2055-28

Joint ICTP/IAEA School on Physics and Technology of Fast Reactors Systems

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Advanced partitioning of MOX fuels by aqueous route (2)

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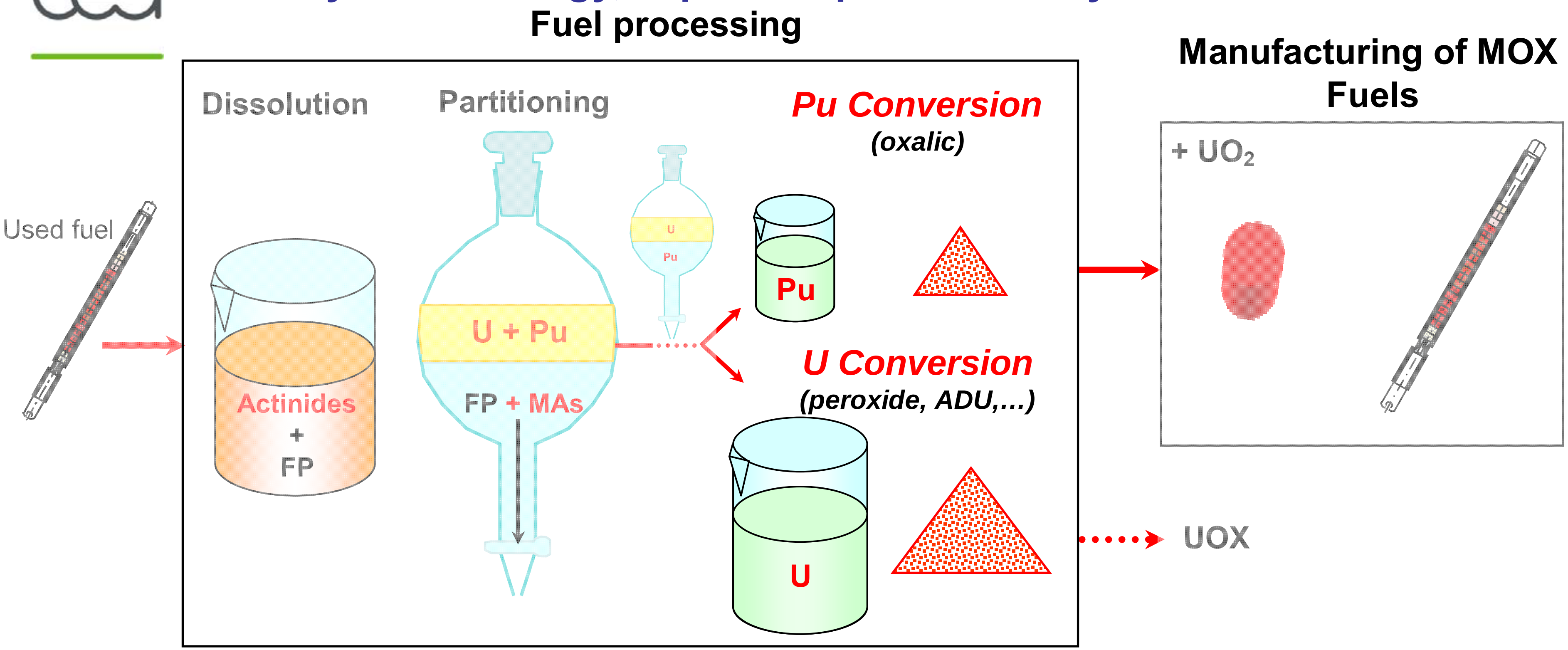
General consideration on existing industrial conversion for U-Pu

- The PUREX process – AREVA's UP2 & UP3 (La Hague)
  - **Hydrometallurgy, separated purification cycles for U and Pu**



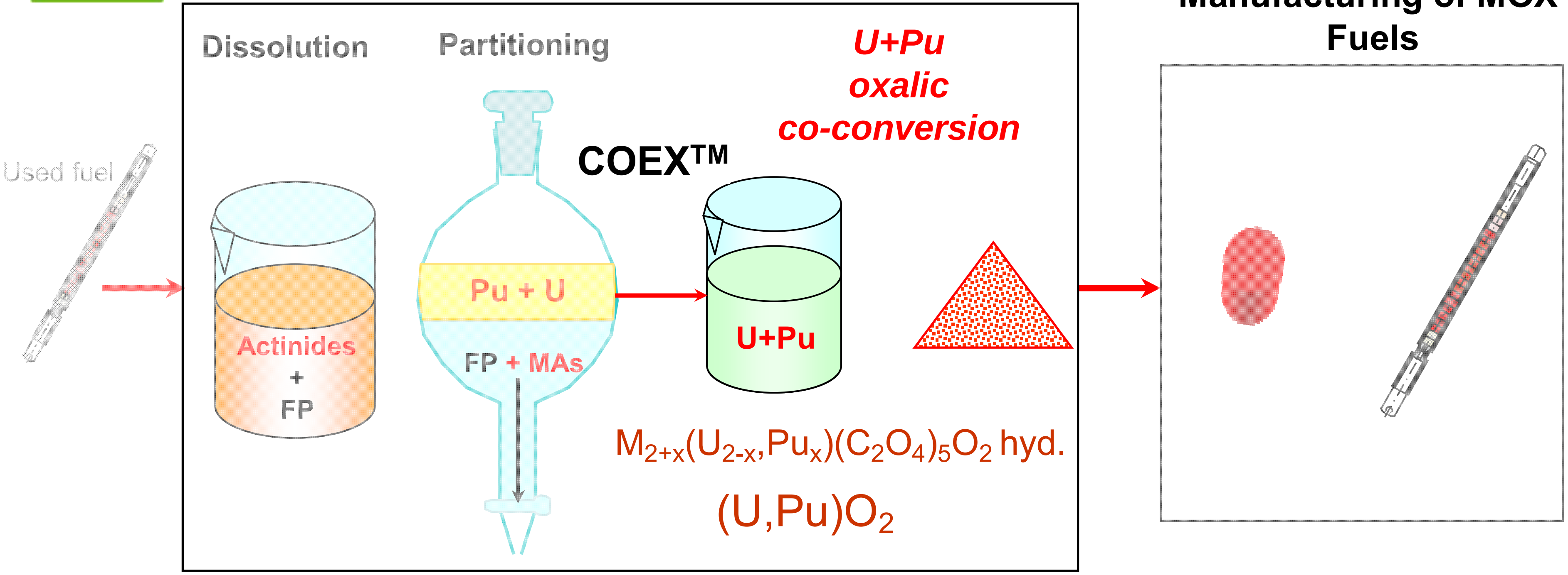
  - **Conversion**
    - For uranium and plutonium : conversion of each actinide into oxide (following a specific purification cycle), *via* continuous oxalic precipitation for Pu and continuous peroxide precipitation (in France) for U.
    - PuO_2 = one of the starting material for MOX fabrication (MELOX process).

