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Educational, Scientific and
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International Atomic Energy Agency

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Joint ICTP-IAEA Advanced Workshop on Multi-Scale Modelling for Characterization and Basic Understanding of Radiation Damage Mechanisms in Materials

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Nuclear fuel behavior under irradiation: introduction to multi-scale modeling and experimental characterization

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Modeling and experiments for improving nuclear fuel performance : *Numerical and experimental simulation at the atomic scale*

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Nuclear Energy Division
Fuel Study Department

CADARACHE

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Outline

Modeling and experiments for improving nuclear fuel performance : numerical and experimental simulation at the atomic scale

Part 1

Nuclear fuel behavior under irradiation: introduction to multi-scale modeling and experimental characterization

Part 2

Numerical simulation techniques for nuclear fuels at the atomistic scale: electronic structure calculations and empirical potentials

Part 3

Numerical simulation of transport properties in nuclear fuels : from the atomistic scale to the mesoscopic scale

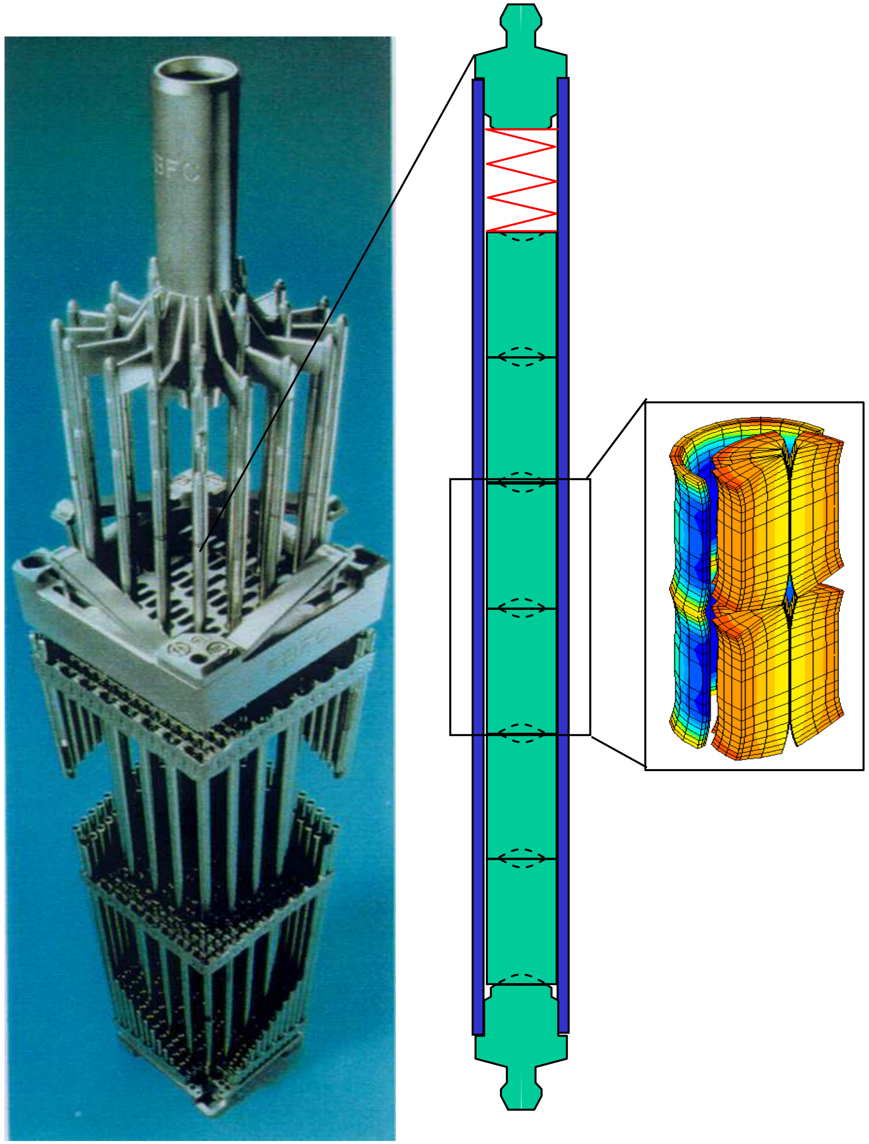
Part 4

Experimental simulation of nuclear fuels: separate effect studies

Part 1

Nuclear fuel behavior under irradiation: introduction to multi-scale modeling and experimental characterization

Nuclear fuels



Materials of interest:

UO_2, $(U,Pu)O_2$

oxide fuels

standard nuclear fuels in PWR

(U,Pu)C, (U,Pu)N

carbide, nitride fuels

$(U,Pu,MA)O_2$

Minor Actinide (MA) containing fuels

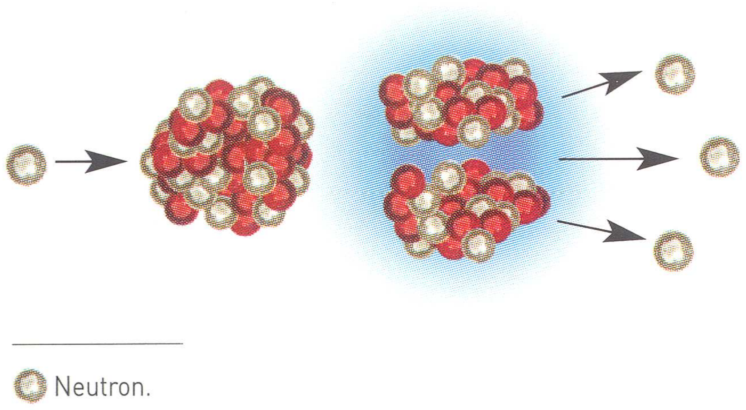
transmutation targets

UAl, PuAl, UZr, UPuZr, U_3Si_2, UMo

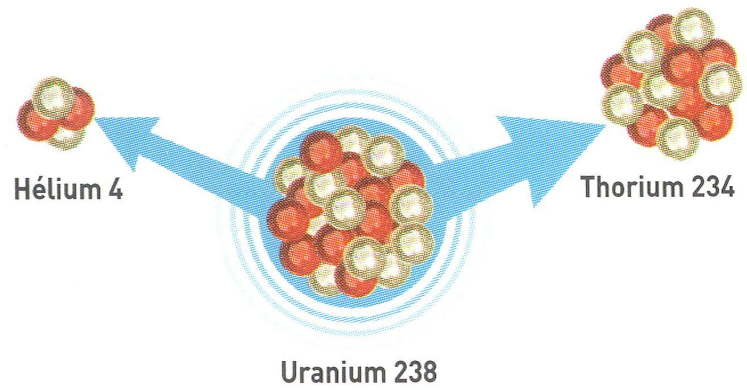
metallic fuels

Irradiation damage in nuclear fuels

fission



alpha decay



Fission products **Helium**

Volatile elements (Kr, Xe, I…)

Dilution ?
Bubble precipitation ?
Swelling ?

Recoil nuclei

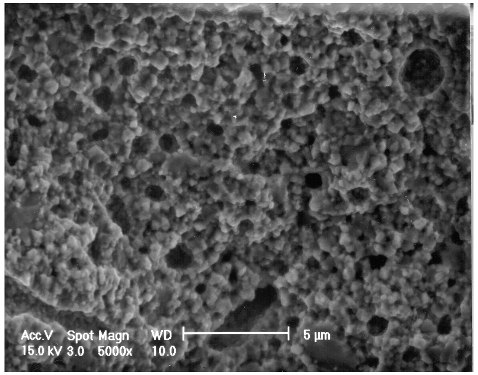
→ Collision cascades

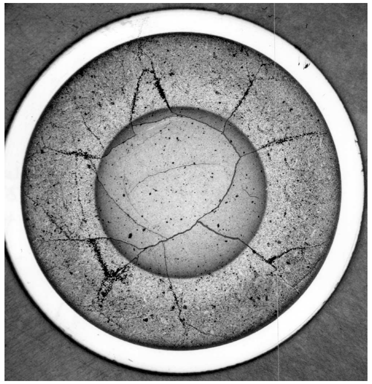
Point defects

Stability ?
Structural changes ?

Fuel under irradiation: a complex behavior

- **Fuel under irradiation :**
  - a very complex behavior, a changing material
  - few transformations as illustration !





