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# CHALLENGES RELATED TO MINOR ACTINIDE BEARING FUELS

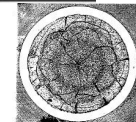
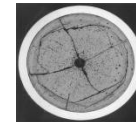
**Dominique WARIN**

***Atomic Energy and Alternative Energy Commission***

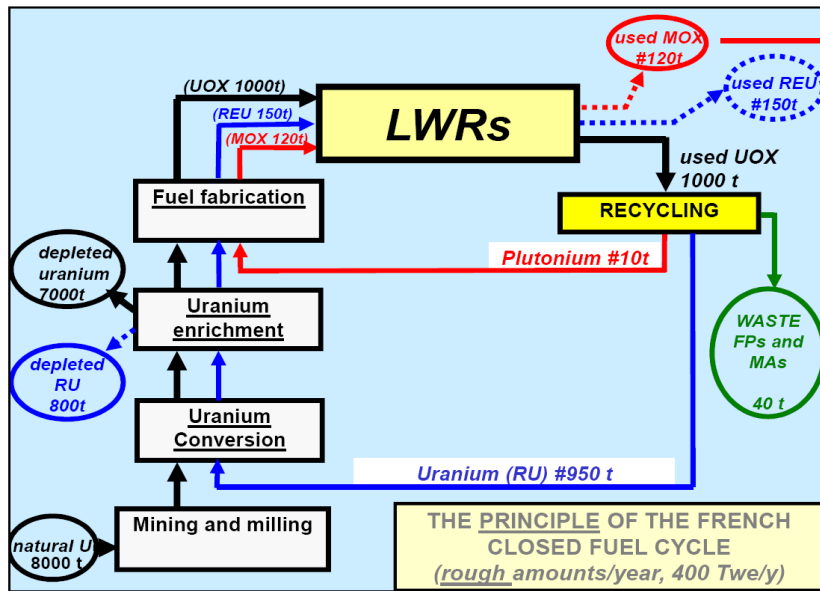
European Nuclear Conference 2012 – Manchester, UK  
December 9-12, 2012

## OUTLINE OF THE PRESENTATION

- The context for transmutation
- **The specificities** of MA bearing fuels
- Knowledge from irradiation experiments
  - Fast reactor : **homogeneous** transmutation mode
  - Fast reactor : **heterogeneous** transmutation mode
  - **Oxide** / metal, carbide or nitride
- **Fabrication** of MA bearing fuels
- **Modeling** of MA bearing fuels
- Conclusion