General considerations on « existing » industrial conversion for (U,Pu)

- Advanced processes for Gen III+/IV fuel cycle (e.g. COEX^{TM})
  - **Ex: hydrometallurgy, co-processing of uranium and plutonium**

Fuel processing

Manufacturing of MOX Fuels



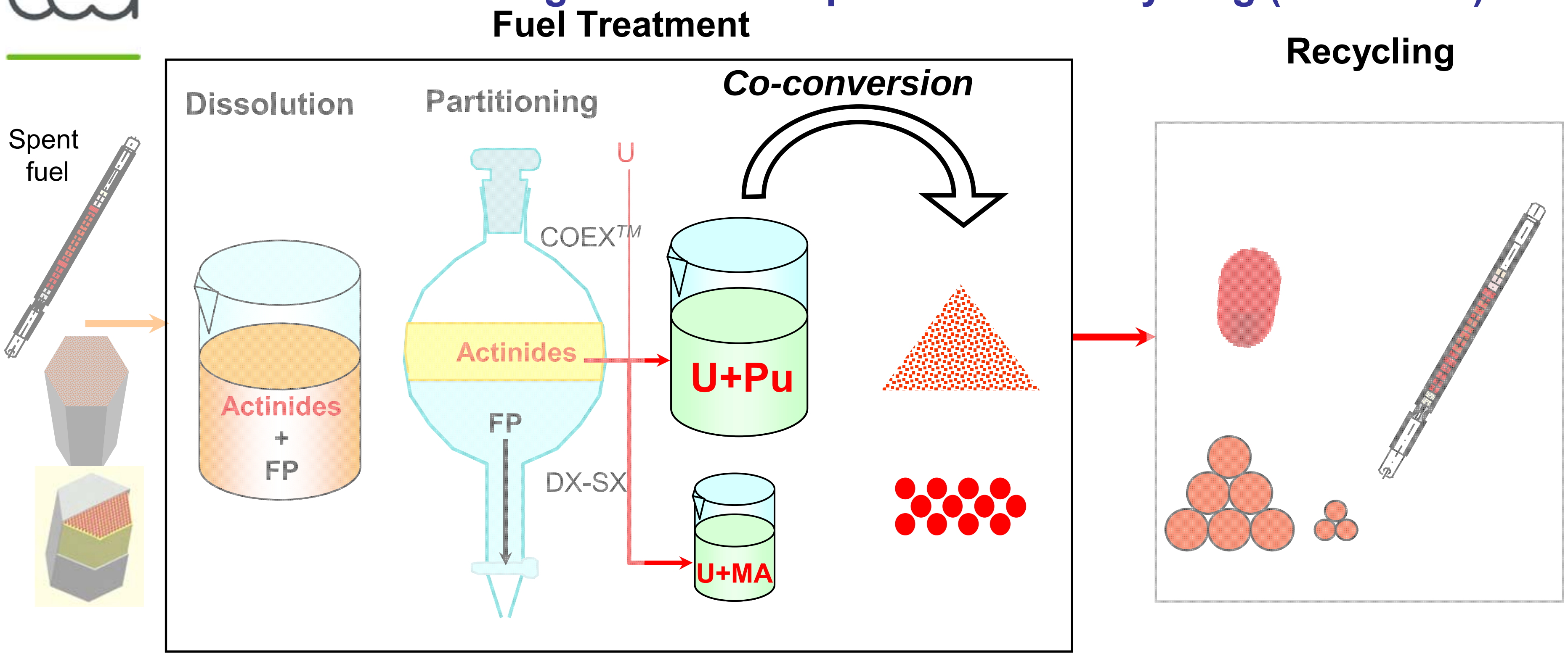
$M_{2+x}(U_{2-x},Pu_x)(C_2O_4)_5O_2$ hyd.

$(U,Pu)O_2$

  - **Co-conversion**
    - Co-conversion of U and Pu into oxide following the partitioning steps and more integrated to the fuel refabrication (MOX).
    - U+Pu co-conversion product $(U,Pu)O_2$ = fuel precursor (LWR then FR).

General considerations on future conversion, for MAs

- Innovative processes: e.g. COEX^{TM} + MAs Transmutation (het.)
  - **U+Pu comanagement and separated MAs recycling (het. mode)**