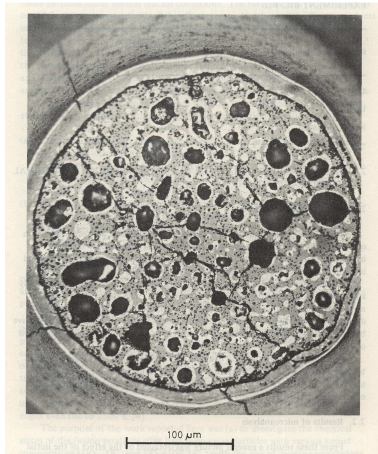
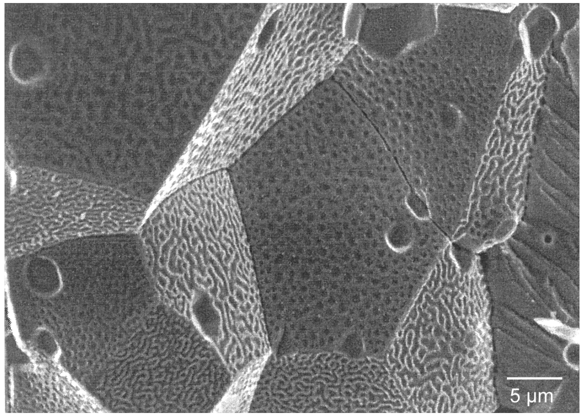
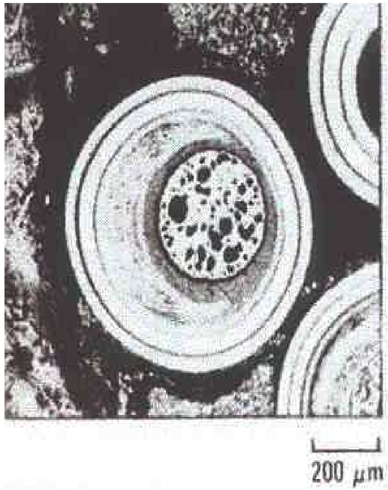
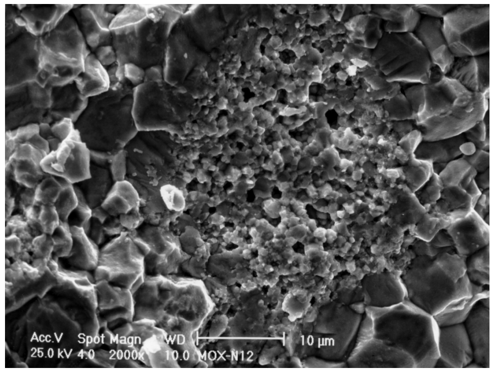


FIG.1. Micrograph of an irradiated kernel with an initial composition of 90% UO_2–10% UC_2 after a burn-up of 69% FIMA.



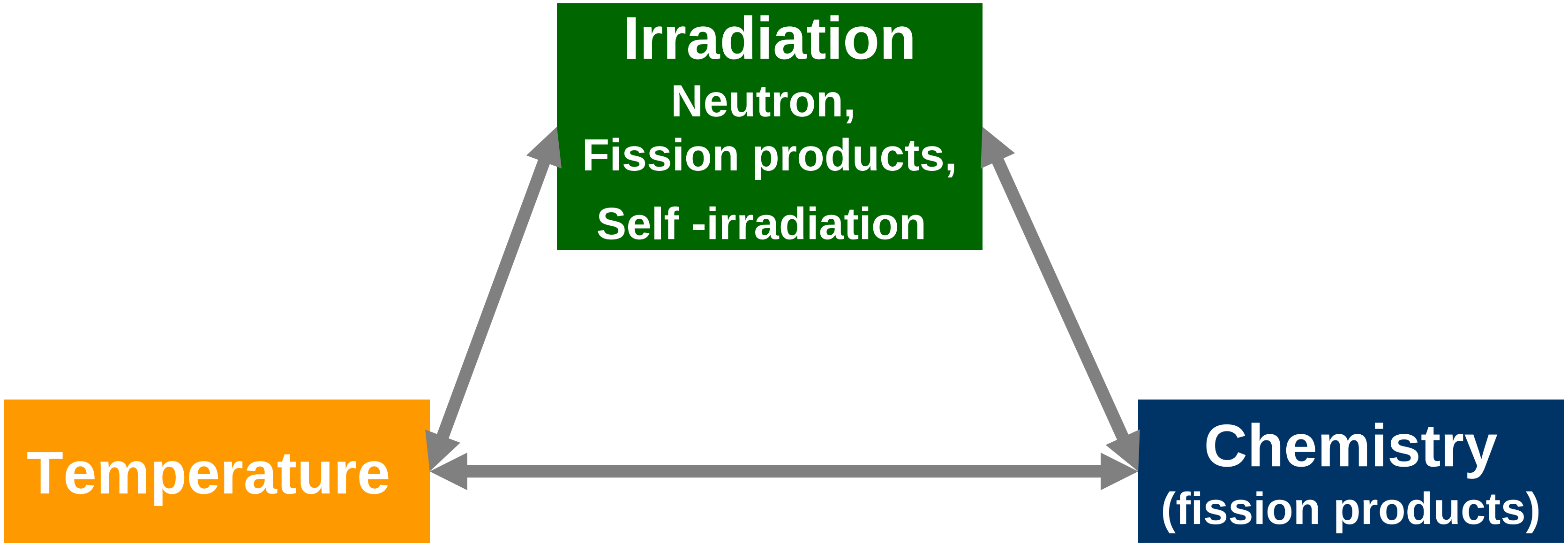




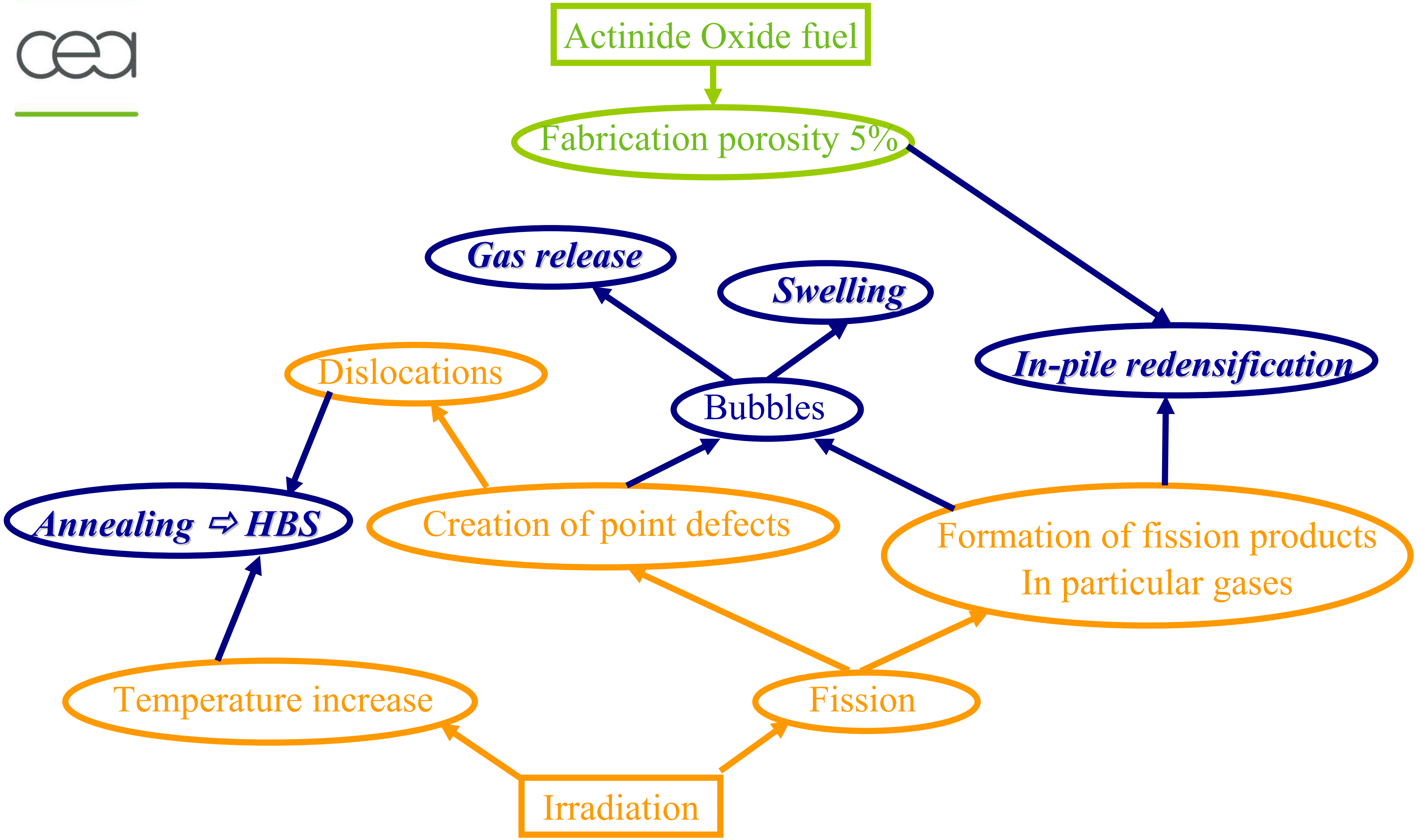
Fuel under irradiation: a complex behavior

