(q)$$



Near stoichiometry Kr incorporated in $V_{UO_2}^0$ or V_{UO}^{2-}



For as implanted 0.5 at.% Kr samples

- Kr atoms surrounded by U and O atoms only (**no Kr aggregate**)
- Kr atoms as single atoms inside neutral **bound Schottky defects** with O vacancies aligned along the (100) direction (BSD1)

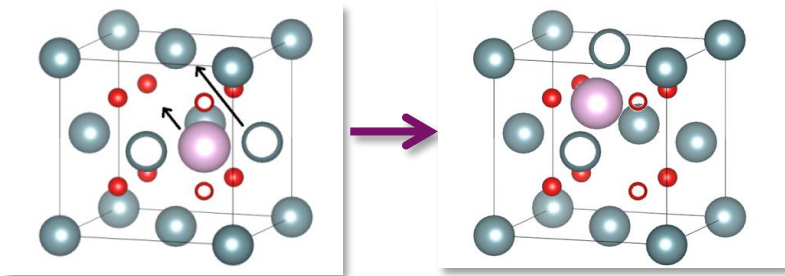
→ Nano-cavities observed by TEM after rare gas implantation are empty

Ph. Martin *et al.* J. Nucl. Mat. 466, 379 (2015)

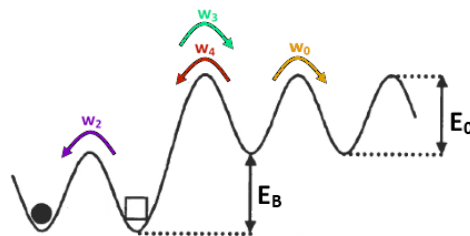
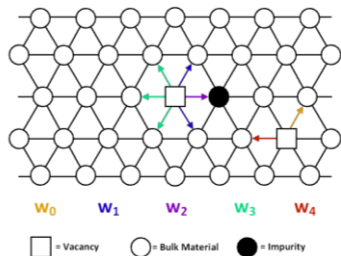
E. Vathonne, PhD thesis (2014)

Migration of Point defects and Fission Gases in UO₂

Elementary mechanisms and energy (NEB, ab initio)
 Attempt jump frequencies (empirical potential)



Approximation : no interaction beyond 2nd neighbours
 ⇨ 5-frequency model



Kr migration mechanism depends on stoichiometry

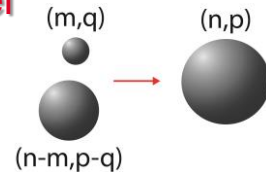
- UO_{2-x} : Schottky defect and charged U vacancy
- UO₂ : charged U-O divacancy and charged U vacancy
- UO_{2+x} : 2 charged U vacancies

→ similar to previous study of Xe

[Andersson *et al* PRB 84, 054105 (2011)]

E. Vathonne *et al.*, Inorg. Chem. 56, 125 (2017)
 R. Skorek *et al*, DIMAT 2011 323-325, 209 (2012)
 S. Maillard *et al.*, F-BRIDGE deliverable D225 (2012)
 A. Le Prioux, Ph.D. Thesis (2015-2017)

Data needed to parametrize a Cluster Dynamics model



$$\frac{dC(n,p)}{dt} =$$

$$\sum_{(m,q) \text{ mobile}} J_{(n-m,p-q)+(m,q) \leftrightarrow (n,p)}$$

Absorption of mobile cluster (m,q) by cluster (n,p)

$$- \sum_{(m,q) \text{ mobile}} J_{(n,p)+(m,q) \leftrightarrow (n+m,p+q)}$$

Emission of mobile cluster (m,q) from cluster (n,p)

$$+G(n,p) \quad \text{Source term}$$

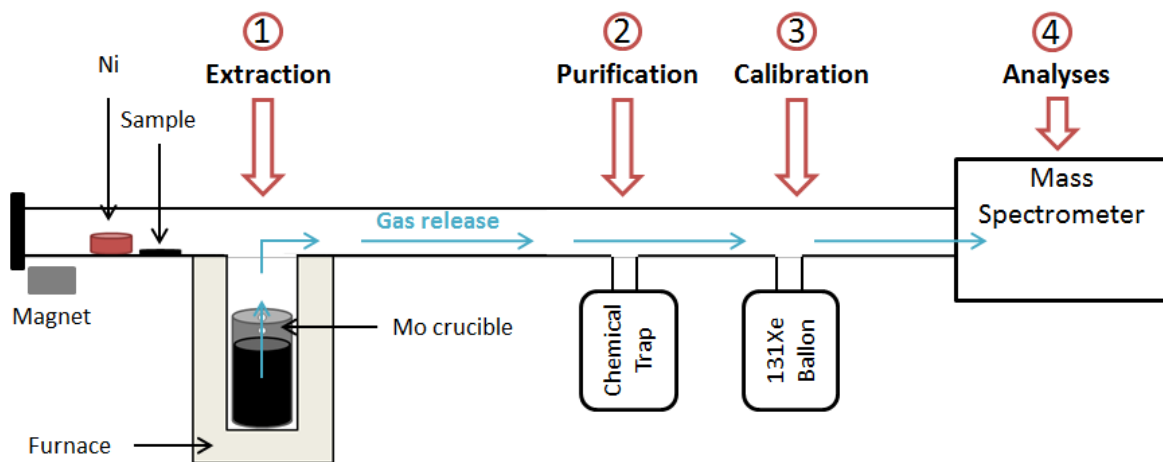
$$-K(n,p) \quad \text{Sink term}$$

Comprehensive framework to calculate defect and defect cluster (cavities / bubbles / dislocation loops) concentrations over time, with time scale and length scale appropriate for fuel study (*years, grains*)

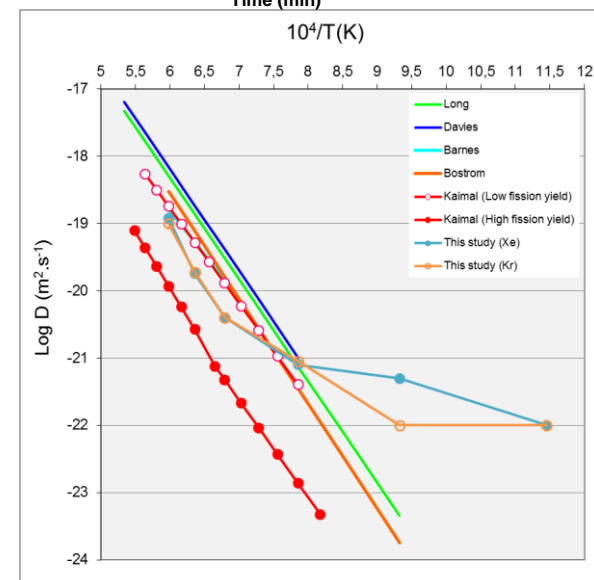
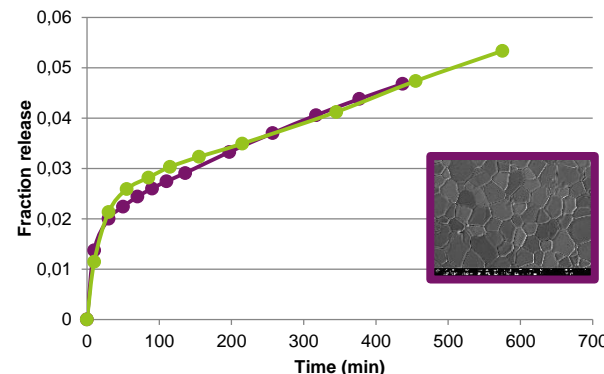
Cluster dynamics to interpret TDS exp. with Xe or Kr implanted fresh UO_2

- Ion implantation coupled to **Thermal Desorption Spectrometry** on **PIAGARA** Platform: Analyze low content of rare gases (detection limit $\sim 10^7$ atoms)
- **Isothermal release** of xenon or krypton from UO_2