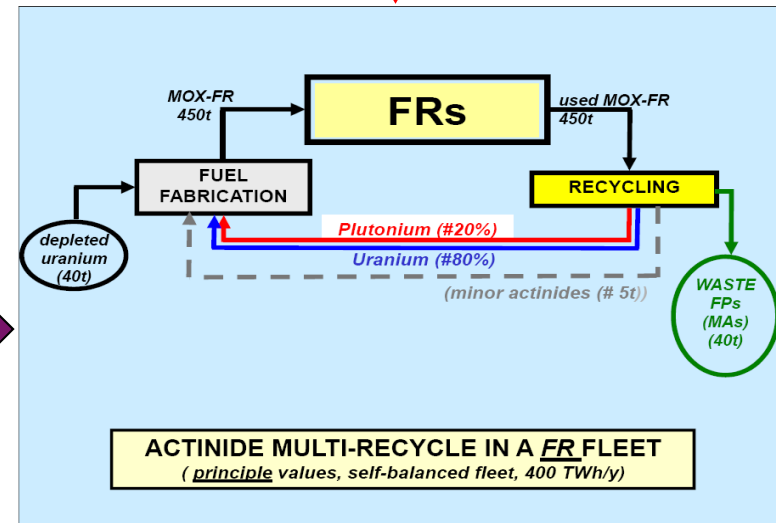
# FROM LWR RECYCLING TO FR RECYCLING



1. Pu stored in MOX LWR spent fuel recycled in MOX SFR to start the SFRs deployment
2. MA recycled in SFR

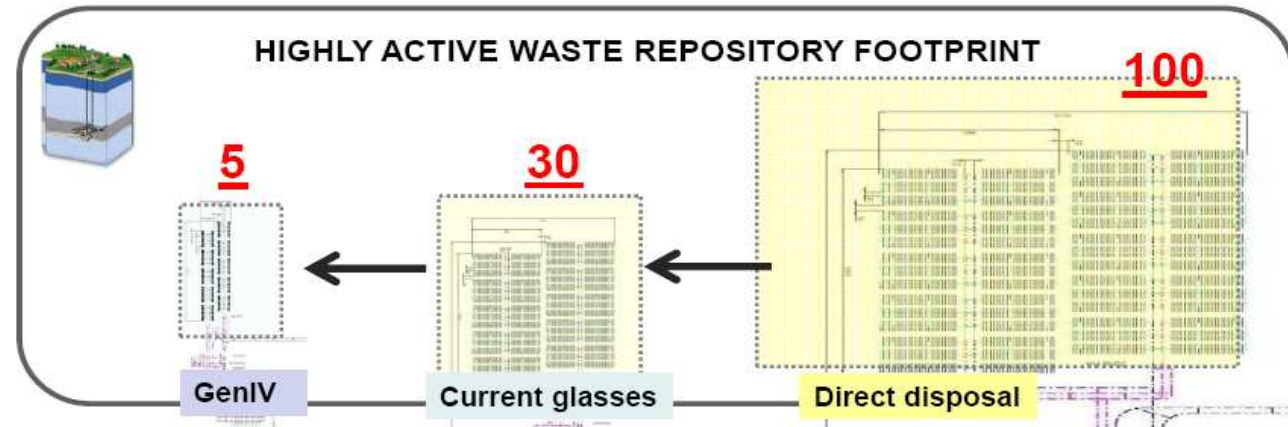
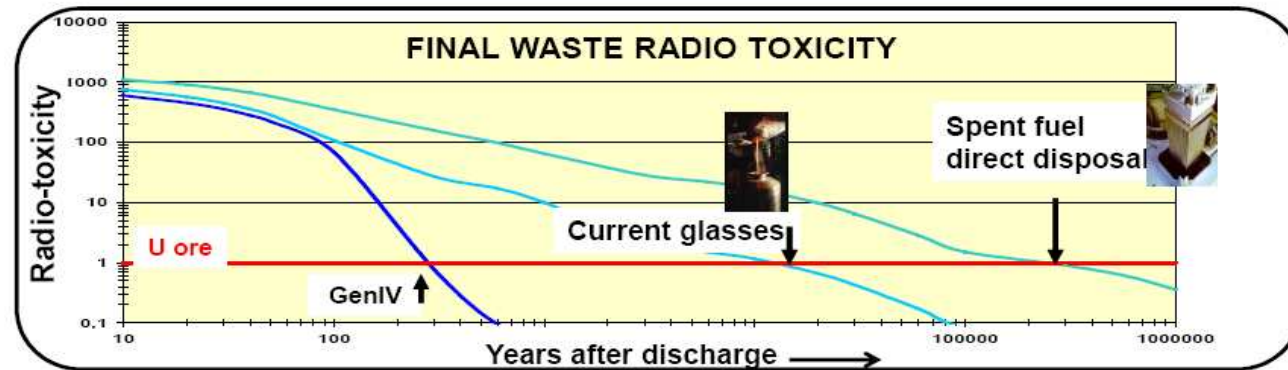
- Scenarios have to be flexible

Both systems can coexist during a transition phase

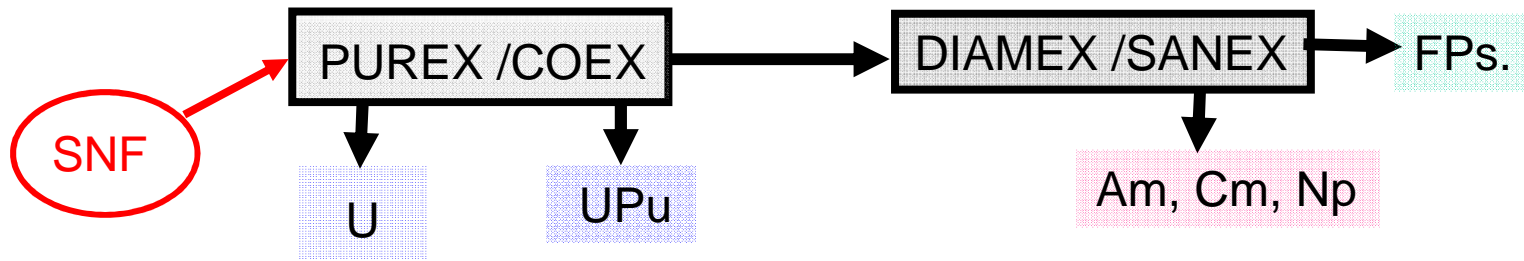


# GENERATION IV SYSTEMS WASTES

Transmutation with **GEN IV Fast Reactor** systems will allow to avoid MAs in the waste repository : **significantly lower radio-toxicity life time and lower footprint**