Complex **phenomena** involved

Strong coupling between various effects

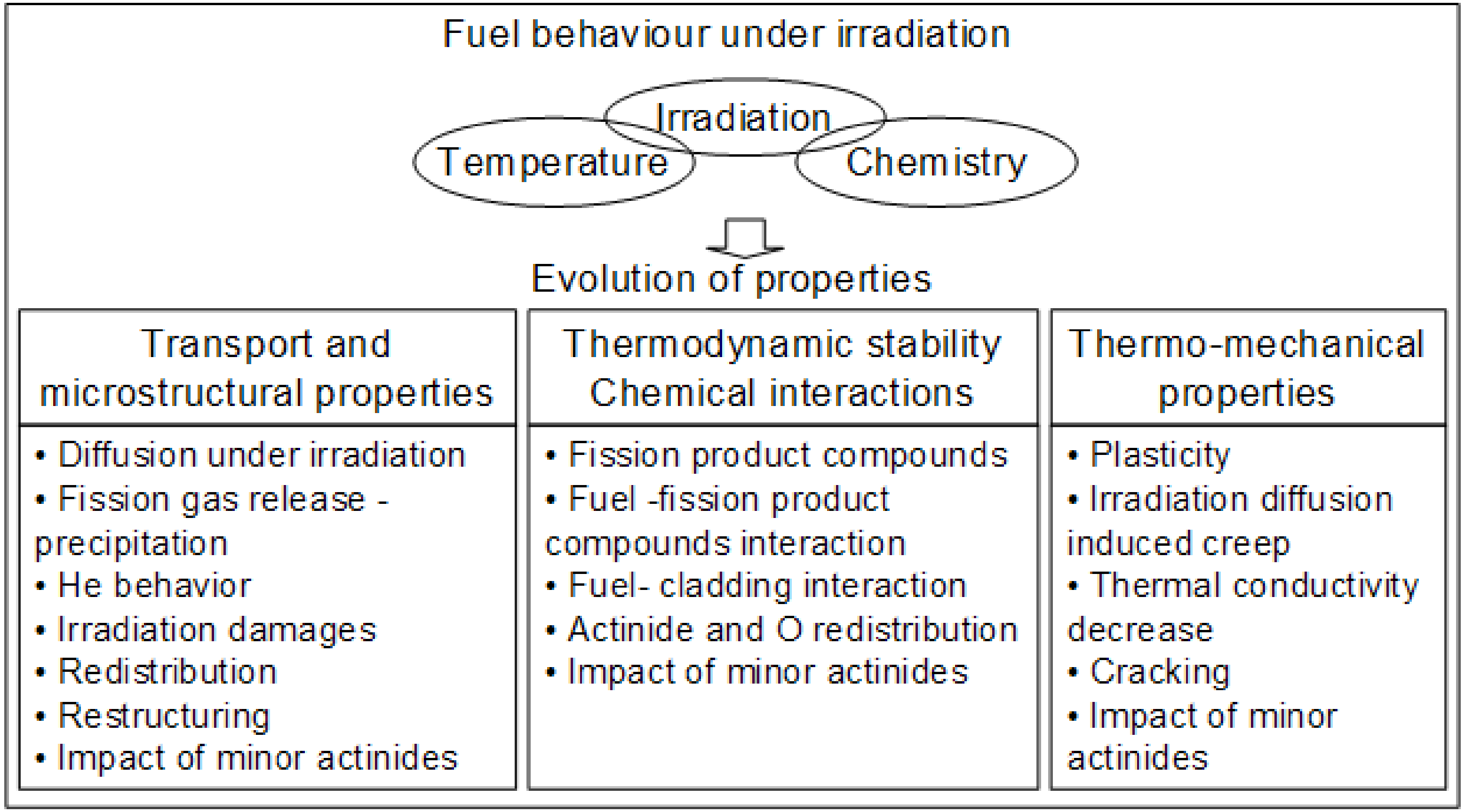


Fuel under irradiation: a complex behavior



Fuel under irradiation: a complex behavior

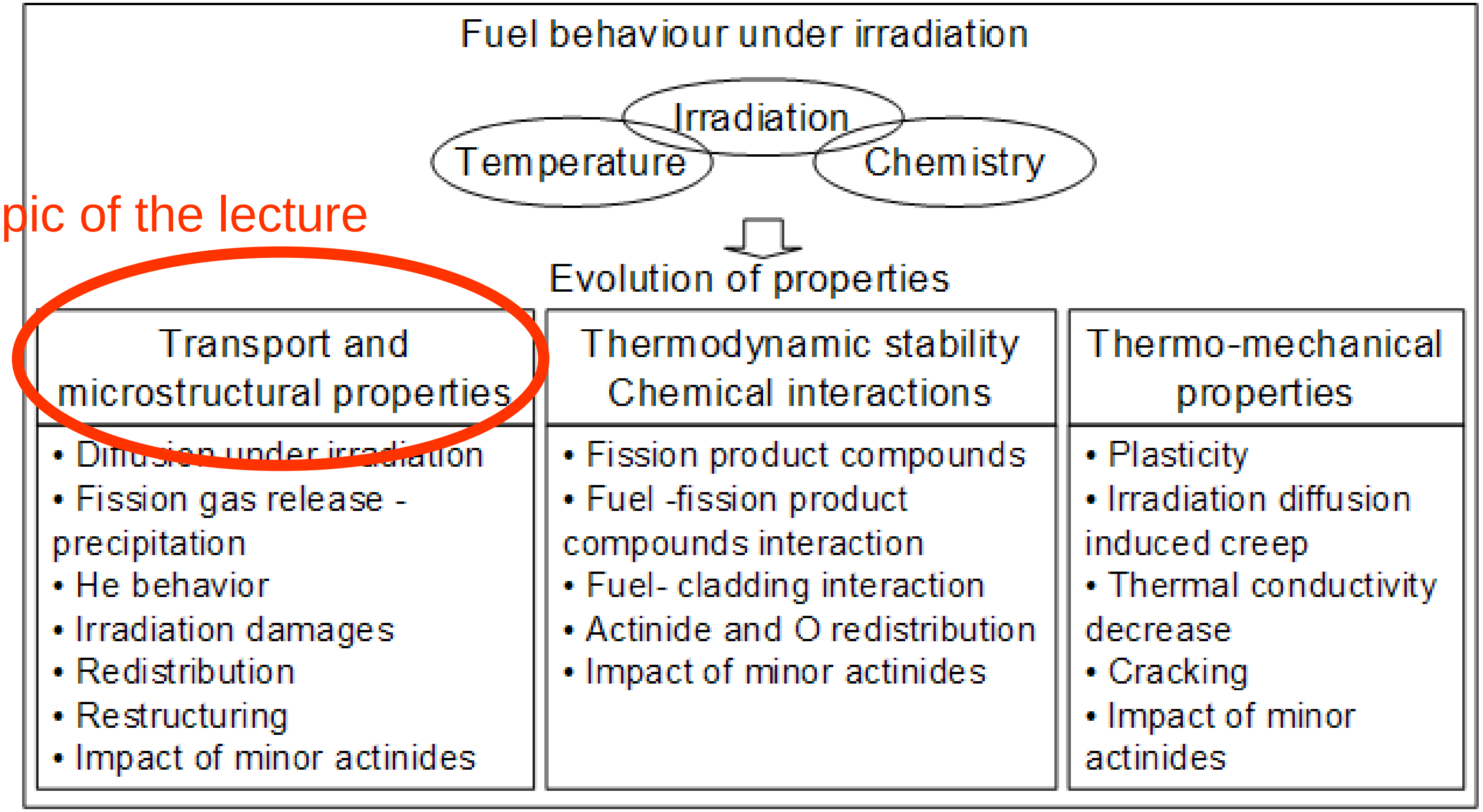
Complex **phenomena** involved



Fuel under irradiation: a complex behavior

Complex **phenomena** involved

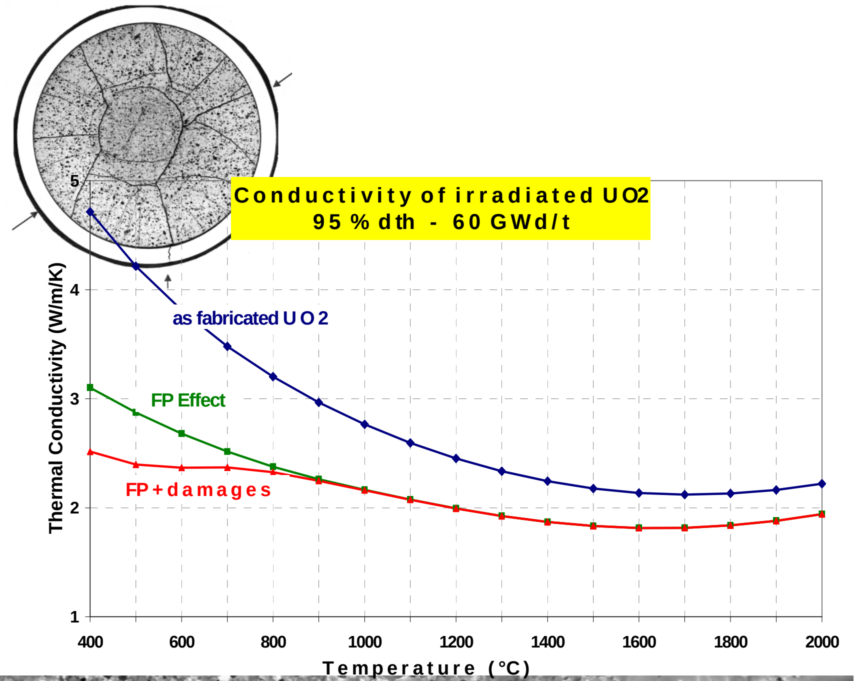
Main topic of the lecture

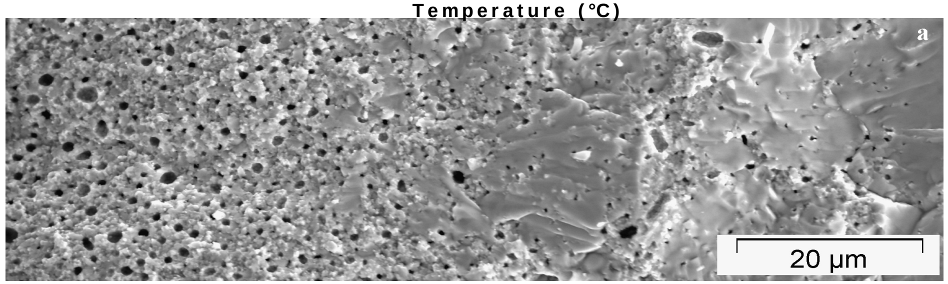


Fuel under irradiation: a complex behavior

• Physical and chemical transformations

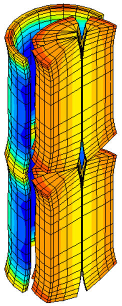
- Mechanical changes (cracking)
- Thermal changes (thermal conductivity)
- Structural changes (phase stability)
- Micro-structural changes (defects, HBS structure)
- Species migration
- Formation of fission product compounds…





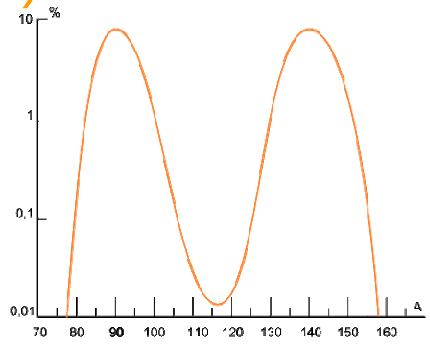
SEM Fractograph at periphery of a UO_2 pellet (73 GWd/t) J. Noirot 07

Some basic limiting phenomena in LWRs fuels



- **Pellet Cladding Interaction**
  - Clad strains induced by pellet expansion
  - during power ramps
- **Behaviour of Fission Products and Fission Gas**

fissions ➔ 1 third of FP are rare gas atoms (Xe + Kr)



Distribution des produits de fission de l'uranium-235

Axe Y: échelle logarithmique

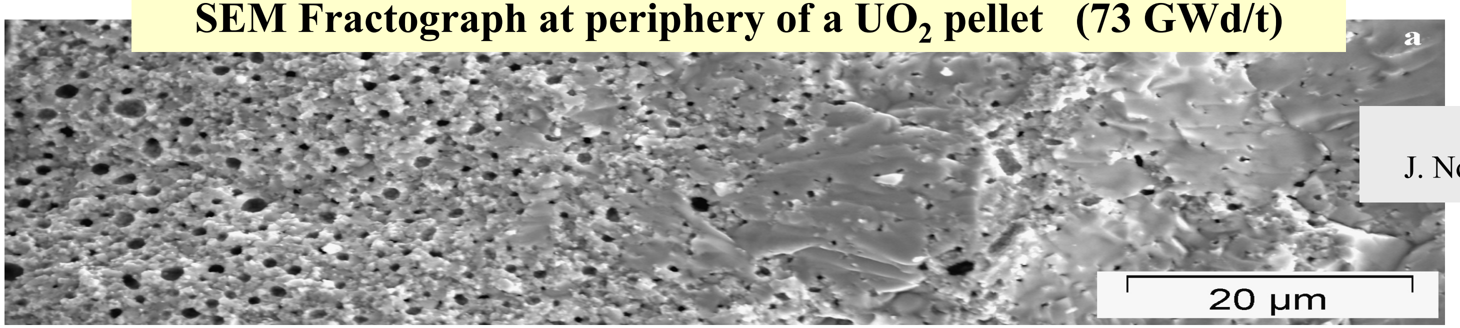
  - Increase of inner pressure due to gas release
- **High Burn-up effects**

Complete transformation of UO_2 microstructure

subdivision of grains 10 µm ➔ 0.2 µm

  - High Burn-up Structure (HBS) or Rim effect

SEM Fractograph at periphery of a UO_2 pellet (73 GWd/t)



J. Noirot 07