CENBG
CEMHTI
IPNL



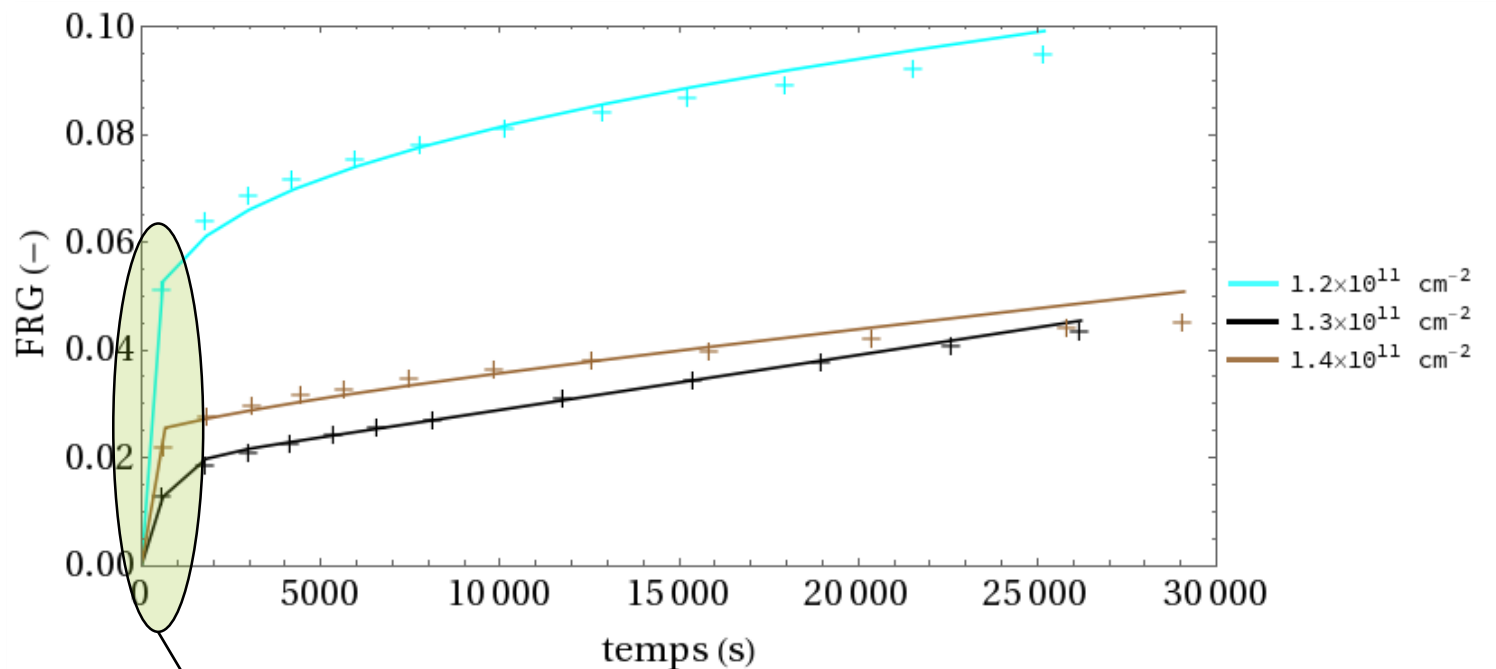
Isothermal release at 1300°C of Xe 129 implanted in UO_2 in polycrystal (purple) or single crystal (green)



- Arrhenius law for low contents of Xe and Kr implanted UO_2
→ Determination of **intragranular atomic Kr/Xe diffusion coeff**

Cluster dynamics to interpret TDS exp. with Xe or Kr implanted virgin UO_2

- Mobile species: I_{U} , V_{U} , I_{Xe} , $V_{2\text{U}}$, $\text{XeV}_{2\text{U}}$ —→ Mobilities from atomistic simulation
- C_v^0 : point defects concentration induced by implantation
- Initially Xe in interstitial site



800 keV Xe
implanted UO_2
1573 K annealing

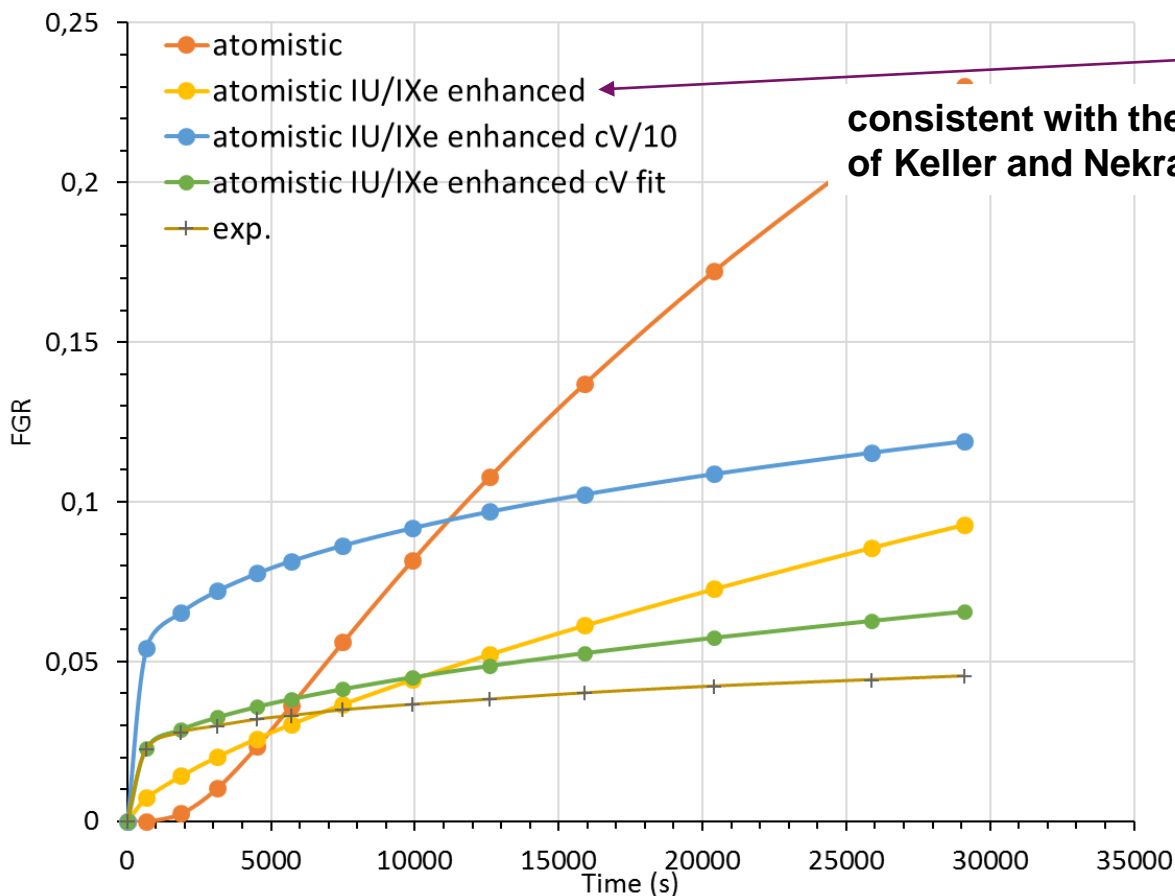
Initial burst release not reproducible → interpretation is difficult

Migration of Point defects and Fission Gases in UO_2

Cluster dynamics to interpret TDS exp. with Xe or Kr implanted virgin UO_2

- Mobile species: $I_U, V_U, I_{Xe}, V_{2U}, XeV_{2U}$ → Mobilities from atomistic simulation
- C_v^0 : point defects concentration induced by implantation
- Initially Xe in interstitial site

Parameters	Ab initio
E_m^{VU} (eV)	4,72
E_m^{IU} (eV)	0,7 (4,1)
E_m^{IXe} (eV)	2,2 (5,75)
$E_m^{XeV_{2U}}$ (eV)	3,50
$E_m^{V_{2U}}$ (eV)	2,84
$(D_0)^{VU}$ (cm ² /s)	0,00712
$(D_0)^{IU}$ (cm ² /s)	0,03 (N.C.)
$(D_0)^{IXe}$ (cm ² /s)	0,03 (0,0075)
$(D_0)^{XeV_{2U}}$ (cm ² /s)	0,148
$(D_0)^{V_{2U}}$ (cm ² /s)	0,545