# MA PARTITIONING AND TRANSMUTATION



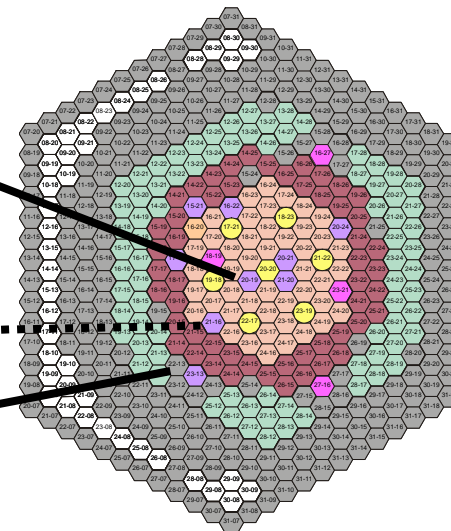
Innovative partitioning processes based on the design of new extractants

MA recovery processes have been successfully experimented, (kgs-scale, genuine spent fuel)

**HOMOGENEOUS** # 2% MA diluted in the fuel

**HETEROGENEOUS:**

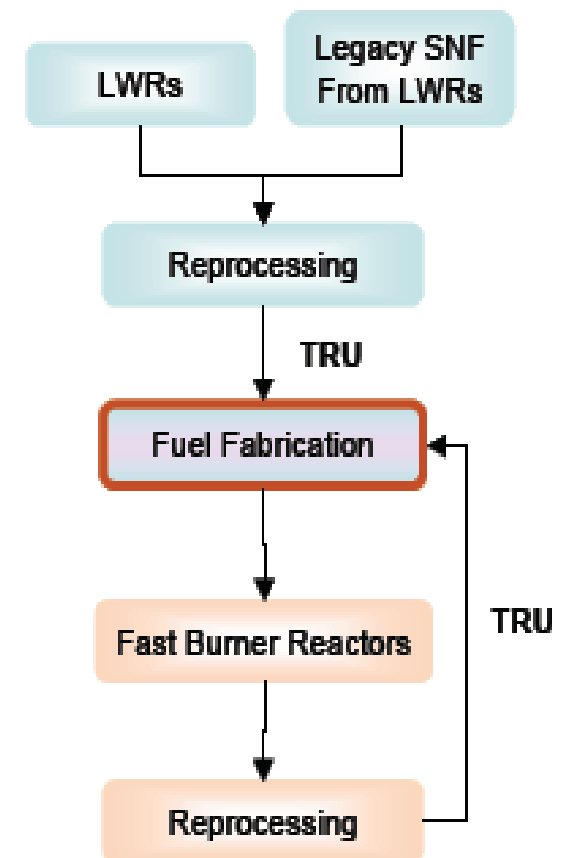
- « once through targets » (inert matrix)
- multi-recycled «MA bearing blankets » (# 10-20% MA on UO<sub>2</sub>)



## THE SPECIFICITIES OF MA BEARING FUELS

Transmutation fuel development is considerably more challenging than for conventional fuels :

- Multiple elements in the fuel
  - U, Pu, Np, Am, Cm
- Varying thermodynamic properties
  - e.g. High vapor pressure of Am
- Impurities from separation process
  - e.g. High lanthanide carryover
- High burnup requirements
- High helium production during irradiation
- Remote fabrication & quality control
- Fuel must be qualified for a variable range of composition
  - Age and burnup of LWR SNF
  - Changes through multiple passes in FR
  - Variable conversion ratio for FR