Fuel under irradiation: a complex behavior

- Goal : improve our **understanding and our capability to predict** the fuel behavior

  - Need to **de-correlate** the complex **phenomena** involved
  - deeper description of phenomena : **towards the atomistic level**

Coupling between :

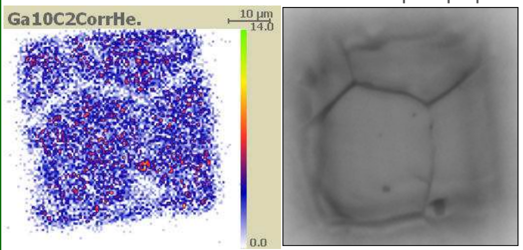
- Post irradiation examinations (PIE) after power plant irradiations or specific MTR irradiations
- Separated effect studies
- Modeling and characterization at the relevant scale.

Multi-scale modeling and characterization to understand the fuel behavior

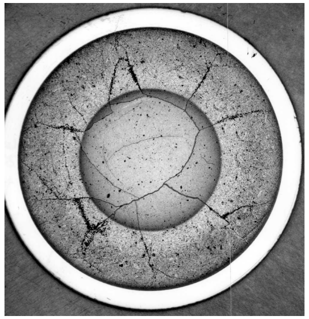
Experimental simulation
Separate effect studies

Cartographie en µ-NRA

Microscopie optique

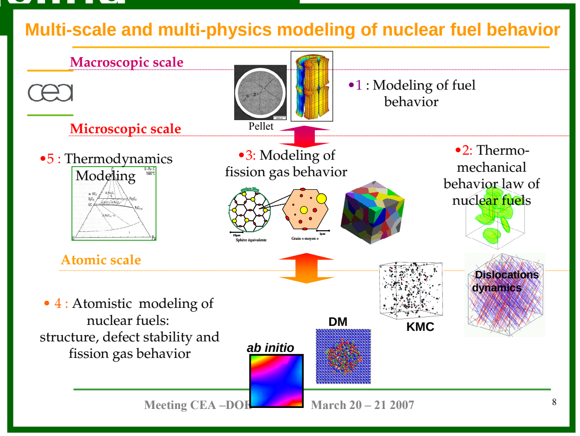


Post irradiation examinations



Multi-scale characterization

Multi-scale modeling

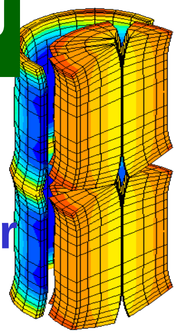


Behavior of current and future fuels

Materials under irradiation

understanding

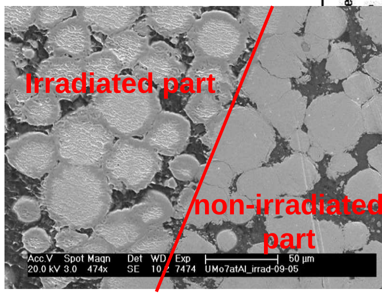
Simulation - validation for the prediction of the in-pile fuel behavior

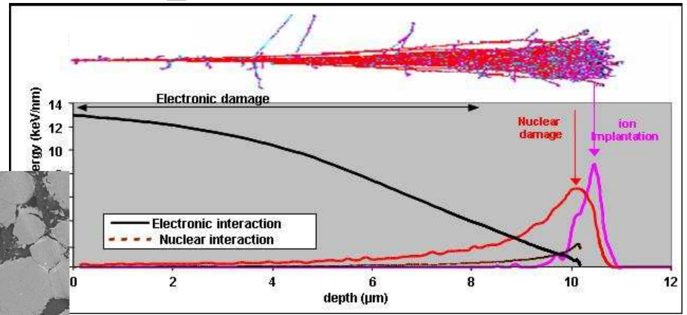


- **Improve performance of current fuels**
- **Design future fuels**

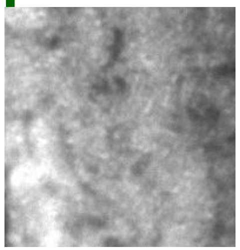
Separated effects studies : the approach

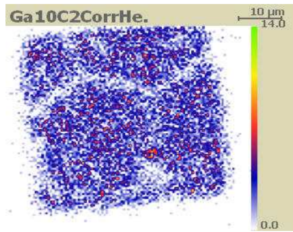
- Non active model materials such as UO_2
- Ion **implantation** to simulate FP
- **Thermal treatment or** heavy ion **irradiation**

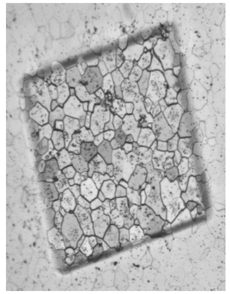




- **Characterization with a large panel of dedicated techniques** (SIMS, RBS, NRA, TEM, XAS)