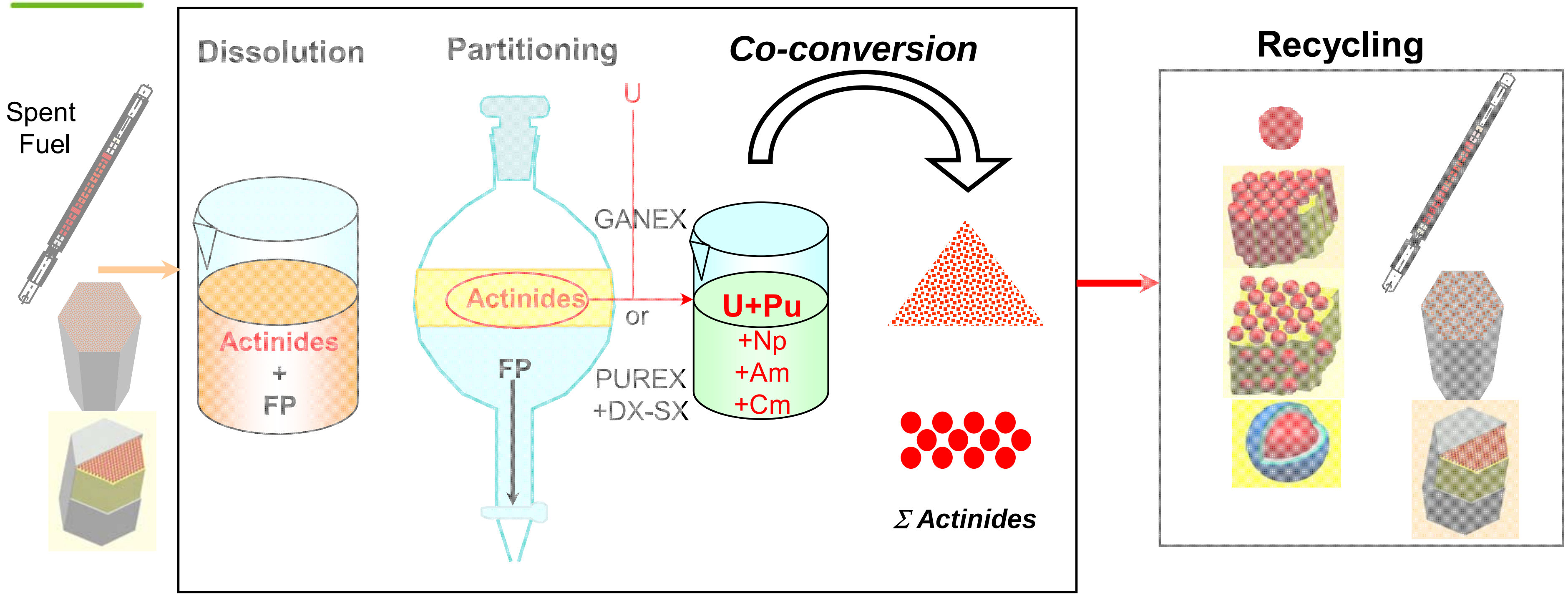


  - **Mixed compounds – a major UPu stream + a specific MAs stream**
- **e.g. oxalic process:**
- $M_{2+x}(U_{2-x},Pu_x)(C_2O_4)_5O_2$ **hyd. +** $M_{2+x}((U,Np)_{2-x},(Am,Cm)_x)(C_2O_4)_5O_2$ **hyd.**
- $(U,Pu)O_2$ **+** $(U,MA)O_{2-x}$

General considerations on future conversion, for MAs

- Advanced processes: e.g. GANEX with MAs transmut. (homogeneous)
  - **Homogeneous recycling of all the actinides together**

Fuel treatment



  - **One mixed actinides end-product – One fuel precursor**

- **e.g. oxalic process:**
- **$M_{2+x}((U,Np)_{2-x},(Pu, Am,Cm)_x)(C_2O_4)_5O_2$ hyd.**
- **$(U,Np,Pu,Am,Cm)O_{2-x}$**

Methods of coconverting actinides into oxide

- Conversion and coconversion existing/possible methods
  - **Coprecipitation + Thermal treatment**

Oxalic	Pu(IV), Pu(III), An(VI)
Hydroxide	U(VI), Pu(IV), An(III)
Carbonate	U et Pu(VI), An(V)
Peroxide…	U(VI), Pu(IV)

  - **Sol-Gel + Thermal Treatment** **U(VI), Pu(IV), An(III), An(V)**
    - Colloidal Sol-Gel
    - Internal or external gelation (gelating additives)
  - **Thermal Denitration** **U(VI), Pu(IV)**
    - Direct Denitration with/without additives
    - Plasma Denitration
    - MW Denitration
    - Denitration within a template matrix (e.g. ceramic beads)

⇨ New requirements for $\text{Gen}^{III}/\text{Gen}^{IV}$ systems

- Economy of resources (recycling of U and Pu)
- Increased proliferation resistance (co-processing of actinides)
- **MAs** management (↘ long-term radiotoxicity and volume of waste)

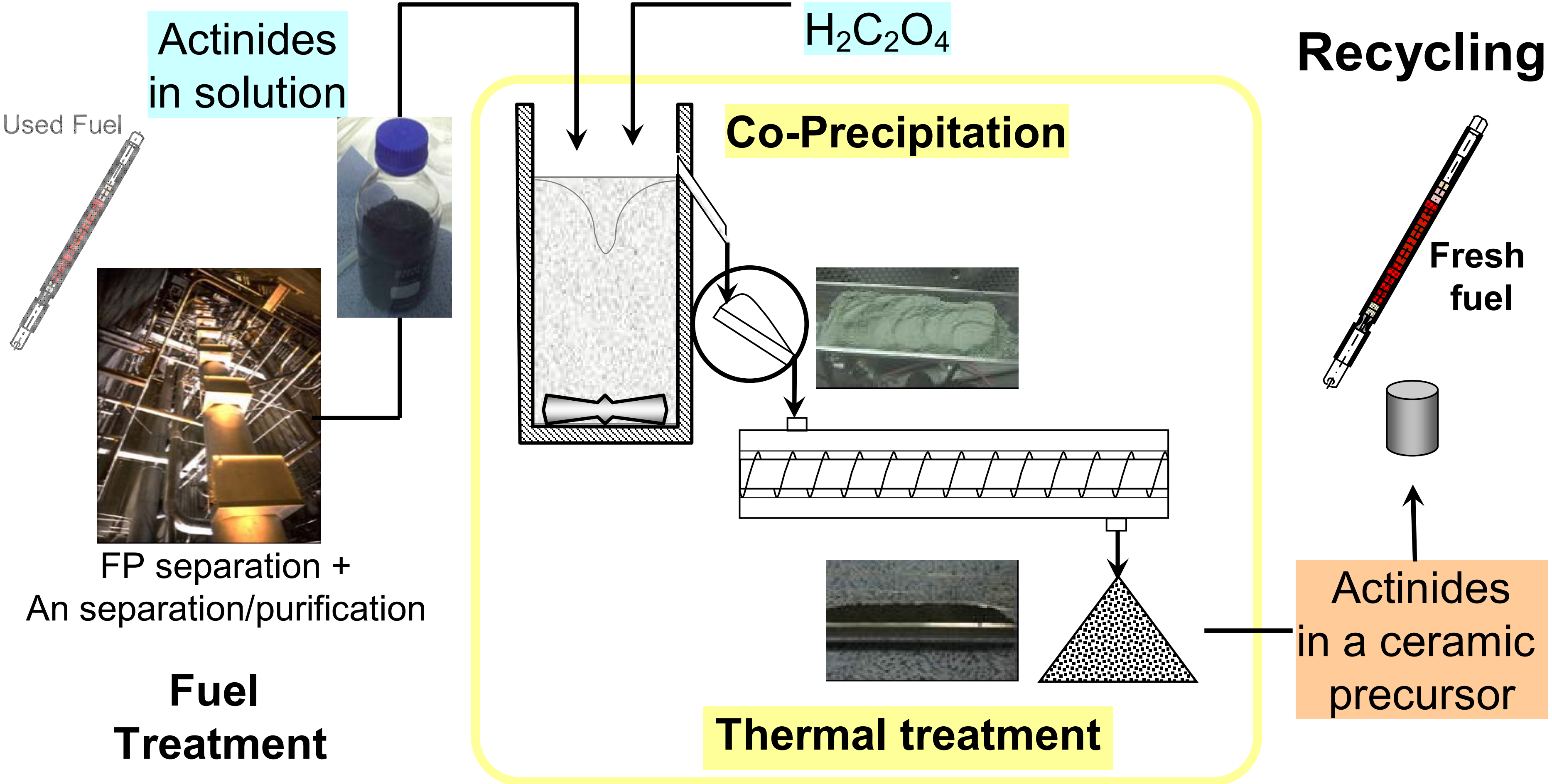
  High activity and remote operation, thermal and radiolysis effect…