# THE SPECIFICITIES OF MA BEARING FUELS

## ■ Thermal, physics and chemistry properties of MA-UPuO<sub>2</sub> / UPuO<sub>2</sub> :

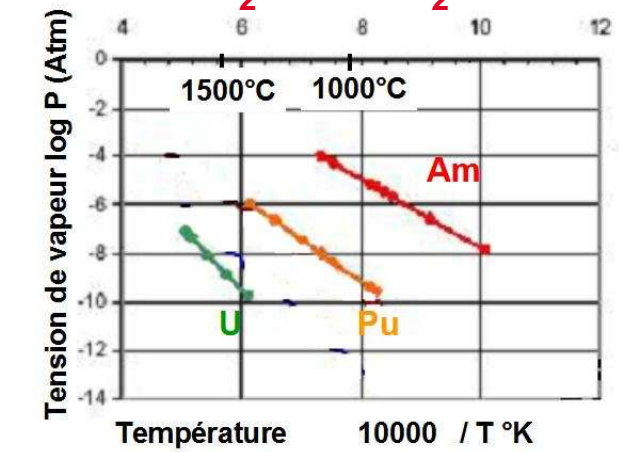
- Melting temperature : lower
- Thermal conductivity : lower
- Volatility : higher
- Oxygen potential : higher

## ■ Thermal and radio toxicity conditions :

- Thermal power : higher :  
**P<sub>th</sub> MA target = ~10 x P<sub>th</sub> LWR-MOX**
- Neutron emissivity : higher : MA target = ~2000 x LWR-MOX

## ■ High He production : alpha decay of <sup>242</sup>Cm , coming from beta decay of <sup>241</sup>Am

- ⇒ **MAs recycling has impacts on :**
- Fabrication, handling, transport, control,... conditions
  - In pile behavior



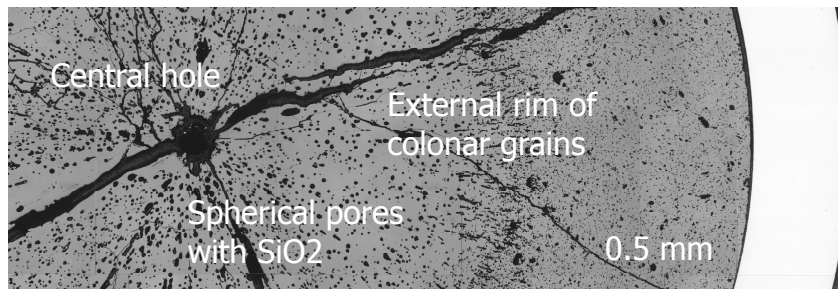
# MA BEARING FUEL FOR HOMOGENEOUS RECYCLING : IN FR PILE BEHAVIOR

## ■ Am1 in JOYO :



■  $P_{LIN} = 416$  to  $430$  W/cm

■ Irradiation : 10 minutes, and 24h

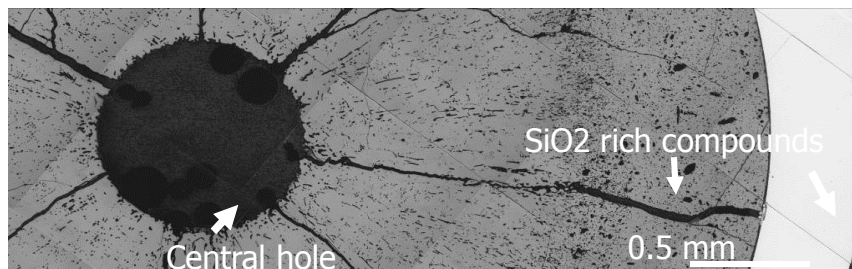


### First test, 10 min

- SiO<sub>2</sub>-MOX precipitates (around the central hole)
- Formation of spherical pores, moving to the center



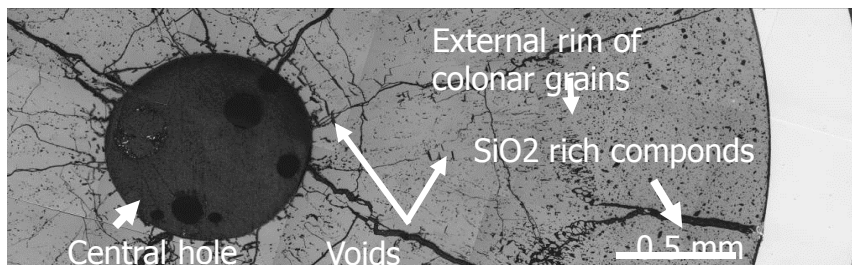
O/M 1.98



### Second test, 24 h

- SiO<sub>2</sub> rich compounds, next to the cracks
- Formation of holes, moving to the center
- Large central hole is formed