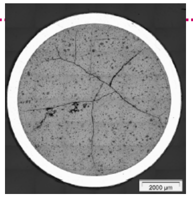




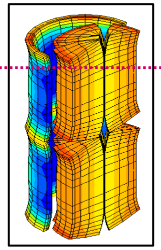
- **Large scientific facilities** (particle accelerators and synchrotron radiation)

Multi-scale modeling of nuclear fuel behavior

Macroscopic scale



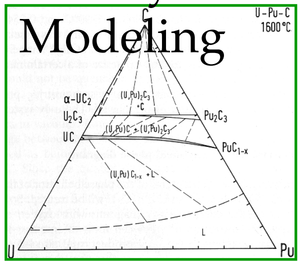
Pellet



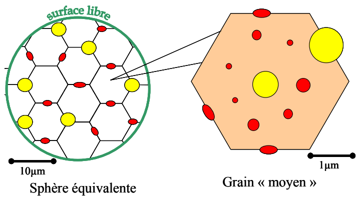
- Modeling of fuel behavior

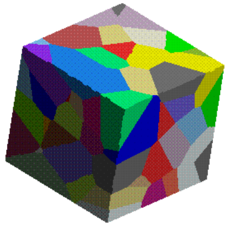
Microscopic scale

- Thermodynamics Modeling

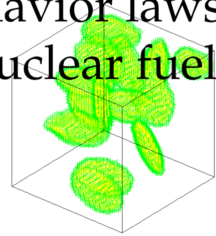


- Modeling of fission gas behavior





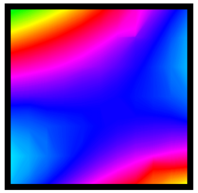
- Thermo-mechanical behavior laws of nuclear fuels



Atomic scale

- Atomistic modeling of nuclear fuels: structure, defect stability and fission gas behavior

ab initio



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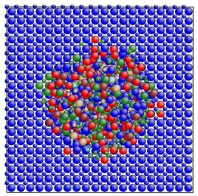

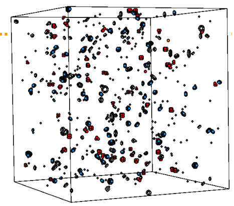

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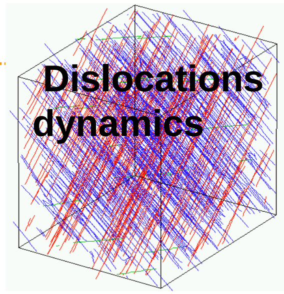
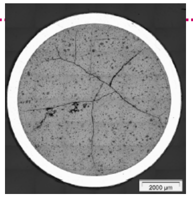


Multi-scale modeling of nuclear fuel behavior

Macroscopic scale



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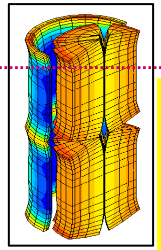
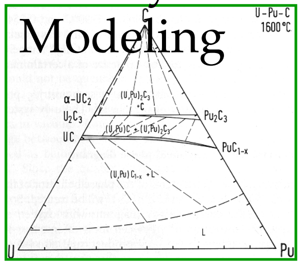


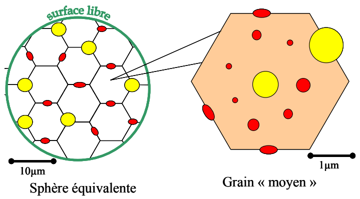
- **Modeling of fuel behavior**

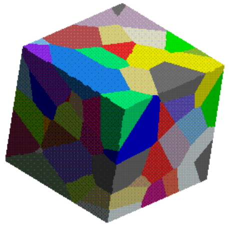
Microscopic scale

- Thermodynamics Modeling



- Modeling of fission gas behavior





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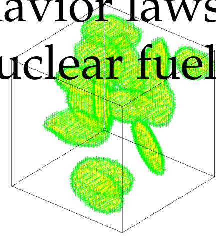

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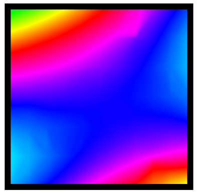

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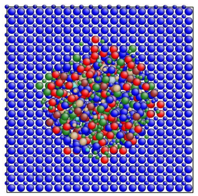

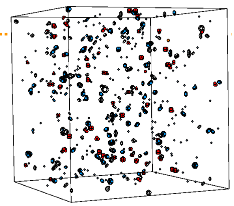

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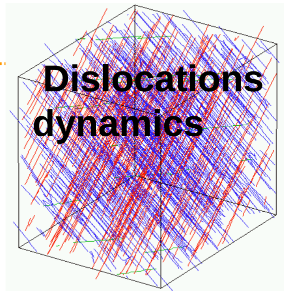
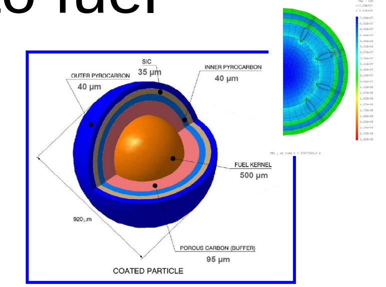


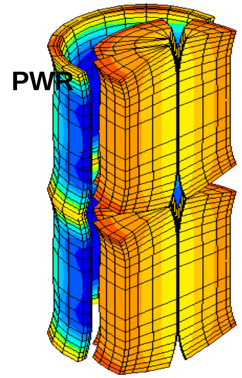
PLEIADES: An Advanced Fuel Performance Code

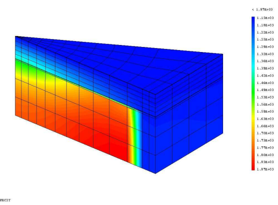
- Multi Reactor Simulation Platform for Fuel Performance Modeling at CEA

PLEIADES

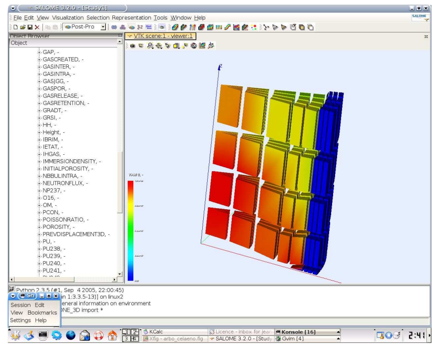
- Software environment dedicated to fuel behavior modeling including all fuel types and reactor concepts







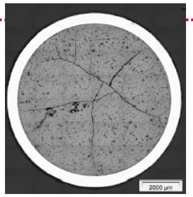
- Set of applications for each reactor concept (**SFR, HTR, GFR, MTR**) adapted to the needs of “user” projects and integrating their modeling (industrial as well as research applications)



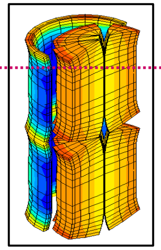
- Capitalization within the same software environment

Multi-scale modeling of nuclear fuel behavior

Macroscopic scale



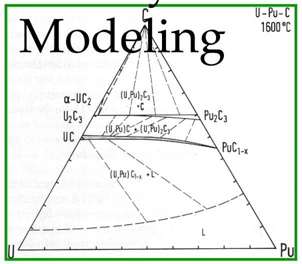
Pellet



• Modeling of fuel behavior

Microscopic scale

• Thermodynamics Modeling



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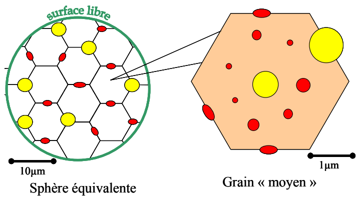

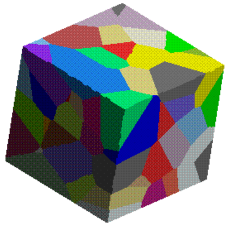

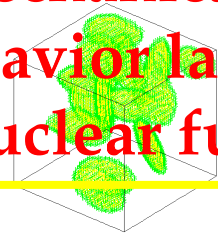

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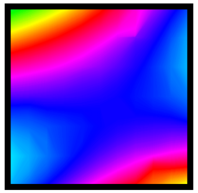
• Atomistic modeling of nuclear fuels: structure, defect stability and fission gas behavior

ab initio



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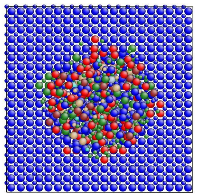

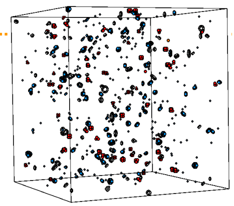

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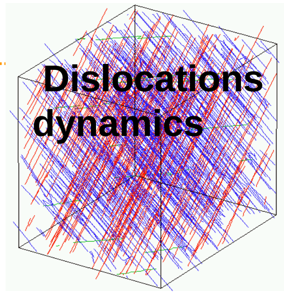
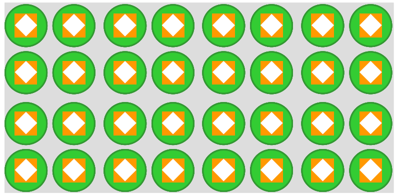