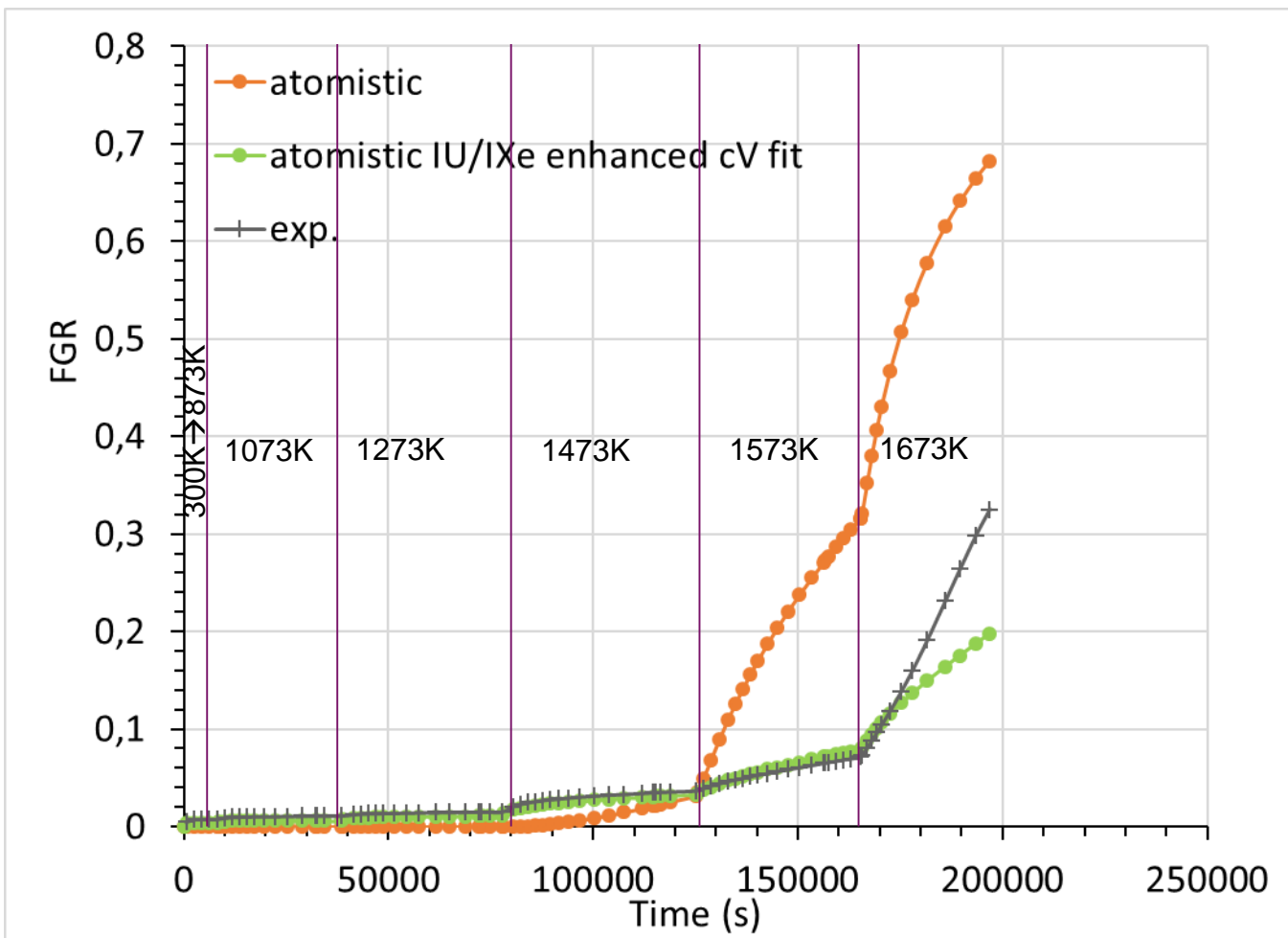


$$C_v^0 = 7 \cdot 10^{18} \text{ I/V per cm}^3$$

$$\rightarrow \text{Fit} : 1,7 \cdot 10^{18} \text{ I/V per cm}^3$$

800 keV Xe implanted UO_2
1573 K annealing

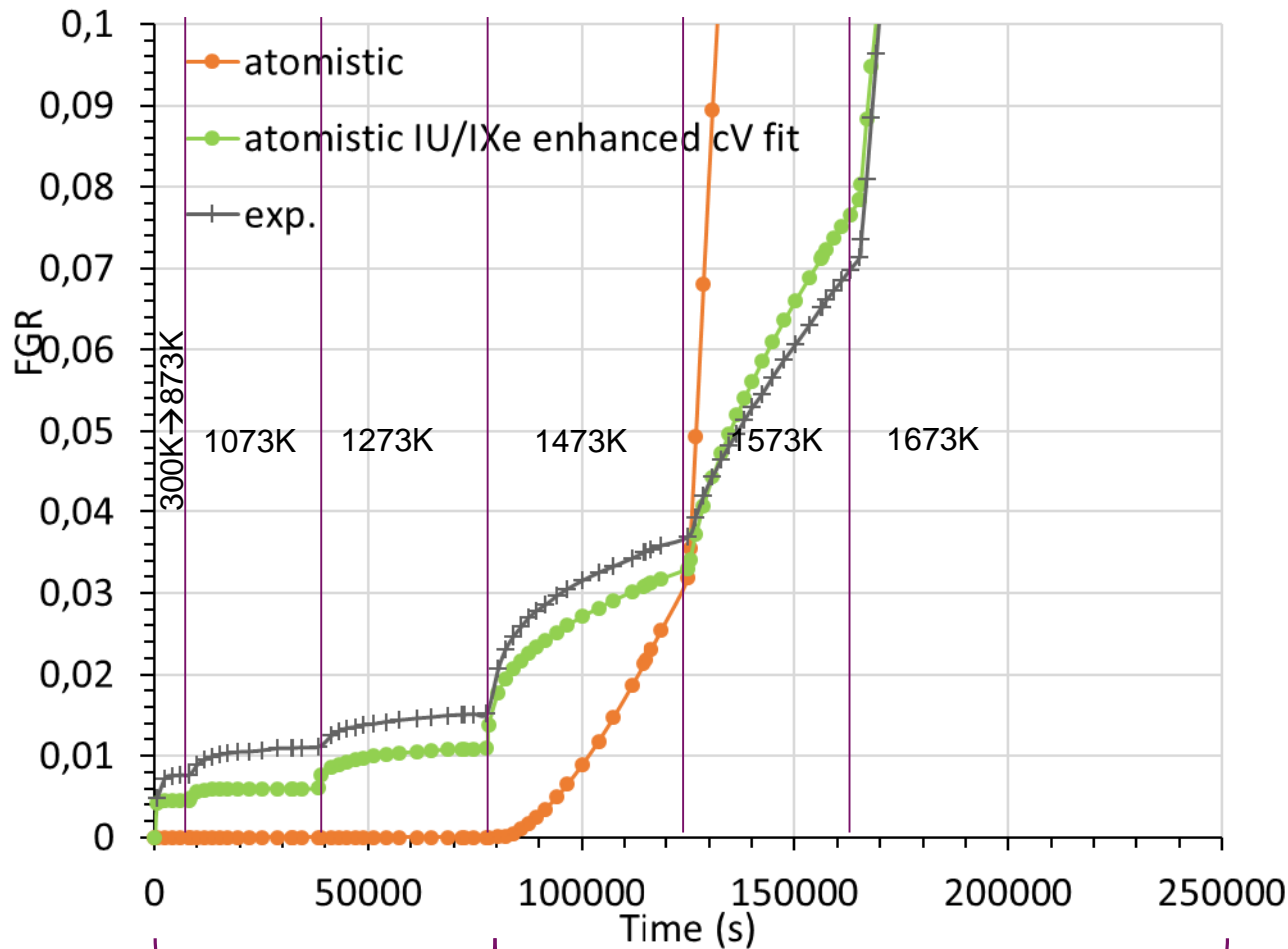
Cluster dynamics to interpret TDS exp. with Xe or Kr implanted virgin UO_2



800 keV Xe implanted
 UO_2
 $1,4 \cdot 10^{11}$ ions/cm²
isochronous annealing

Migration of Point defects and Fission Gases in UO_2

Cluster dynamics to interpret TDS exp. with Xe or Kr implanted virgin UO_2



800 keV Xe implanted
 UO_2
 $1,4 \cdot 10^{11}$ ions/cm²
isochronous annealing

Release of I_{Xe}
kick-out mechanism
 $I_{\text{U}} + \text{XeV}_{\text{U}} \rightarrow I_{\text{Xe}}$

Release of both I_{Xe} and $\text{XeV}_{2\text{U}}$

Extending the approach to the study of intergranular behavior and mixed oxides fuels

- ✓ Combining multiscale modeling and adapted experimental characterization techniques we are able to identify mechanisms or parameters introduced in industrial models
- ✓ Up to now, we focused our investigation to intragranular mechanisms in UO_2

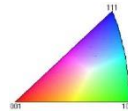
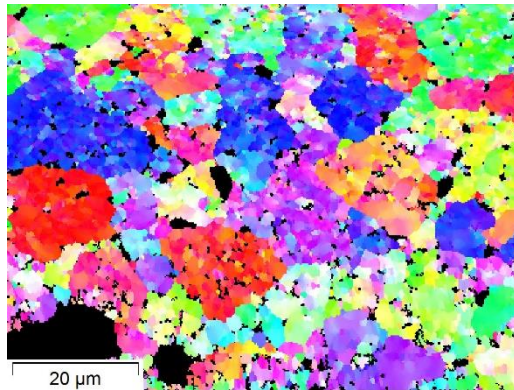
But:

- Experimental evidence that grain boundaries play a key role in fission gas release and microstructure evolution under irradiation (HBS, sub-domains formation inside initial grains,...)
- Fuels for GenIV reactors will be mixed oxides : $(\text{U,Pu})\text{O}_2$ even $(\text{U,Pu,Am})\text{O}_2$

M. Talla Noutack
NuMat P1.100



Methodology developed for UO_2 is being applied to these systems



SEM EBSD
burn-up 73 GWd/tU

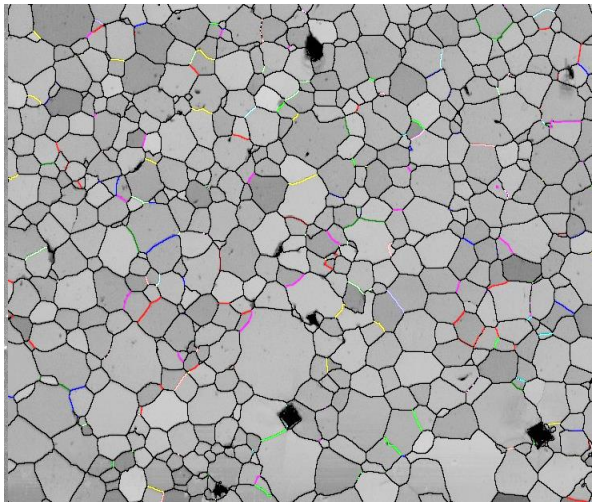
A combined atomistic modeling and
experiment to study UO_2 grain
boundaries properties