O/M 1.95





# MA BEARING FUEL FOR HOMOGENEOUS RECYCLING : IN FR PILE BEHAVIOR

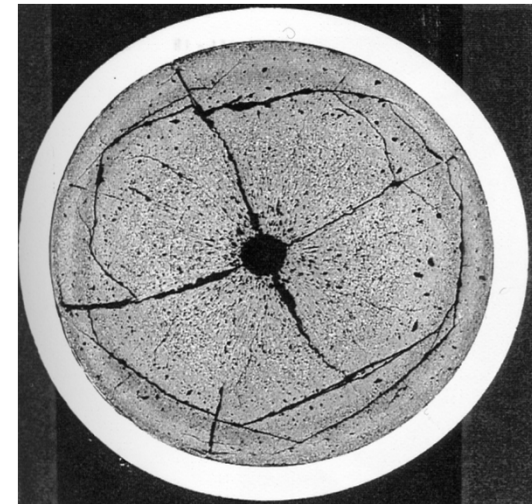
- **SUPERFACT homogeneous** transmutation mode in SFR Phénix :

- $U_{0.74}Pu_{0.24}Am_{0.02}O_2$  and  $U_{0.74}Pu_{0.24}Np_{0.02}O_2$
- 382 efpd,  $P_{LIN} = 380$  to  $325$  W/cm
- Burn up = 6.7 at%

- **Transmutation rate** at mid plan :  
28% for  $^{241}Am$ , and 30% for  $^{237}Np$

- U, Pu, Am and Np radial concentration profiles are very flat : **no specific redistribution of MAs**

- **Microstructure evolution is similar to UPuO<sub>2</sub>**



For still limited power and burn up, **no influence of the MAs concentration on in-pile behavior, except for the He gas release (4 times higher / UPuO<sub>2</sub>)**

# MA BEARING FUEL FOR HETEROGENEOUS RECYCLING : IN FR PILE BEHAVIOR

- **SUPERFACT heterogeneous** transmutation mode in SFR Phénix :

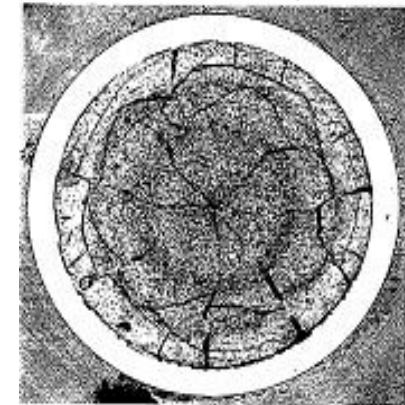
- $U_{0.6}Am_{0.20}Np_{0.20}O_2$ , 4 g MA/cm<sup>3</sup>
- 382 efpd,  $P_{LIN} = 174$  to 273 W/cm
- Burn up = 4.1 at%

- **Limited restructuration of the fuel :**

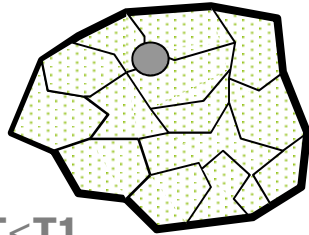
- Limited fuel-cladding interaction
- “Cold” fuel : no formation of central hole
- **High He production and retention**

mm <sup>3</sup> /g fuel	Xe Kr	He
Standard UPuO <sub>2</sub> , 6at%	1220	40
<b>UAmNpO<sub>2</sub>, 4at%</b>	<b>764</b>	<b>2970</b>

No formation of  
central hole



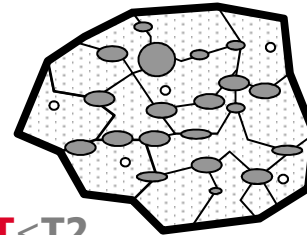
- In a “cold” fuel, **the He behavior in the UO<sub>2</sub> matrix has to be carefully studied** (potential clad failure under operation)



$T < T1$   
500 to 700°C

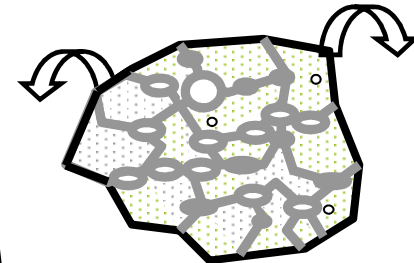
- **Low matrix swelling** : He atoms isolated and no mobile