  Innovative concepts of fuels and reactors…

⇨ **Innovative processes**

Methods of coconverting actinides into oxide

- Oxalic coconversion of Actinides into Mixed Oxide
  - From actinides in spent fuel solution to their recycling into a fresh fuel



Actinides in solution

$H_2C_2O_4$

Recycling

Used Fuel

Co-Precipitation

Fresh fuel

FP separation +
An separation/purification

Fuel Treatment

Actinides in a ceramic precursor

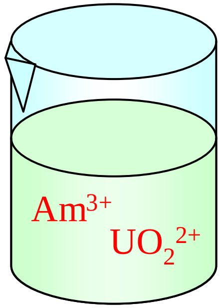
Thermal treatment

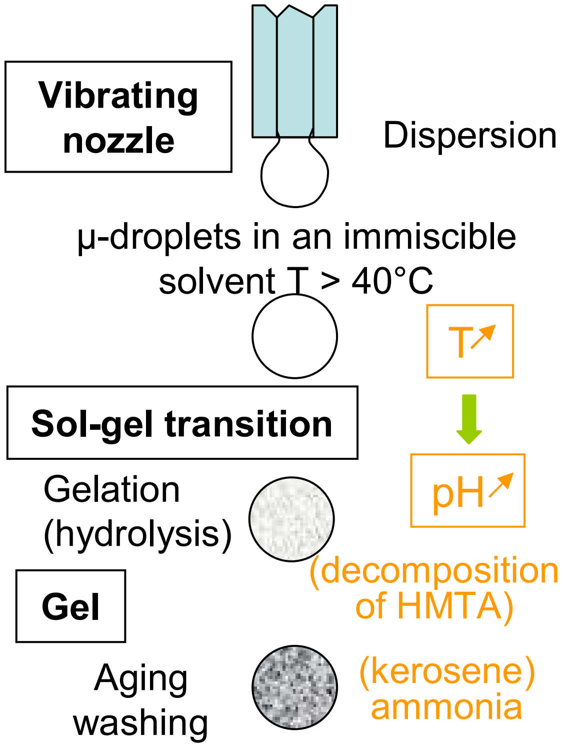
Methods of coconverting actinides into oxide

Sol-gel (e.g. internal cogelation)

Actinides solution

+ HMTA
+ Urea
+ Carbone
(carbide)





Vibrating nozzle

Dispersion

μ-droplets in an immiscible solvent T > 40°C

T↗

Sol-gel transition

Gelation (hydrolysis)

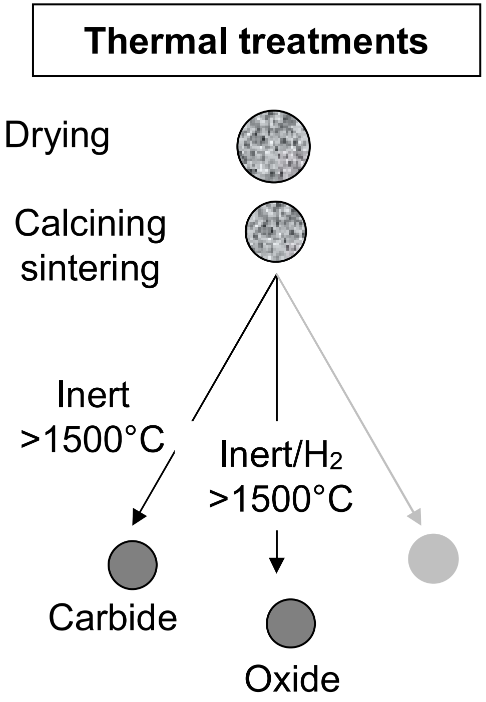
pH↗

(decomposition of HMTA)