A combined atomistic modeling and experiment to study UO_2 grain boundaries properties

Identification of grain boundaries

- Misorientation angle between grains
- Coincidence site lattice (CSL) index

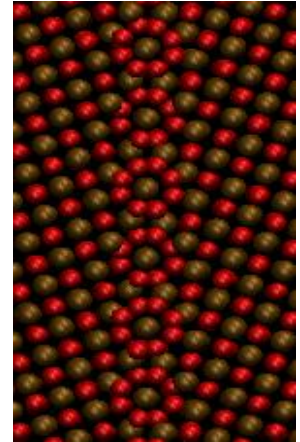
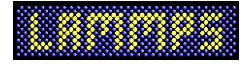
SEM-EBSD map of polycrystalline UO_2 and identification of symmetric GB



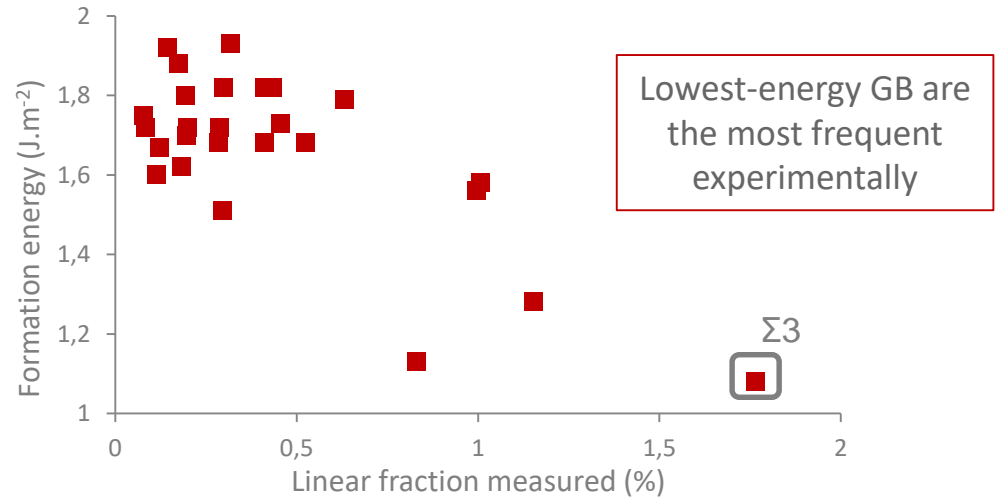
20% of GB are symmetric

Atomic scale investigation

- Large systems: Study using empirical interatomic potentials (validation: complementary electronic structure calculations)
- Construction of GB from 2 grains with given misorientation and relaxation to obtain GB structure and energy



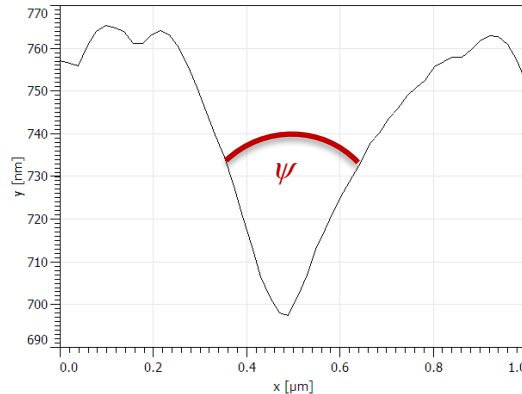
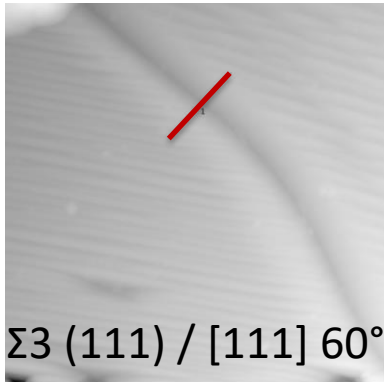
Correlation energies calculated / fractions measured



A combined atomistic modeling and experiment to study UO_2 grain boundaries properties

Experimental characterization

- Formation of a GB groove through thermal etching 4 hours at 1400°C under Ar-5\%H_2
- Determination of ψ angle using AFM (Institut Néel, Grenoble)



ψ angle measured : 92.9°

For a GB \perp
to the surface,

GB Formation energy

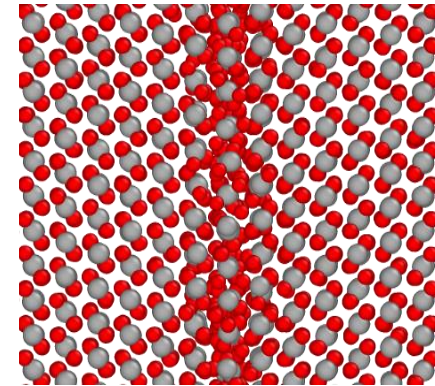
$$\gamma_{\text{gb}} = 2\gamma_s \cos\left(\frac{\psi}{2}\right)$$

Cleavage energy

(energy needed to open the GB)

$$\gamma_c = \gamma_{\text{gb}} - 2\gamma_s$$

Atomic scale modelling



LAMMPS

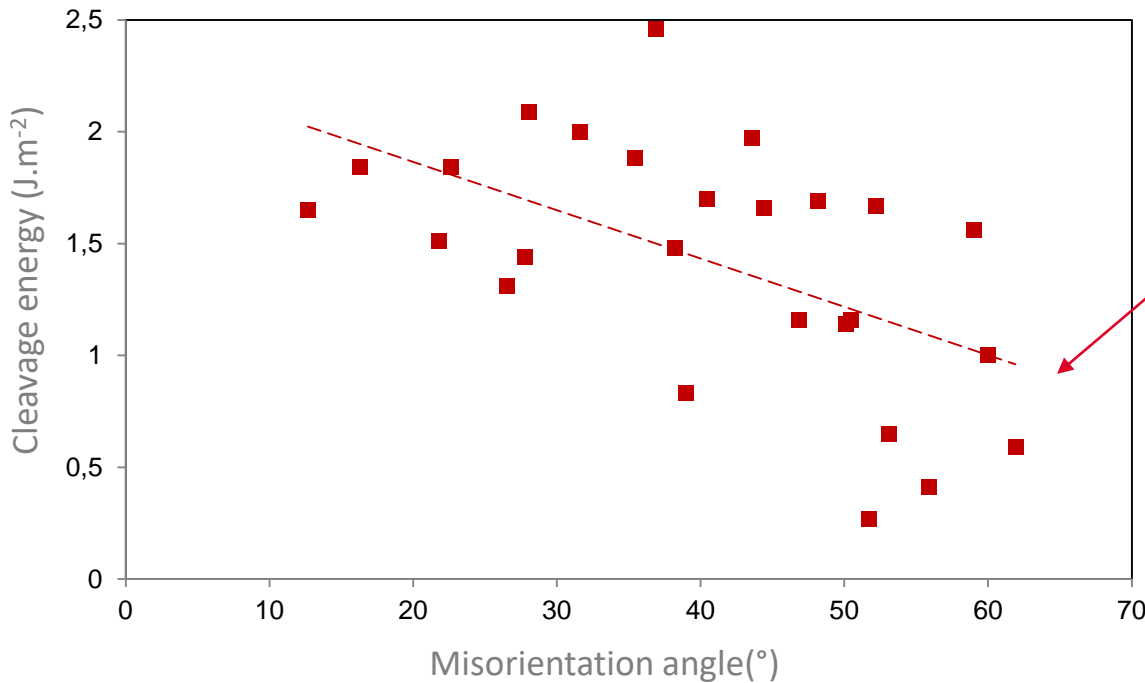
At 1400°C

Formation energy: $\gamma_{\text{gb}} = 1.38 \text{ J.m}^{-2}$

Surface energy (GB plane): $\gamma_s = 1 \text{ J.m}^{-2}$

ψ angle calculated : 92.7°

A combined atomistic modeling and experiment to study UO_2 grain boundaries properties

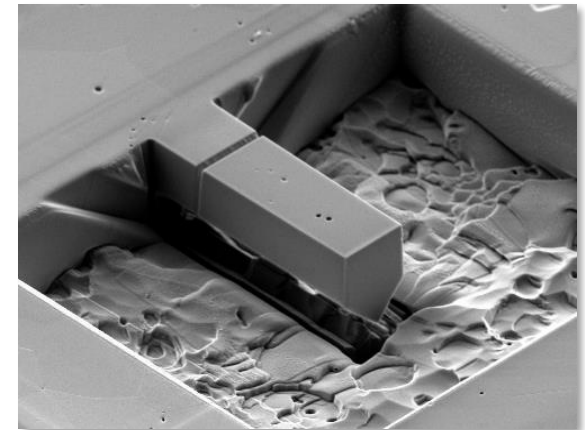


A correlation is observed between cleavage energy and misorientation angle

R. Henry
NuMat 07.18

Perspectives

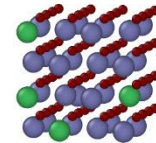
- Validate correlation on more experimental tests
- Extend simulations to general GB and determine impact of temperature
- Establish a link between cleavage energy and toughness through micro-mechanical testing (nano-indenter)
- Defects/FG segregation and diffusion at GB and sub-domains interfaces