## MA bearing blanket



$T1 < T < T2$

- **Potential significant swelling**
- *Fuel-clad interaction risk*



$T2 < T$   
1100 to 1400°C

- Moderated swelling due to high He release

- Conditions of **swelling-release** has to be known quantitatively , in **order to design an heterogeneous MA transmutation pin** :
  - On going experimental program (in the HFR and Osiris MTRs, with  $UO_2$  matrix + Am concentrations, **open/closed porosity**) in order to obtain **experimental data in support of fuel design**
  - Second experimental phase : **for optimizing the microstructure**
  - Third phase : **high burn up experiments of representative fuel elements** under operating and qualifying conditions

# POSSIBLE TRANSMUTATION FUEL

- Mixed (U,Pu): **Oxide (SFR, GFR, LFR...)** versus **Carbide (GFR, ...), Nitride (SFR), Metal (SFR, ...)**

- Two different technologies:

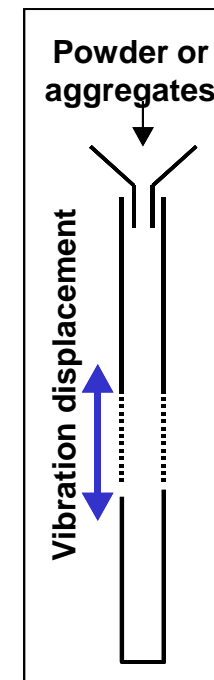
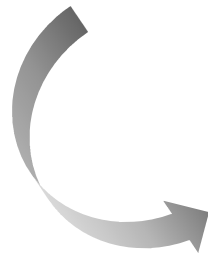
- **Hydrometallurgy:** powders to pellets in fuel pin

- **Pyrochemistry:**

- well adapted to metal fuel

- aggregates by Vibropack in fuel pin

- metal fuel casting



## OXIDE VERSUS METAL, CARBIDE OR NITRIDE ?

### ■ Metal :

- A satisfactory option for SFR, good knowledge in USA
- Large on going experiment program in US Labs, using Idaho-ATR (UPuAmZr) and SFR Phénix (PuAmZr)

### ■ Carbide :

- An option for SFR, “almost” reference for Gas cooled GFR
- UPuC knowledge in Europe (limited) and in India (large)
- **No irradiation tests so far of MA-UPuC**
- **Pyro phoricity of divided carbide materials**

### ■ Nitride :

- An option for SFR, UPuN knowledge in Japan, Europe and USA
- **Complexity of fabrication process** (carbothermic reduction, N15 enrichment ?)
- **MA-nitride stability as a function of temperature**
- PuAmZrN and PuAmN irradiated in SFR Phénix

## Laboratory scale process

- **Synthesis of MA compound powders**, starting from separated MA nitric solution (interface co-conversion)
- A promising process : **the oxalic co-precipitation**, calcination, then direct-powder or  $UO_2$ -diluted powder pelletizing

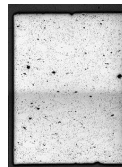


Pu

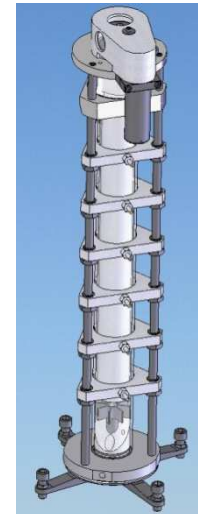
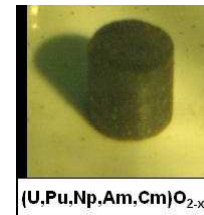
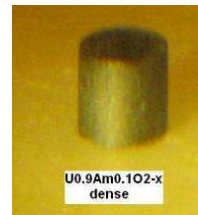


U and Pu

COPIX (UPu) $O_2$  irradiation test in Phenix, 2008-2009



U, Pu, Np, Am,..Cm



- **Characteristics of the powders** : physico-chemistry, purity, flowability, **sintering properties**,...
- **Technology** : continuous precipitation apparatus: vortex effect, pulsed column,...
- **Remote handling operations**