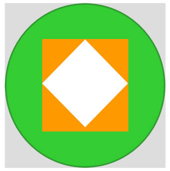
Dislocations dynamics

Thermo-mechanical laws : Homogenization/localization techniques

Heterogeneous domain



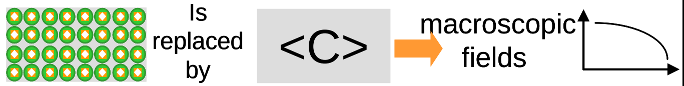
Hypothesis :
An elementary representative volume exists



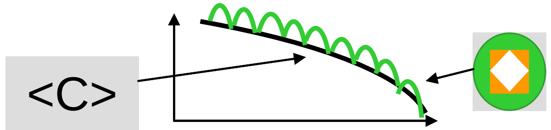
Homogenization
Calculations on elementary representative volume

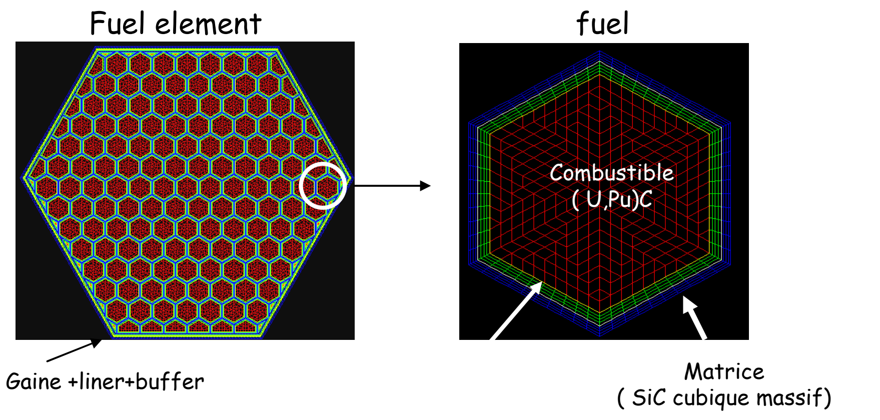
Effective properties

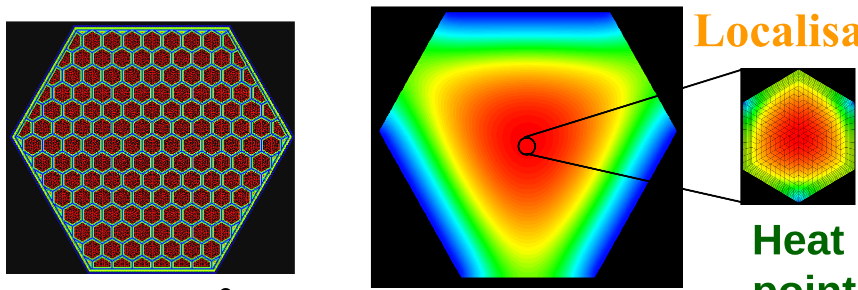
Homogenous problem solved



Localization step







Localisation

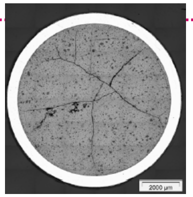
$T_{Hélium}$ = 750 °C

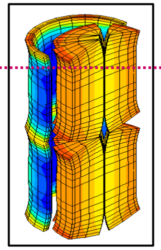
Heat points study

Homogeneous calculations

Multi-scale modeling of nuclear fuel behavior

Macroscopic scale



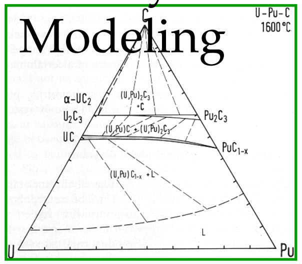


Pellet

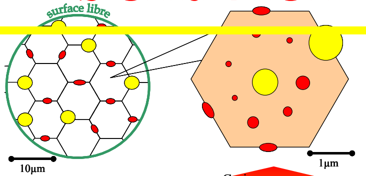
• Modeling of fuel behavior

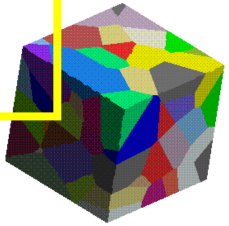
Microscopic scale

• Thermodynamics Modeling

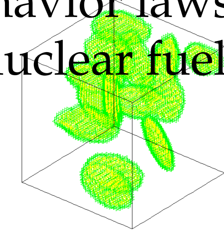


• Modeling of fission gas behavior





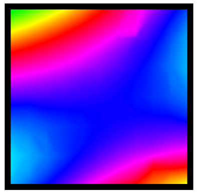
• Thermo-mechanical behavior laws of nuclear fuels



Atomic scale

• Atomistic modeling of nuclear fuels: structure, defect stability and fission gas behavior

ab initio



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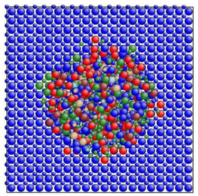

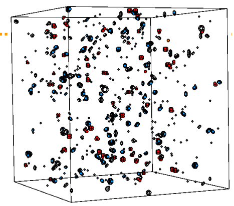

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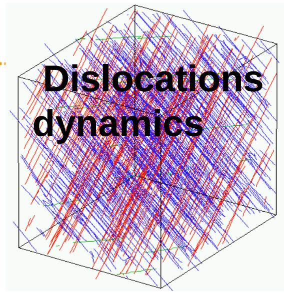
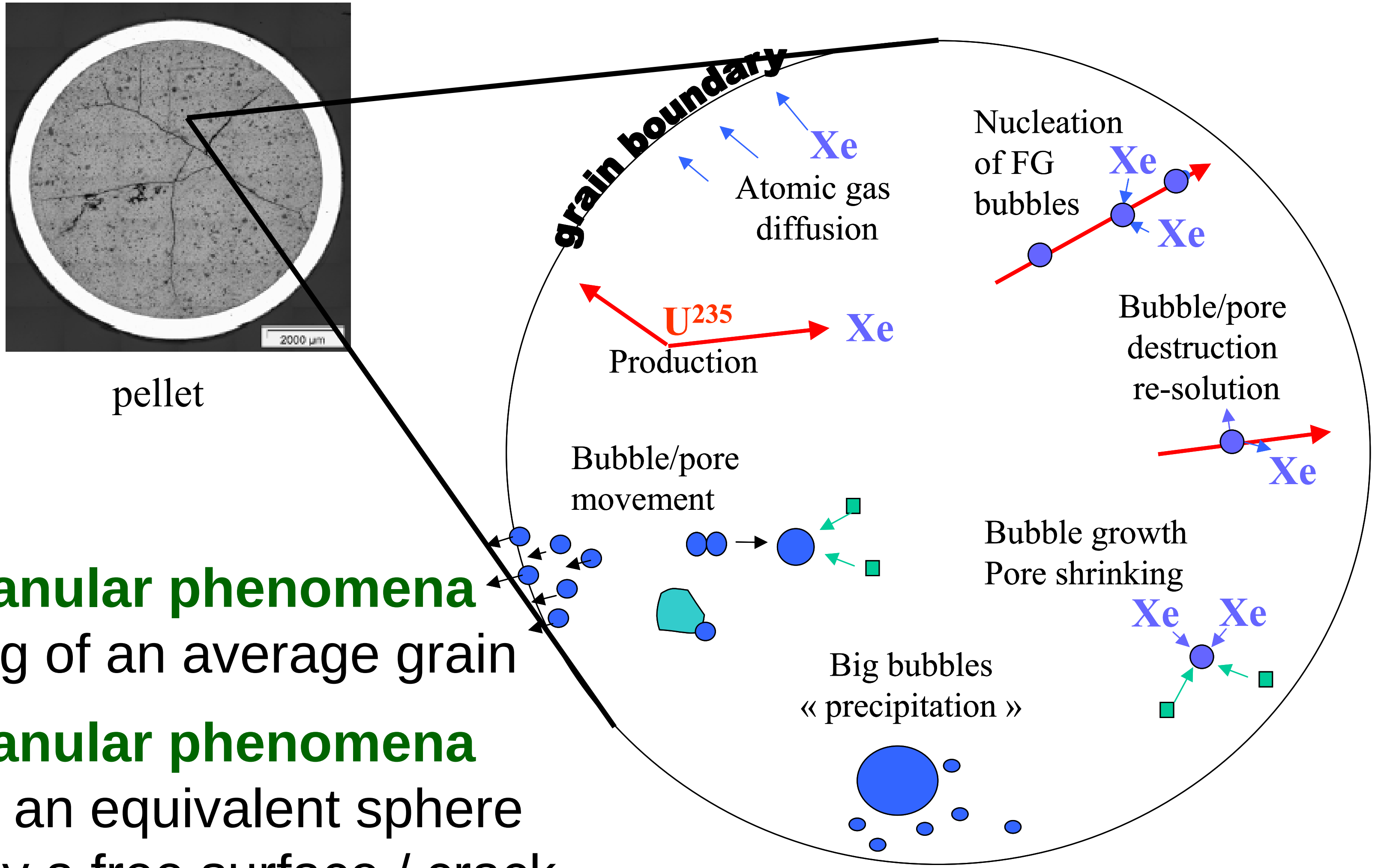