Gel

Aging washing

(kerosene) ammonia



Thermal treatments

Drying

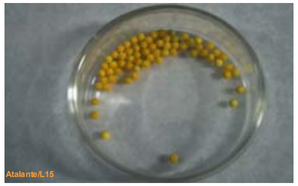
Calcining sintering

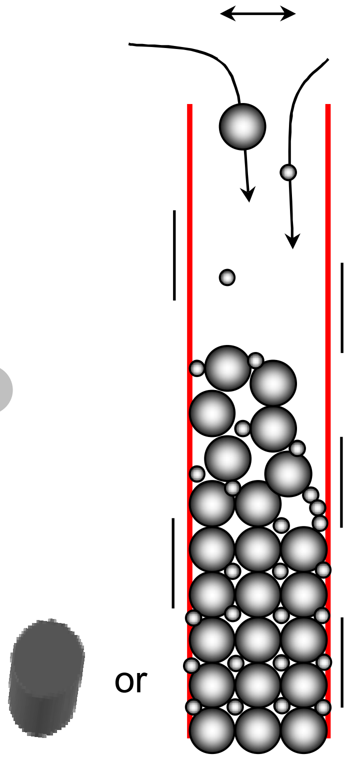
Inert >1500°C

Inert/H_2 >1500°C

Carbide

Oxide

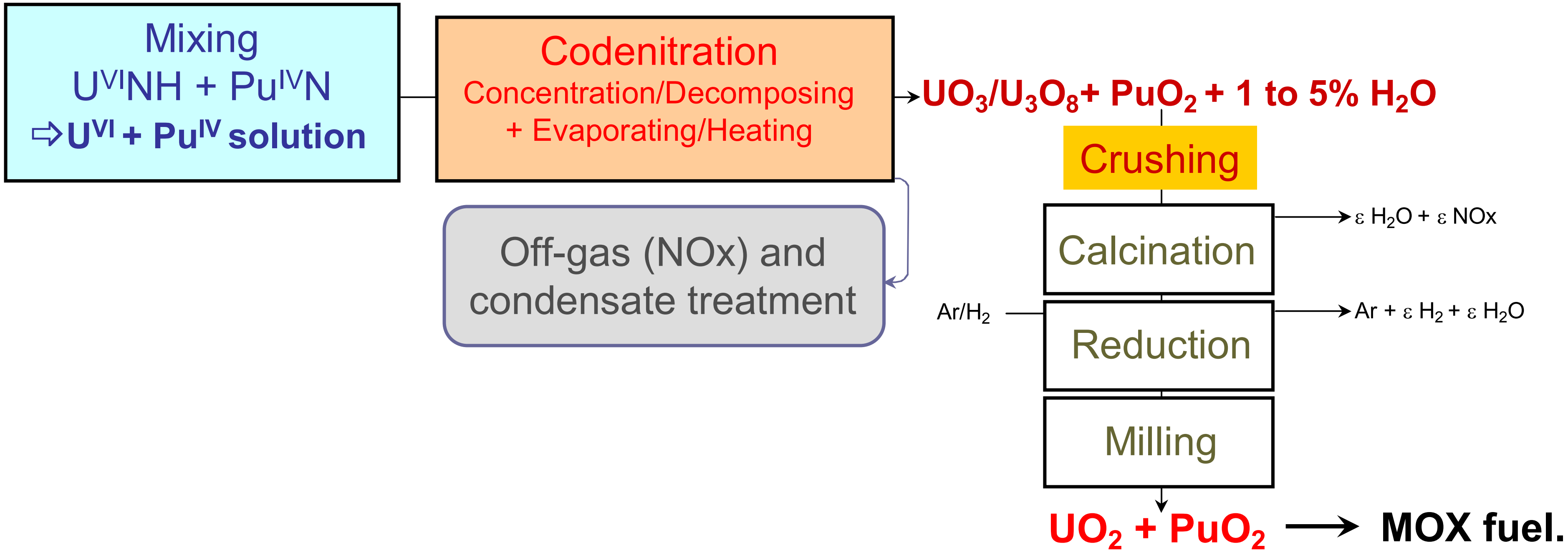




- **Few applications for only U+Pu processing within a LWR and/or Na-FR fuel cycle (relative complexity of the process).**

Methods of co-converting actinides into oxide

- Thermal codenitration
  - e.g. co-denitration of uranium and plutonium by MicroWave heating (Japanese JAEA technology)



Mixing
$U^{VI}NH + Pu^{IV}N$
⇨ **$U^{VI} + Pu^{IV}$ solution**

Codenitration
Concentration/Decomposing
+ Evaporating/Heating

Off-gas (NOx) and condensate treatment

$UO_3/U_3O_8 + PuO_2$ + 1 to 5% H_2O

Crushing

Calcination → $\varepsilon\ H_2O + \varepsilon\ NOx$

Ar/H_2 → Reduction → $Ar + \varepsilon\ H_2 + \varepsilon\ H_2O$

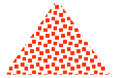
Milling

$UO_2 + PuO_2$ → MOX fuel.

  - Recent industrialization in the Rokkasho Mura plant
  - U and Pu still separated (no solid solution)

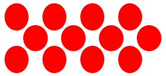
Methods of coconverting actinides into oxide

- Past and present work involving MAs
  - Main process developments (1965-2005)