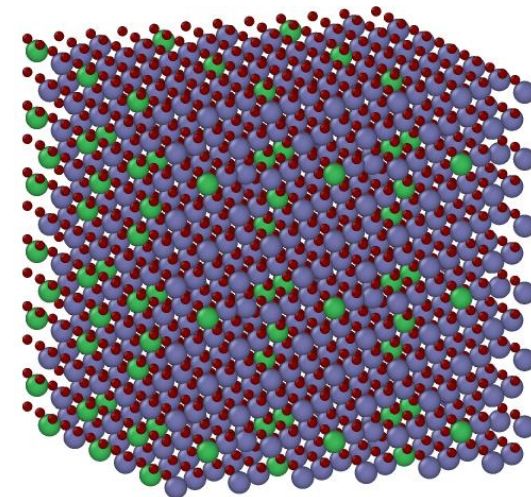
Atomistic simulations to produce
MOX thermodynamic data integrated
in Fuel Performance codes

Data needed for fuel performance codes and thermodynamic databases to evaluate margin to melting

- Lattice parameters, enthalpies, heat capacities, thermal expansion vs temperature, stoichiometry, Pu content for $(U,Pu)O_2$
- Complementary to measurements in hot labs that are scarce due to the complexity of such experiments
- **Difficulty** : solid solutions with wide range of compositions numerous local configurations must be considered
- Use of electronic structure and empirical potential calculations (molecular dynamics and Monte Carlo) to combine accuracy and large quantity of calculations

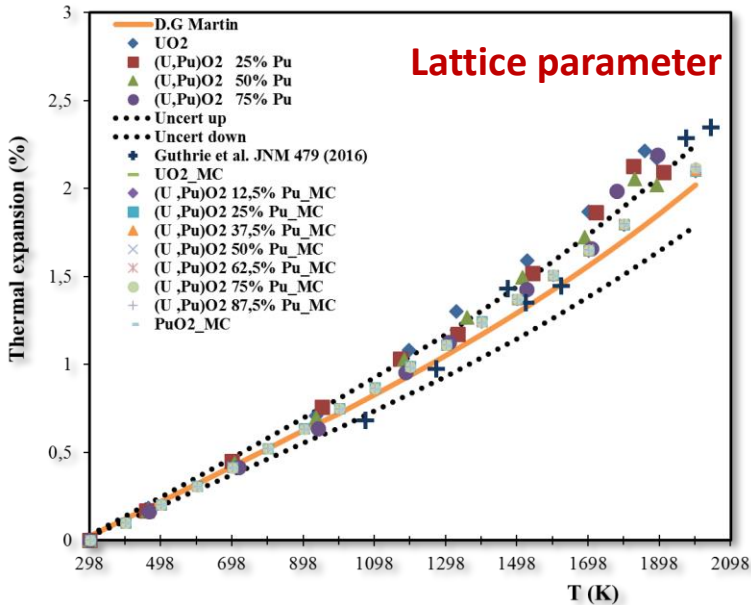


96-atom SQS supercell for electronic structure calculations



2592-atom supercell for empirical potentials

Code GIBBS (CNRS, IFP-EN, Paris Sud)



Similar trends as recommendations drawn from measurements for UO₂ and PuO₂

D.G. Martin *et al.*, J. Nucl. Mater. 152, 94 (1988)

J. Fink, Int. J. Thermophys. 165, 3 (1982)

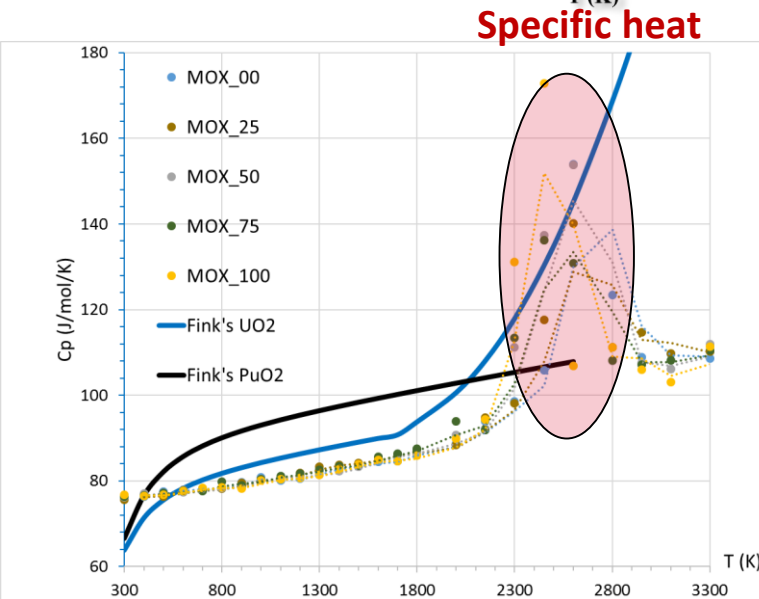
Empirical pot. predicts small Pu content effect

No clear effect from ab initio either (may be at high T)

I. Cheik-Njifon *et al.*, Inorg. Chem. 2018, 57, 10974

I. Cheik-Njifon, PhD thesis (2016-2018)

C. Takoukam-Takoundjou, PhD thesis (2017-2020)



At 2300K (~0.8 Tm) Bredig transition: Fusion of O sublattice

⇒ Change in properties

Should be taken into account in laws used in FPC

- Investigation of operational issues using basic research approach: irradiation damage, margin to fuel melting, microstructure \Rightarrow essential data for fuel performance codes
- Combination of modelling and experiments contributes to get further insight into mechanisms governing the behaviour of nuclear fuels and cladding under irradiation

Future work

- Extend application of models at mesoscopic scale, in particular rate theory methods to study atomic transport and dislocation dynamics for microstructure
- Application of basic research approach on other nuclear fuels: including additives, chemically active fission products, minor actinides

Contributors:

CEA

- DEN, DEC: R. Belin, M. Bertolus, E. Bourasseau, G. Carlot, M. Freyss, S. Maillard, C. Onofri, C. Sabathier
- DEN, DMN: T. Jourdan
- PhDs and post-docs: B. Dorado, I. Cheik Njifon, M. Gérardin, A. Germain, A. Ksibi, C. Takoukam Takoundjou, M.S. Talla Noutack, E. Vathonne, J. Wiktor

- DAM: M. Torrent, G. Geneste, B. Amadon, B. Dorado

CNRS

- CEMHTI : M-F. Barthe
- CENBG : E. Gilabert

DOE

- LANL : D. Andersson, M. D. Cooper, C. Stanek, R. Perriot

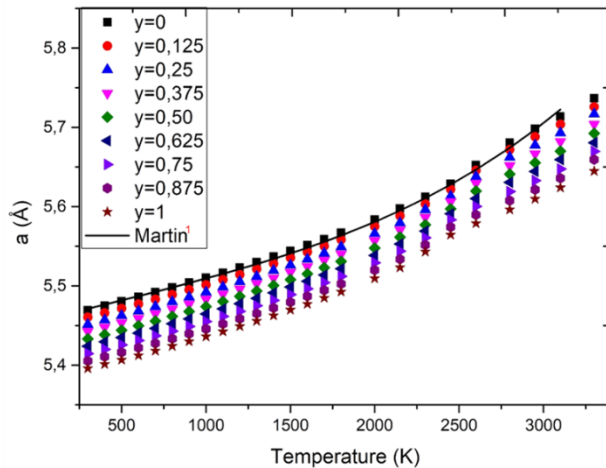
THANK YOU FOR YOUR ATTENTION



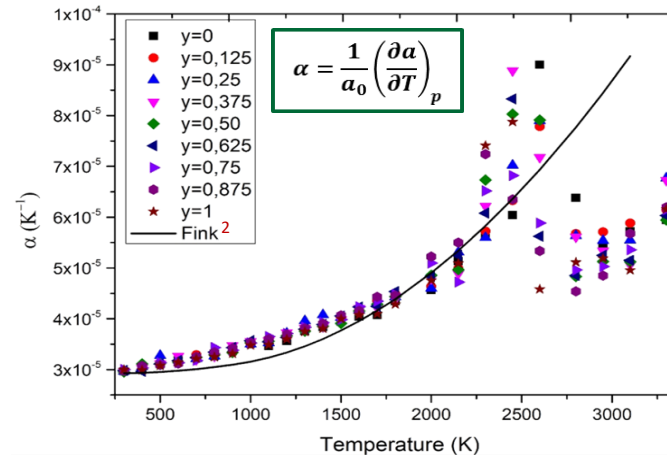
Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Cadarache | 13108 Saint-Paule-lez-Durance Cedex
T. +33 (0)4 42 25 30 34