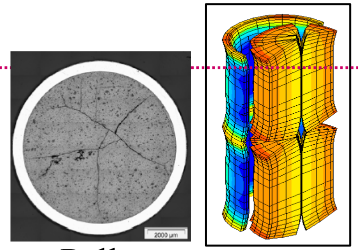
Fission gas behavior in PWR fuel: diffusion models



- **Intragranular phenomena**
  Modeling of an average grain
- **Intergranular phenomena**
  - Use of an equivalent sphere defined by a free surface / crack

Multi-scale modeling of nuclear fuel behavior

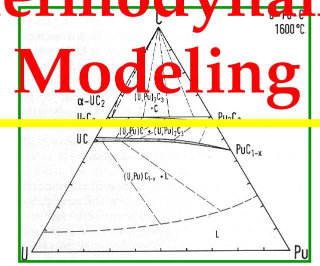
Macroscopic scale


Pellet

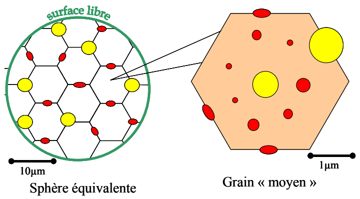
- Modeling of fuel behavior

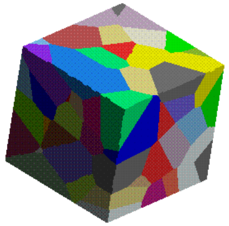
Microscopic scale

- **Thermodynamics Modeling**



- Modeling of fission gas behavior





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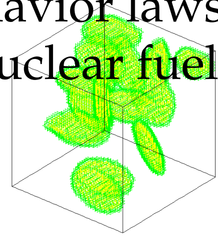
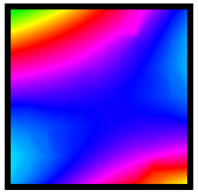


Atomic scale

- Atomistic modeling of nuclear fuels: structure, defect stability and fission gas behavior

ab initio



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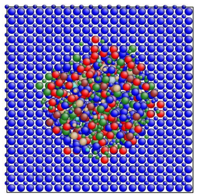

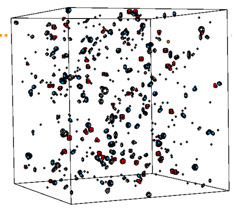

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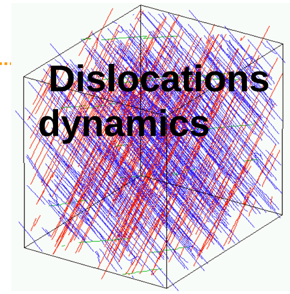
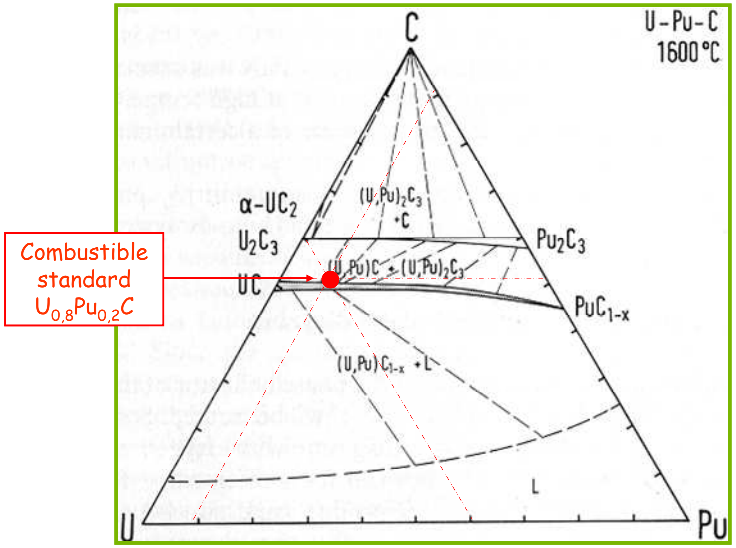


Thermodynamics fuel modeling

- **Thermal and chemical stability of fuels**
  - Phase diagrams of future fuel systems (U, Pu, O, C, N)
  - Impact of minor actinides

- **Chemical interaction of fuel with environment**
  - Cladding, coolant, air, water



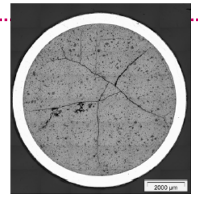
- **Thermodynamics on irradiated fuels**
  - Impact of FP, FP compounds

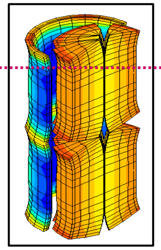
- **Construction of a "Fuel data base" for future fuels**

Guéneau *et al.* J. Nucl. Mater. 344, 191 (2005).

Multi-scale modeling of nuclear fuel behavior

Macroscopic scale



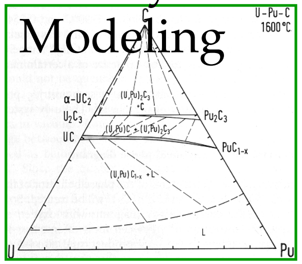


Pellet

- Modeling of fuel behavior

Microscopic scale

- Thermodynamics Modeling



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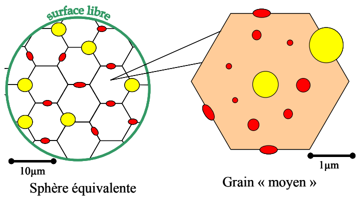

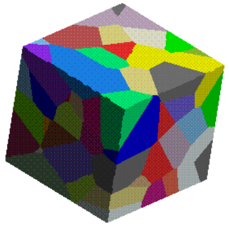

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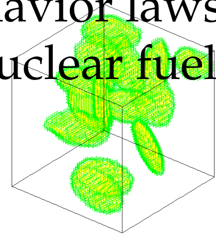
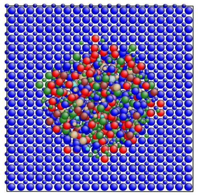


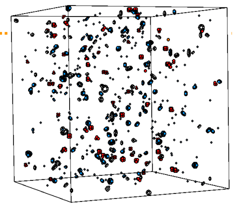
Atomic scale

- **Atomistic modeling of nuclear fuels: structure, defect stability and fission gas behavior**

ab initio

DM





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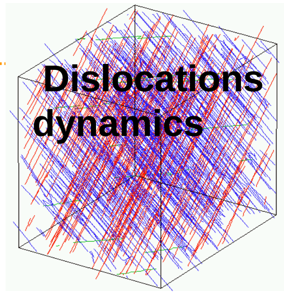


Ab initio modeling of nuclear fuel

Determine and understand physical and chemical properties of fuels at the atomic scale

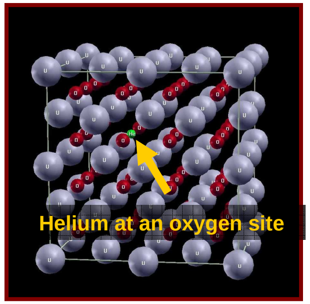
Decouple basic processes

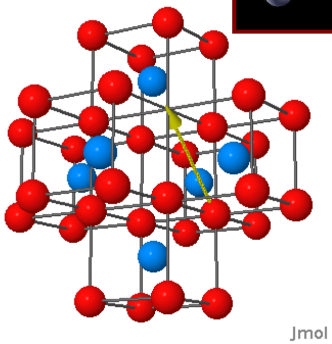
- Stability of a given type of point defect
- Localization of a given fission product
- Migration mechanism of a chemical element

Understanding of the mechanisms involved

Quantify phenomena

- Formation energy of defects
- Incorporation energy of a chemical element
- Structural modification (*swelling*)
- Solubility (*solution energy*)
- Migration (*migration energy*)





Provide basic data ➔ Models at microscopic scale

Classical Molecular Dynamics

Irradiation effects : Displacement cascades in UO_2 :

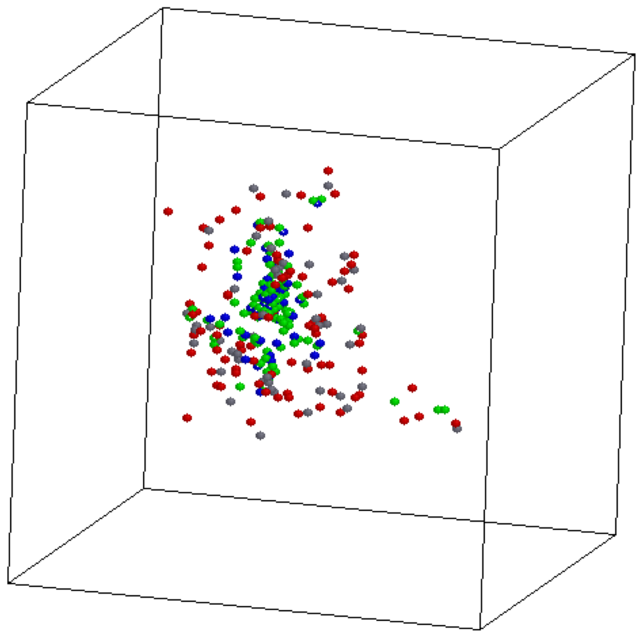
- Concentration of free point defects produced
- Recombining/Clustering of defects: nature, size, number
- Fission product: segregation