➢ **Precipitation**

- oxalic mainly, but no work on other coprecipitation methods

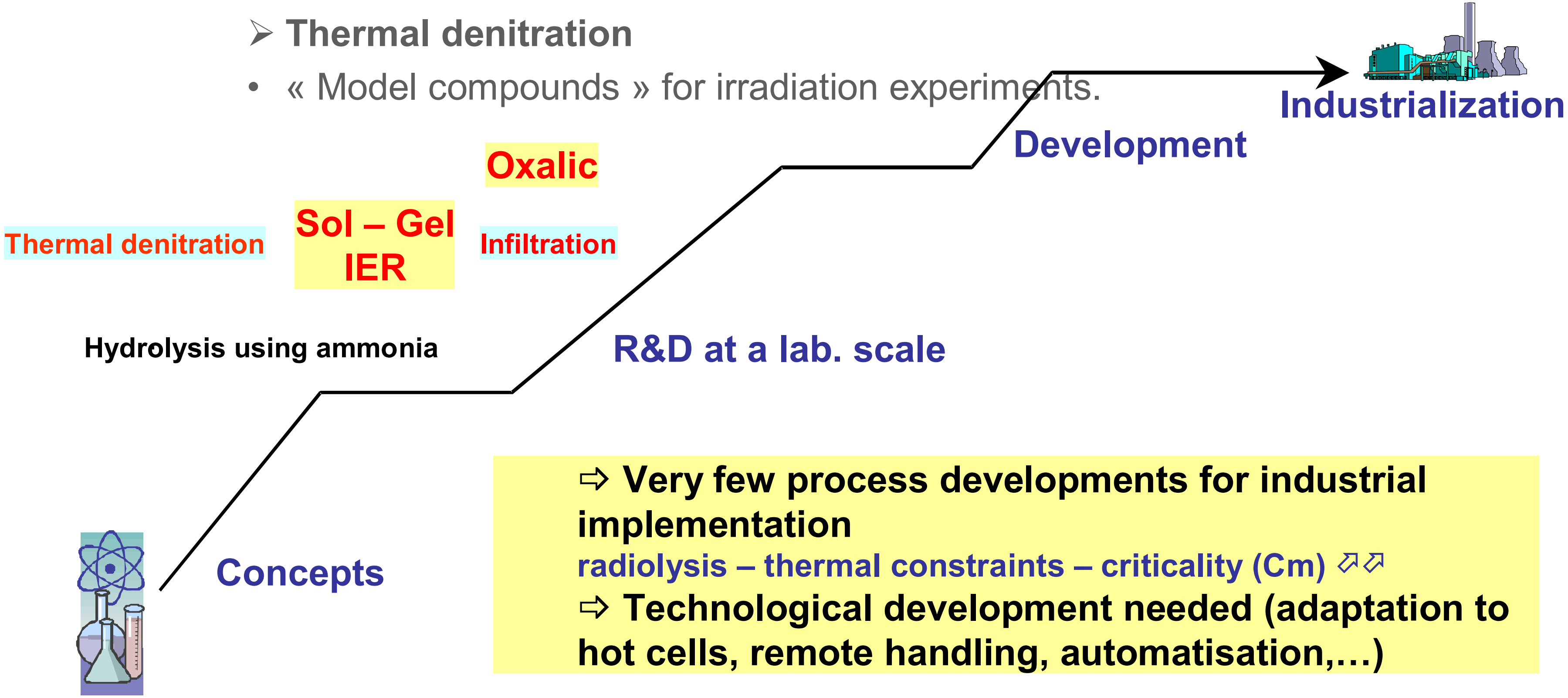


➢ **Sol-gel (or analog methods)**

- « Model compounds » for irradiation experiments (not focused on process)

➢ **Thermal denitration**

- « Model compounds » for irradiation experiments.



⇨ **Very few process developments for industrial implementation**

radiolysis – thermal constraints – criticality (Cm) ⬀⬀

⇨ **Technological development needed (adaptation to hot cells, remote handling, automatisation,…)**

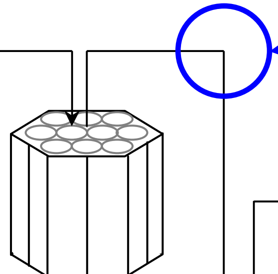
Focus on the oxalic (co)conversion process

- Oxalic coprecipitation
  - **From PUREX to COEX™**

CEA-ATALANTE

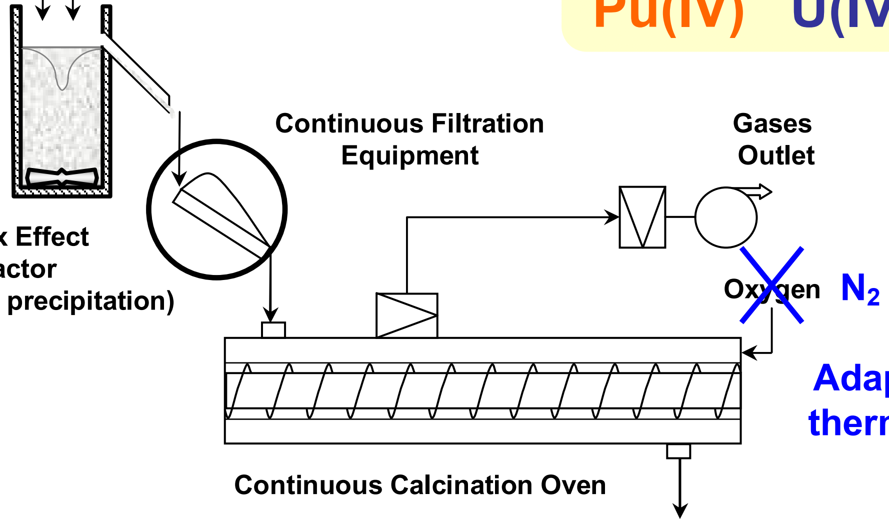
Focus on the oxalic (co)conversion process

- **Conversion PUREX ⇔ Coconversion COEX™** ⇔
  - **Adaptation of the chemistry of mixed U-Pu oxalate/oxide compounds to known technologies used industrially**



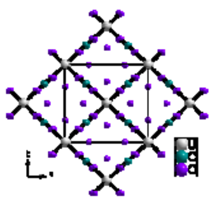
⇔

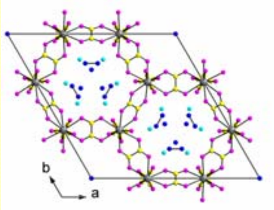
Pu(IV) **U(IV)+Pu(III)**

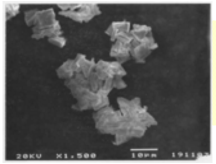


⇔

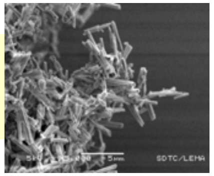
Pu(IV) Oxalate **U(IV)+Pu(III) Oxalate**





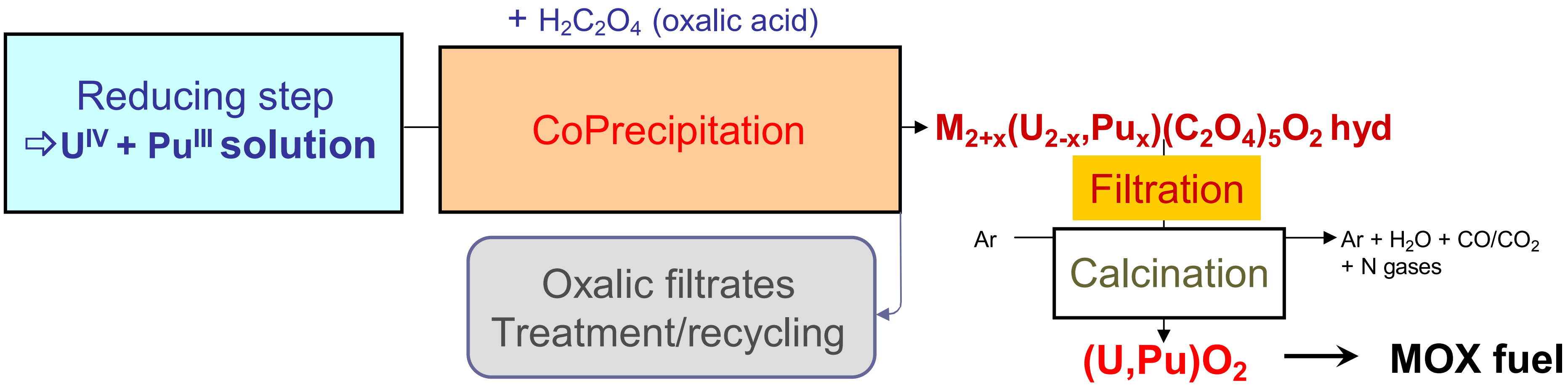


PuO_2 ⇔ $(U,Pu)O_2$



Focus on the oxalic (co)conversion process

- Reference method developed by the CEA with AREVA NC
  - Coconversion of uranium and plutonium *via* an oxalic co-precipitation



+ $H_2C_2O_4$ (oxalic acid)

Reducing step
⇨ **U^{IV} + Pu^{III} solution**

CoPrecipitation

$M_{2+x}(U_{2-x},Pu_x)(C_2O_4)_5O_2$ hyd

Filtration

Ar

Calcination

Ar + H_2O + CO/CO_2 + N gases

Oxalic filtrates Treatment/recycling

$(U,Pu)O_2$ ⟶ **MOX fuel**

- Scientific and technical advantages (GenIII+/GenIV):

⇨ Simplicity (equivalent to Pu(IV) oxalic conversion), continuous operation.