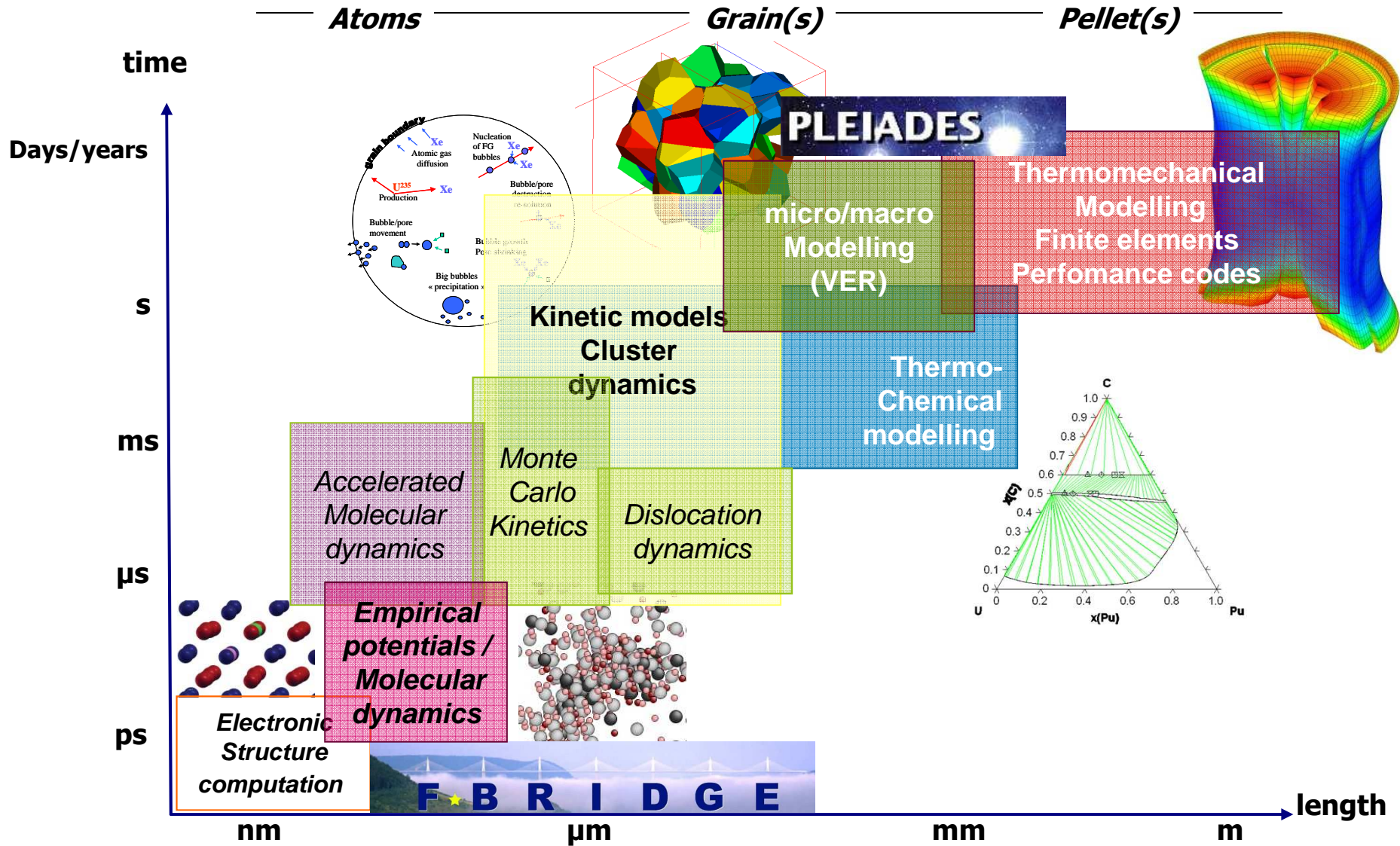
Fuel performance prediction requires understanding of multiple phenomena :

- Thermal conductivity
- Thermal expansion
- Specific heat
- Phase diagrams
- Fission gas formation, behavior and release
- Materials dimensional stability
  - Restructuring, densification, growth, creep and swelling
- Defect formation & migrations
- Diffusion of species
- Radial power distribution
- Fuel-clad gap conductance
- Fuel-clad chemical interactions
- Mechanical properties

*Dynamic properties:*  
Changes with radiation effects, temperature, and time

*Nonlinear effects*  
Initial condition dependence  
(fabrication route)