DEN
DEC
SESC

Lattice parameter

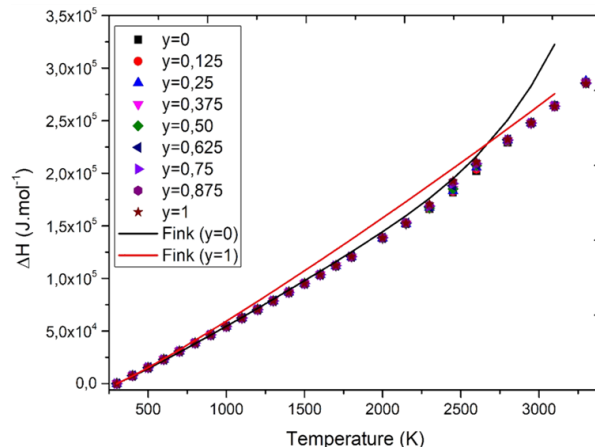


Linear thermal expansion coefficient

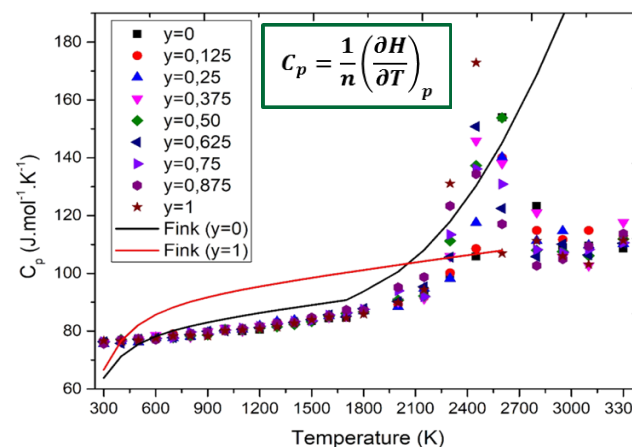


Similar trends as recommendations drawn from measurements for UO_2 and PuO_2
Predictions for variable Pu content

Enthalpy increment



Specific heat



At 2300K (~ 0.8 Tm)
Bredig transition: Fusion of oxygen sublattice
 \Rightarrow Change in properties

Should be taken into account in laws used in FPC

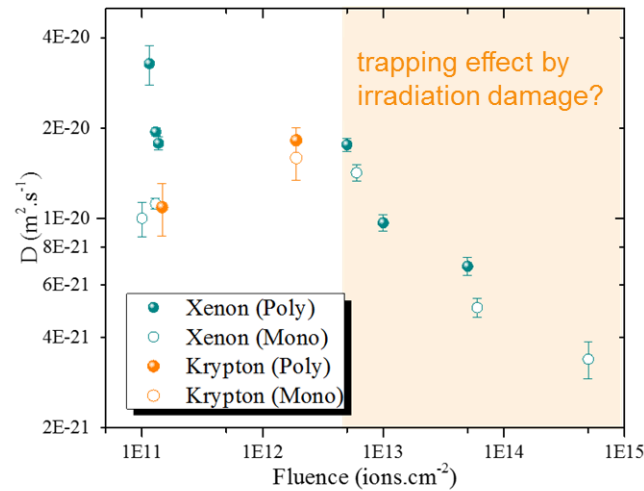
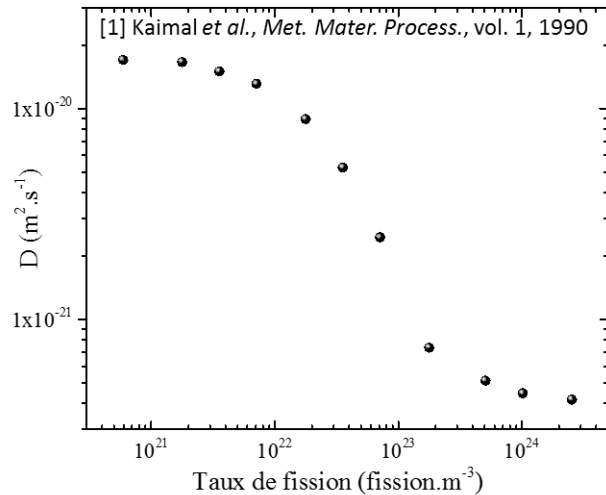
1 D.G. Martin *et al.*, J. Nucl. Mater. 152, 94 (1988)

2 J. Fink, Int. J. Thermophys. 165, 3 (1982)

Point defects and Fission Gas in UO_2 formation/incorporation/migration

Cluster dynamics to interpret TDS experiments

✓ In experiments, decrease of the apparent diffusion coefficient upon fluence of irradiation



✓ Fit of the release isotherms by a simple model based on Fick's 2nd law is coherent with this picture

$$\frac{\partial C(x, t)}{\partial t} = D \frac{\partial^2 C(x, t)}{\partial^2 x} - k_1 \times C(x, t)$$

$$k_1 : \text{trapping coeff} \sim 4\pi r_p C_p D_{2Xe}$$

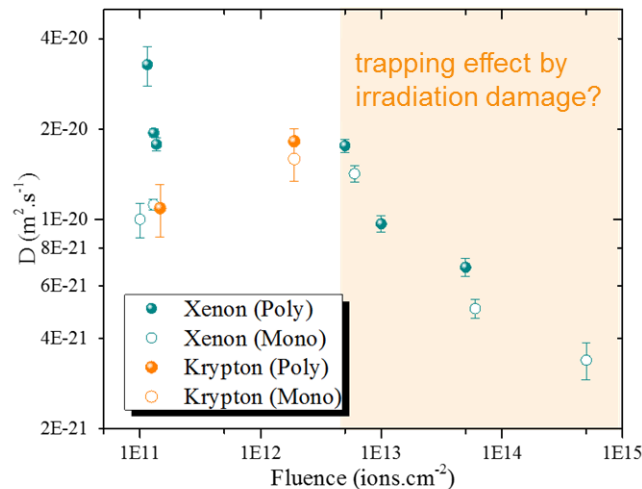
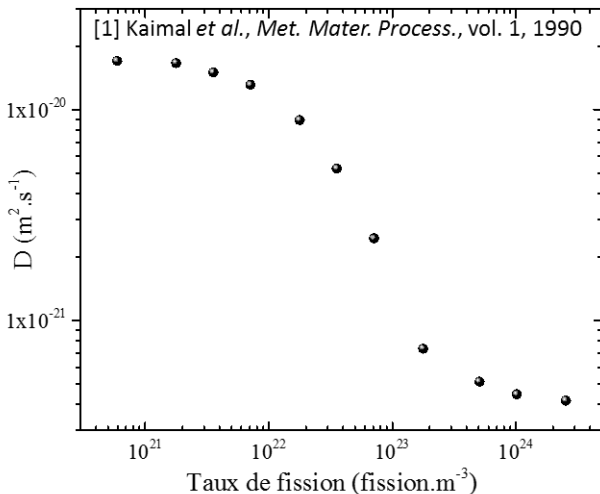
burst effect

$$D(t) = [D_1 - D_{2,intrinsic}] e^{-\frac{t}{\tau}} + D_{2,intrinsic} \longrightarrow \text{intrinsic diffusion coefficient}$$