⇨ Pu distribution homogeneity (oxalate and oxide U,Pu solid solutions).

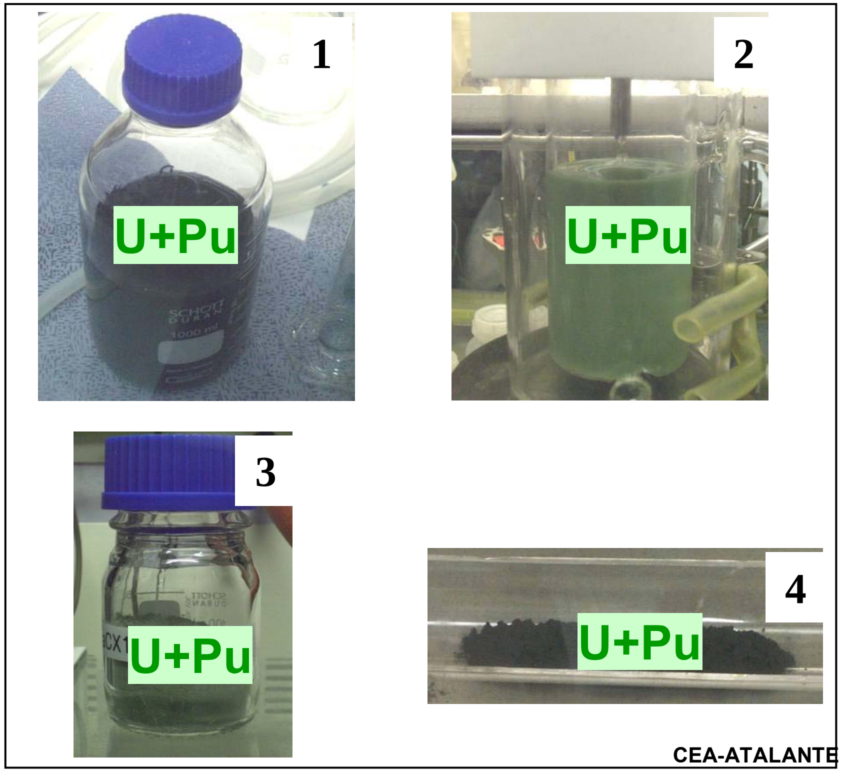
⇨ Supplementary decontamination factor (↗ refining during precipitation).

⇨ O/M ~ 2 without use of H_2.

⇨ Flowability of the powder, specific surface area ~ 3 - 4 m^2/g…

Focus on the oxalic (co)conversion process

- Oxalic coprecipitation
  - Homogeneity and relative cocentrations of U and Pu



CEA-ATALANTE

Oxalic co-conversion of U(IV)-Pu(III) into (U,Pu)O_2 performed at the CEA's Atalante complex.

1: Initial mixture in solution
2: Co-precipitation in a vortex effect reactor
3: Mixed U(IV)-Pu(III) co-precipitate,
4: (U,Pu)O_2 oxide solid solution.

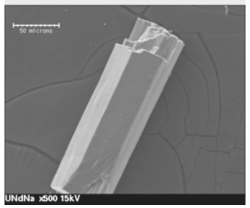
⇨ The plutonium is intimately mixed with uranium, from the co-partitioning step, in solution, up to co-conversion product, a starting material for MOX fabrication.

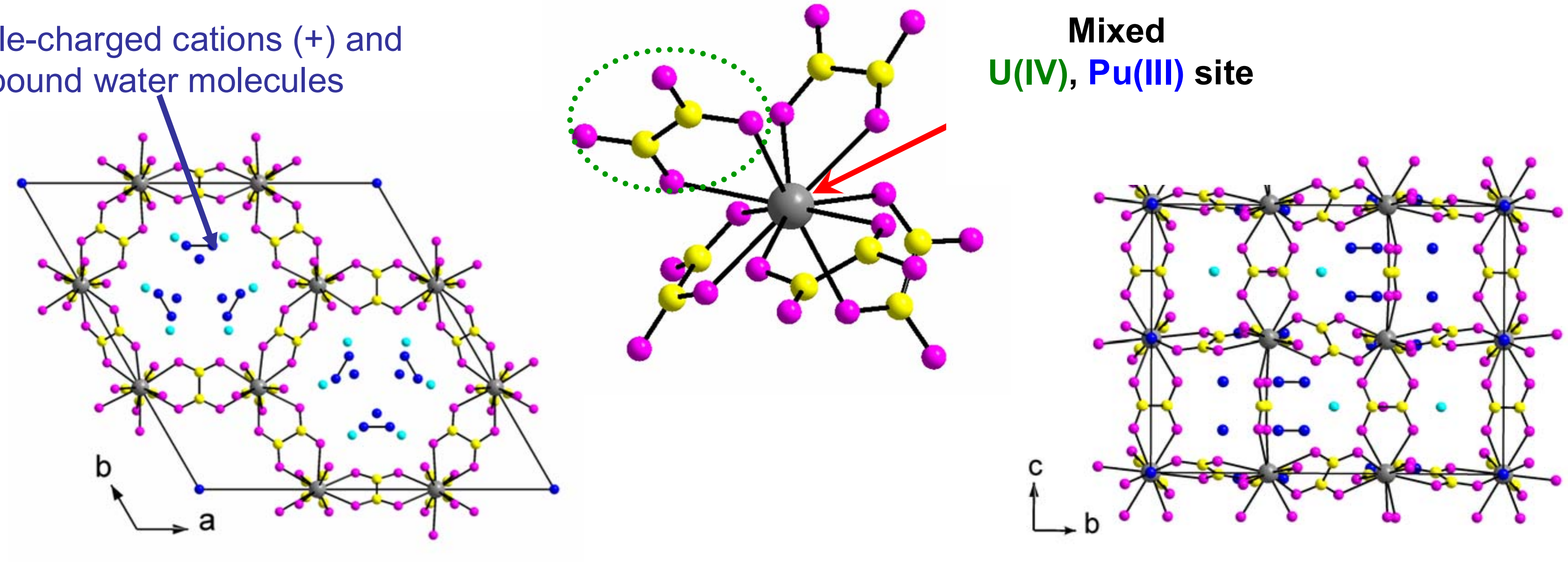
Focus on the oxalic (co)conversion process