Uranium interstitial

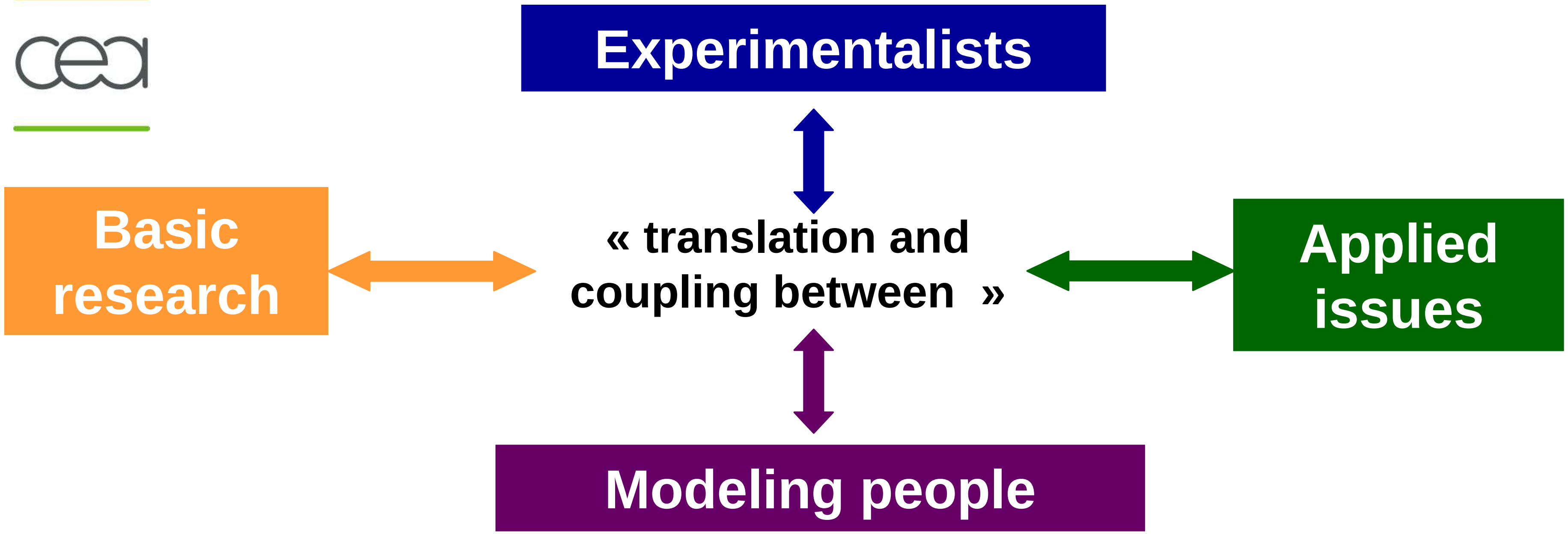
Oxygen interstitial

Oxygen vacancy

Uranium vacancy



Translation language to connect various communities

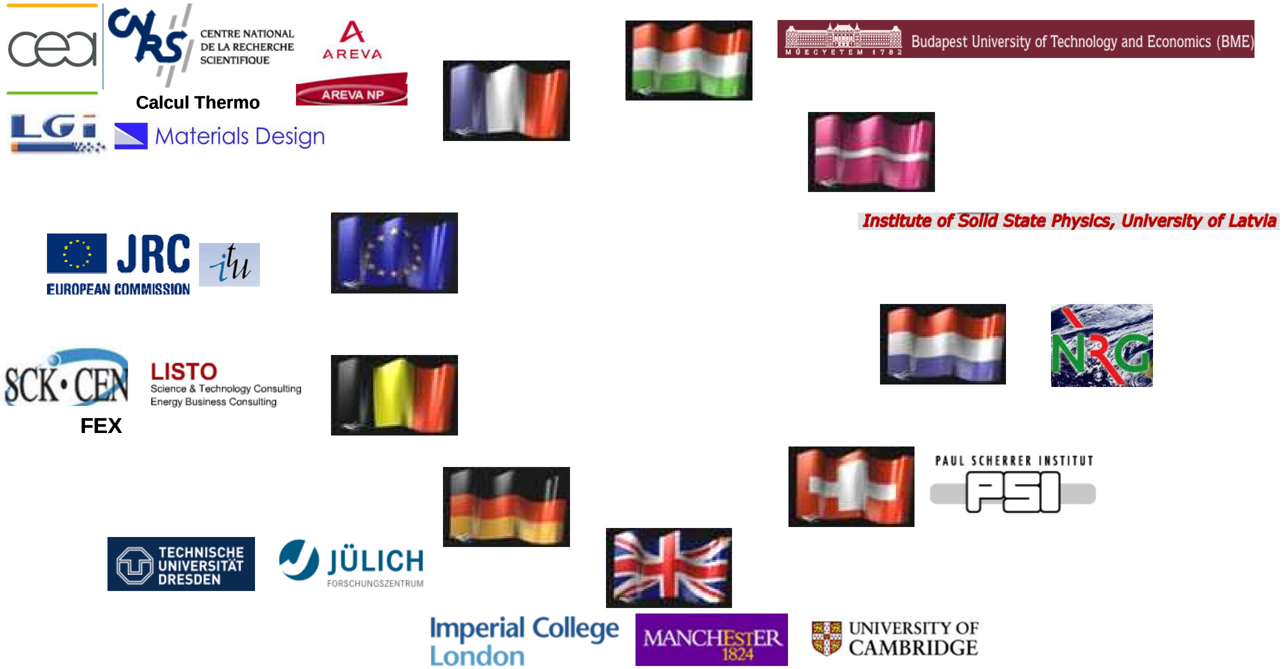


• Instead of asking you what your experimentalist can do for you, ask what you can do for your experimentalist.

• Instead of asking you what your theoretician can do for you, ask what you can do for your theoretician…(B. Schimmelpfennig from the Actinet Theoretical Userlab ThUL).

A European initiative : the F-BRIDGE project

Basic Research in support of Innovative *F*uels Design for the GEn IV systems



F-BRIDGE European project

F B R I D G E

Framework

- Framework of the **Euratom Work programme (FP7)**

- **Area: Advanced Nuclear Systems**
  - improve **efficiency of present systems and fuels**
  - investigate **advanced systems and fuel** in collaboration with efforts of GEN IV, **especially upstream research** (material science, study of fuel cycle and innovative fuels)

- **Topic:** Fission -2007-2.2.1: **Innovative fuels and claddings for generation IV systems**
  - Development and qualification of innovative fuels and claddings
  - Impact: increase efficiency of EU research support to GIF
  - Focusing on cross-cutting and generic issues

F-BRIDGE European project

The needs

- International effort to **increase efficiency in designing innovative fuels** to improve present fuel-cladding systems, to design those for tomorrow

- Up to now fuel development and qualification: successful but long and expensive process essentially based on an **empirical approach**

- Innovative fuel systems: empirical approach **has reached its limit** / difficult to extrapolate to new materials, new environments, or new operating conditions

- **Basic underlying mechanisms** governing manufacturing, behaviour and performance remain poorly understood

- **Challenge** for the next years: complement the empirical approach by a **physically based description** of ceramic fuel and cladding materials

F-BRIDGE European project

Objective : to build a bridge

Integration and Transfer

Basic Research Activities on fuel systems

Technological Issues of the Gen IV Ceramic Fuel Systems

Improvement of a promising Composite Ceramics Concept: the *Sphere-pac Fuel*

Direct impact and feedback on generation IV ceramic fuel systems:

- Fuel design
- Fuel manufacturing
- Irradiation experiments design
- Prediction of the in-pile behavior

F-BRIDGE European project

F ★ B R I D G E

Beyond the state of the art

- Novel approach: brings together numerous significant actors of the nuclear fuel field: scientists, engineers and end-users

- A special team: shall ensure a direct translation of technological issues into basic research investigations, as well as a facilitated transfer of the knowledge acquired to fuel performance codes, design and manufacturing.

- First integrated project on non-metallic nuclear fuels and cladding
  - All relevant length and time scales from the atomic description to macroscopic systems
  - Investigation of important properties of fuel and ceramic cladding materials using a combination of modelling and experiments

- Main contribution : to capitalise on recent advances in both theoretical approaches and experimental techniques to develop fuel behaviour descriptions that have a much sounder physical basis

Part 1 conclusions

- A real need to effectively connect experiment and modelling to improve the knowledge on fuel behaviour under irradiation and design the fuels of tomorrow

- Fuel studies are ongoing to achieve a much deeper understanding description using finer characterization as well as modelling down to the atomic level

- Experiment and modeling have to be coupled and to feed each other

- Experiments need modeling and vice versa