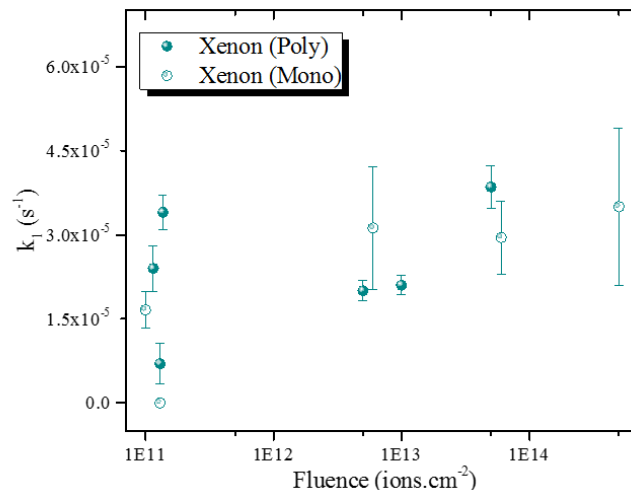
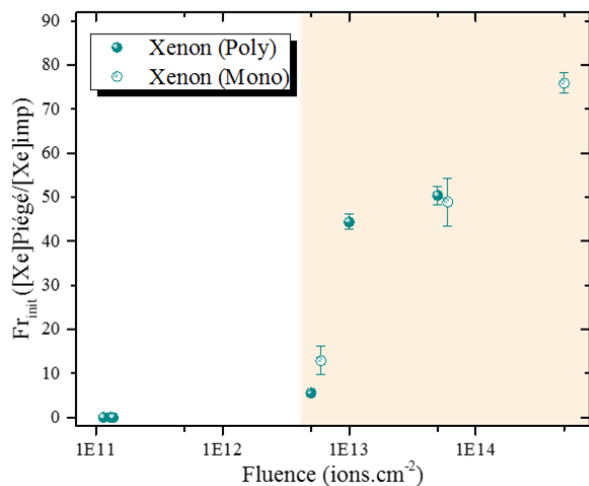
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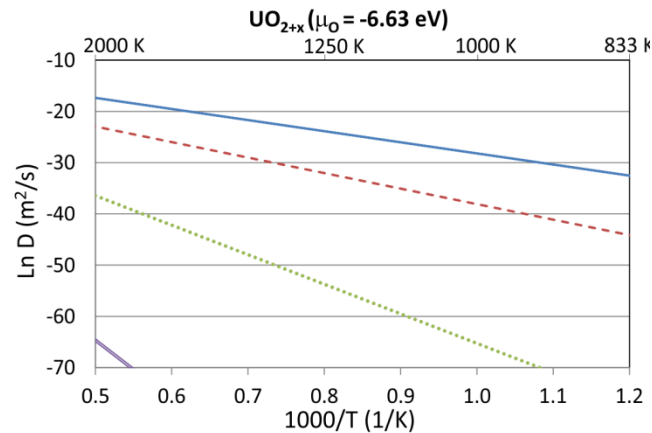
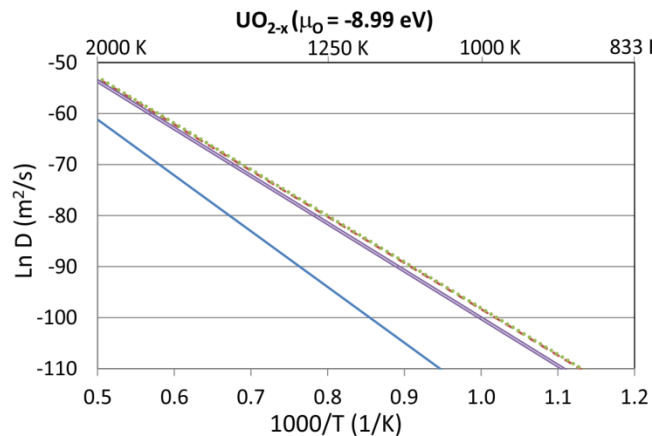
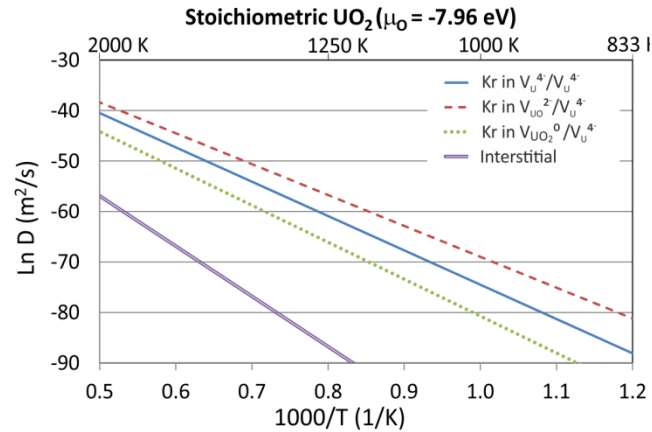
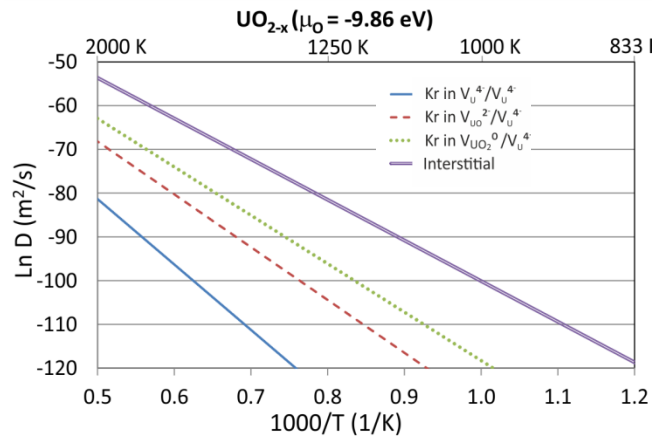


$D_{intrinsic} = 1,9 \cdot 10^{-20} \text{ m}^2 \cdot \text{s}^{-1}$

$k_1 \sim \text{cst}$ during annealing

to check this assumption and determine the mechanisms : CD

Point defects and Fission Gas in UO_2 formation/incorporation/migration



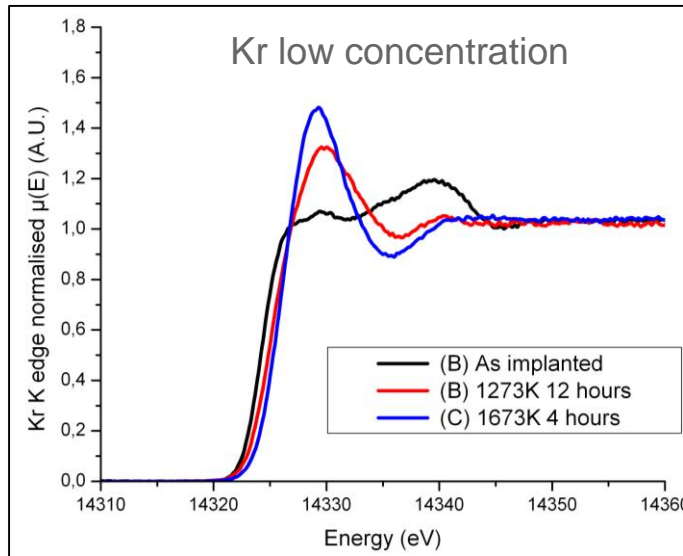
□ Mechanisms and trends with non-stoichiometry similar to previous study of Xe [Andersson et al]

- UO_{2-x} : migration assisted by Schottky and charged V_U $D = 5,35 \times 10^{-8} \times e^{-E_a/kT} \text{ m}^2/\text{s}$ $7,80 < E_a < 8,01 \text{ eV}$
- UO_2 : migration assisted by V_{UO_2} and charged V_U $D = 7,08 \times 10^{-9} \times e^{-E_a/kT} \text{ m}^2/\text{s}$ $4,09 < E_a < 7,80 \text{ eV}$
- UO_{2+x} : migration assisted by 2 charged V_U $D = 3,07 \times 10^{-7} \times e^{-E_a/kT} \text{ m}^2/\text{s}$ $0,73 < E_a < 4,09 \text{ eV}$

□ Combinaison with experiments in progress

Point defects and Fission Gas in UO_2 formation/incorporation/migration

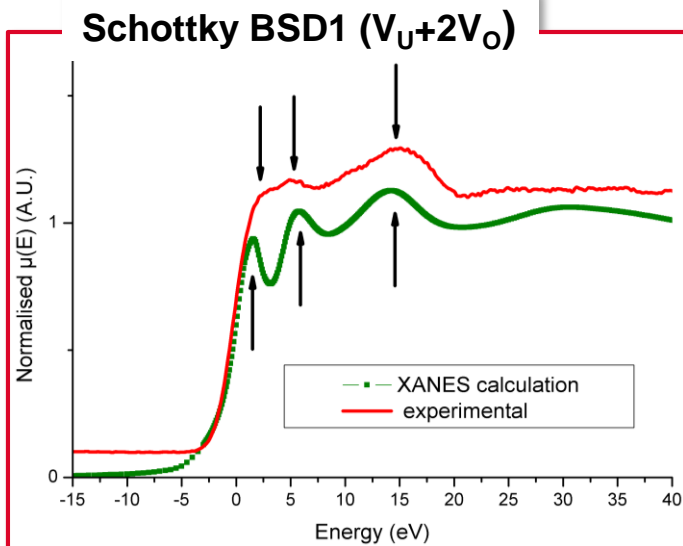
Kr incorporation site from XANES experiments



How to analyse XANES spectra?