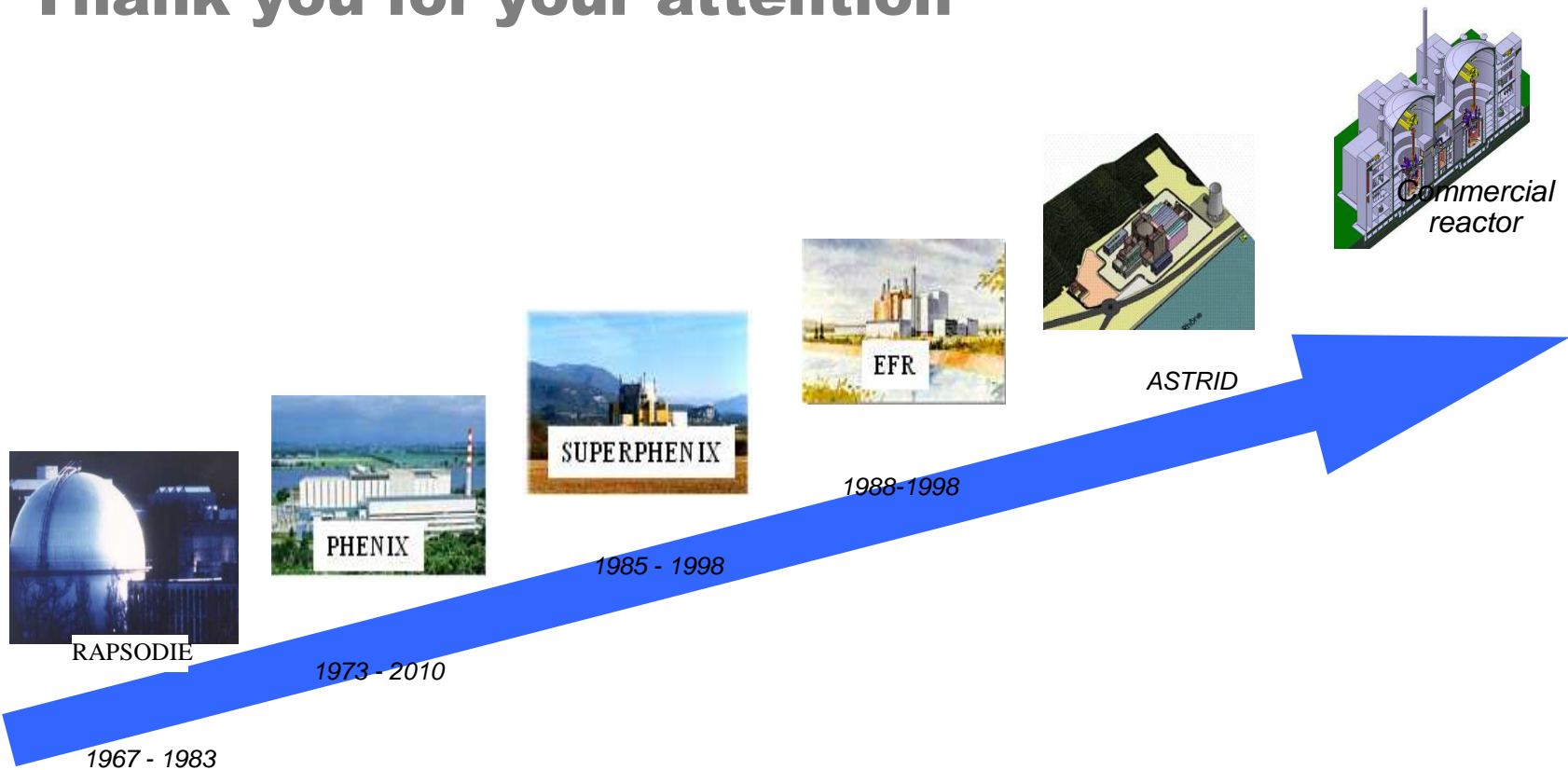
# (TRANSMUTATION) FUEL MULTI-SCALE MODELING





- **Homogeneous** transmutation mode :
  - Satisfactory in pile behavior, “similar” to standard  $\text{UPuO}_2$
  - Irradiation tests at high burn up still necessary (Monju, ASTRID,...)
  - **Towards the qualification**, at pin scale
  
- **Heterogeneous** transmutation mode :
  - In pile behavior still to be fully known
  - Concept to be optimized : microstructures, pin design, irradiation conditions,...
  - **In pile irradiations : data, feasibility** (HFR, Osiris, ATR, Joyo, RJH, Monju, ASTRID,...) ; **EC projects**
  
- Accuracy and validation of **nuclear data**
  
- **Fabrication operation : remote handling** operations at significant scale
  
- **Modeling** development
  
- **Curium issue ?**

# Thank you for your attention



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Manchester  
December 11, 2012

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