To determine the **local environment** of the probed atom, the XANES spectra are modelled using the FDMNES code [Bunău *et al.*, J. Phys.. Cond. Mat. 21 (2009) 345501]

The **atomic coordinates of the atoms surrounding the rare gas atoms** are required as input for the fit of the spectra



Only BSD1 gives a good agreement

DFT+*U* local environment close to XANES results

For as implanted 0.5 at.% Kr samples

- Kr atoms surrounded by U and O atoms only (**no Kr aggregate**)
- Kr atoms as single atoms inside neutral **bound Schottky defects** with O vacancies aligned along the (100) direction (BSD1)

→ **Nano-cavities observed by TEM after rare gas implantation are empty**

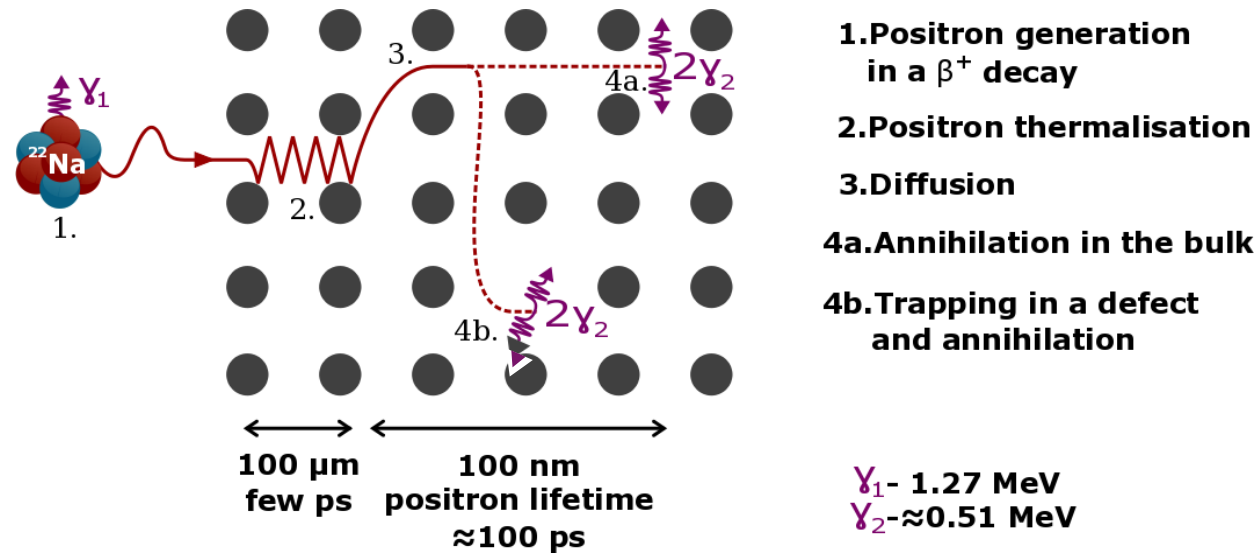
Point defects and Fission Gas in UO_2 formation/incorporation/migration

Positron lifetimes calculation and comparison to PALS exp.

Non-destructive experimental method used to study **vacancies in solids**

Evolution of defects with temperature (ionisation, clustering, annealing)

Neutral and **negative defects** can be detected and distinguished, while **positive** ones cannot be detected



$$\frac{1}{\tau} = \frac{\pi c r_0^2}{e} \int_{R^3} d^3 \vec{r} \left[n(\vec{r}) n^+(\vec{r}) \Gamma(n, n^+) \right]$$

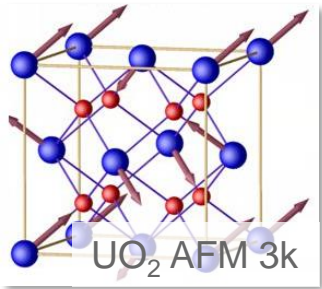
Electronic and positronic densities are needed

➔ **Electronic structure calculations (TC-DFT)**

Atomistic simulation

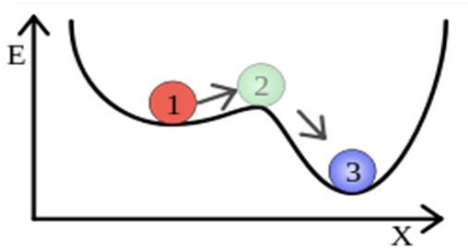
Actinide compounds

- Complex systems:
various phases, non-stoichiometry,
magnetism



Ab initio simulation

- ⇒ DFT+U (VdW-DF+U)
well adapted for FG in actinides fuels
- ⇒ **Occupation matrix control (OMC)** scheme



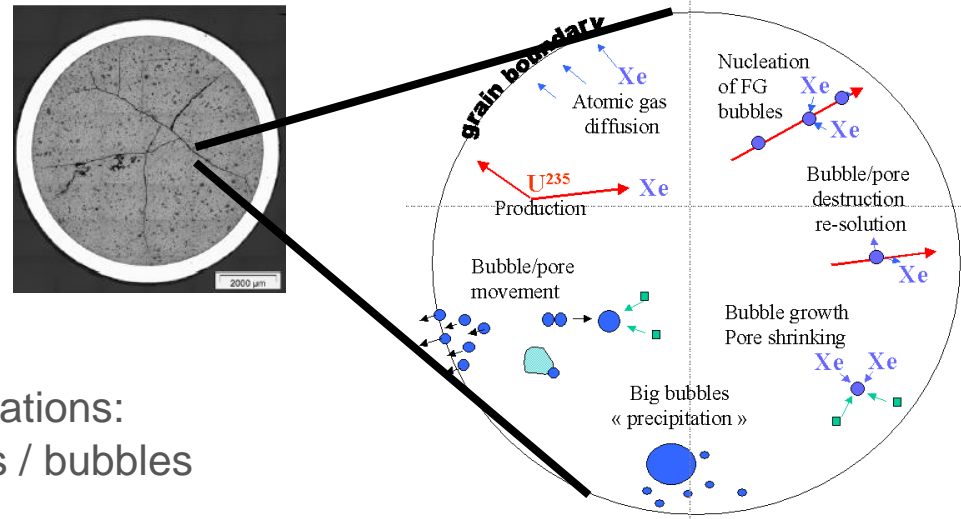
- Actinide cations with varying oxidation state : U^{3+} , U^{4+} , U^{5+} in UO_2
 Pu^{3+} , Pu^{4+} in PuO_2
- Electrons $5f$ localized in numerous compounds
⇒ **Strong electronic correlations**

Empirical potential-based simulation

- ⇒ **Classical molecular dynamics**
- ⇒ **Molecular Monte Carlo**

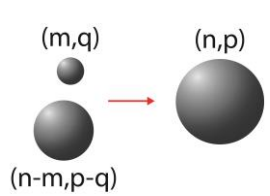
CLUSTER DYNAMICS

Modelling the evolution of the microstructure under in-pile or ion irradiation



Set of differential equations on species concentrations: calculation of defect and defect clusters (cavities / bubbles / interstitial loops) concentrations over time

$$\frac{dC(n, p)}{dt} = \sum_{(m, q)_{\text{mobile}}} J_{(n-m, p-q) + (m, q) \leftrightarrow (n, p)} - \sum_{(m, q)_{\text{mobile}}} J_{(n, p) + (m, q) \leftrightarrow (n+m, p+q)} + G(n, p) - K(n, p)$$



Absorption of mobile cluster (m, q) by cluster (n, p)

Emission of mobile cluster (m, q) from cluster (n, p)

Source term

Sink term (for mobile clusters)

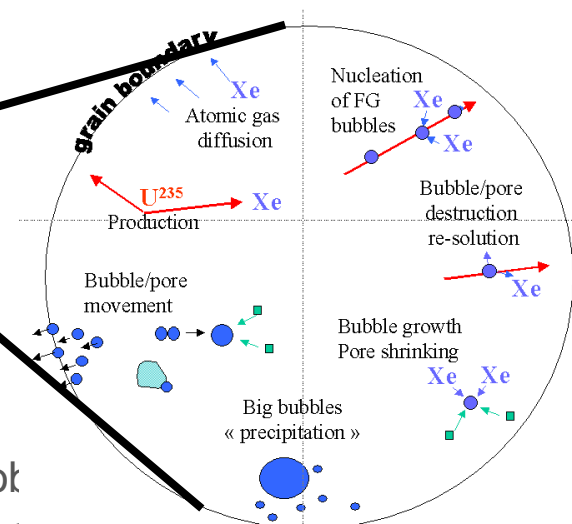
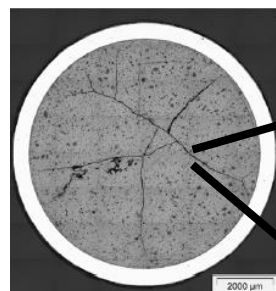
DFT and empirical potential calculations can provide input data needed

Simulation of FG behaviour at the grain scale

- For guaranteeing fuel rod integrity, in-pile FG behavior is of major concern



- Essential since these populations control a large number of properties and the macroscopic evolution of the material



- **MARGARET** code within the **PLEIADES** platform at CEA: Most of simulated (gas concentration, gas release, swelling, porosity) but some phenomena are modelled **empirically**, e.g. **bubble sink strength** and **bubble size distribution** and **irradiation defects not taken into account**

- **Cluster Dynamics (rate theory model)**: Comprehensive framework to calculate defect and defect cluster (cavities / bubbles / dislocation loops) concentrations over time, with time scale and length scale appropriate for fuel study (*years, grains*)

→ **Study using cluster dynamics of intragranular fission gas behaviour**

- **code CRESCENDO (co-developped EDF/CEA)**
- **DFT-based parametrization**
- **Separate effects experimental studies to validate**

Static: Nudged elastic band (*NEB*)

- $T = 0 \text{ K}$
- Discretization and optimization of the migration path
 - Migration barrier E_m
 - Migration mechanism

Activation energy for diffusion :

$E_a \sim E_f + E_m$ via point defect model

$D = D_0 \exp(E_a / kT)$

Prefactor D_0 from vibration modes (phonons), in presence of defects governing the diffusion. Calculations using **empirical potentials** more adapted