- Coprecipitation: structural data
  - Unprecedented mixed U(IV)-Pu(III) oxalate structures with a solid solution domain: $(0 < x \leq 1)$

Crystalline structure: hexagonal space group $P6_3/mmc$

$(M^+)_{2+x}U^{IV}_{2-x}Pu^{III}_x(C_2O_4)_5.nH_2O$





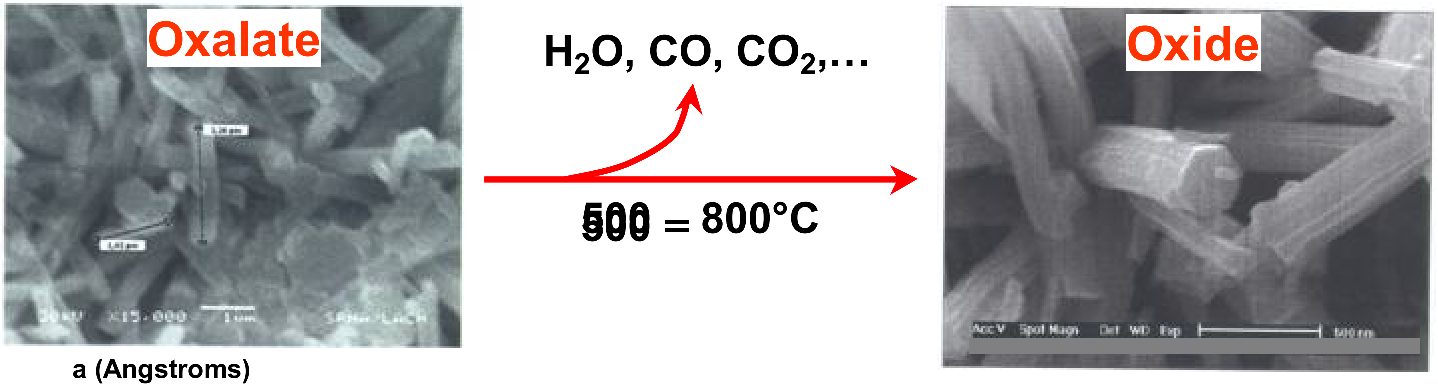
Single-charged cations (+) and bound water molecules

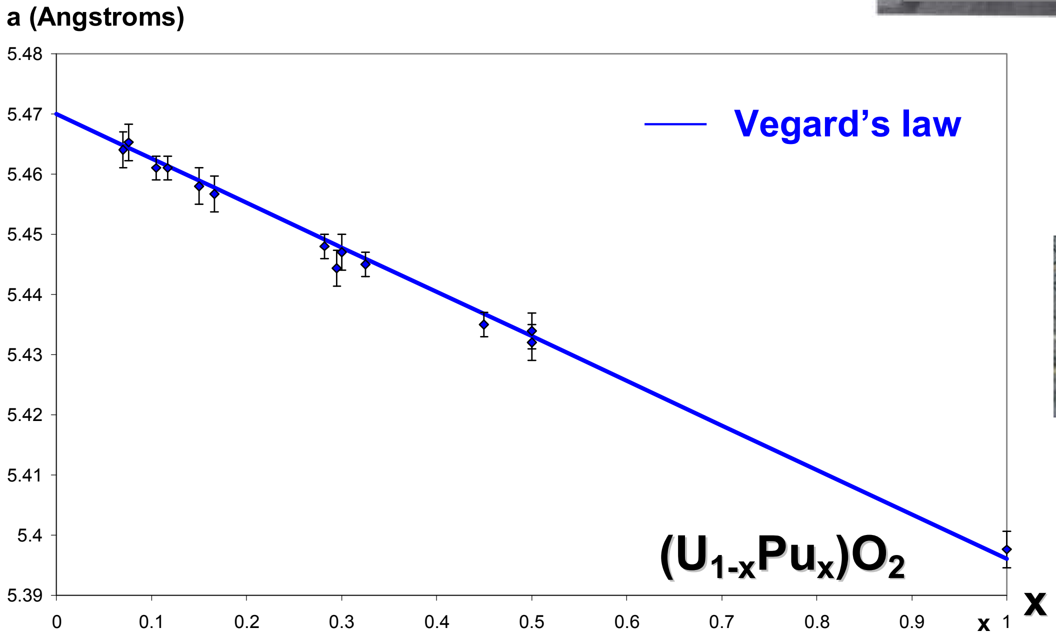
Mixed U(IV), Pu(III) site

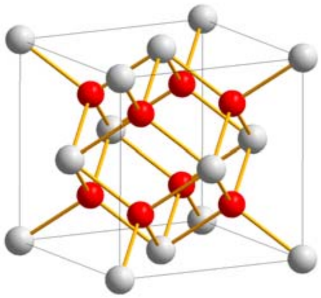
⇨ homogeneous mixing of U(IV) and Pu(III) at molecular scale.
⇨ homogeneous composition of the powder (single-phased co-precipitate).

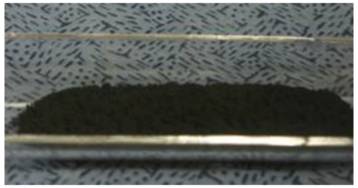
Focus on the oxalic (co)conversion process

- Thermal treatment (calcination): preserving the homogeneity
- $(M^+)_{2+x}U^{IV}_{2-x}Pu^{III}_x(C_2O_4)_5.nH_2O$ $(U_{1-x},Pu_x)O_2$









Solid solutions (e.g. $(U,Pu)O_2$), for mol % Pu between 0 and 50

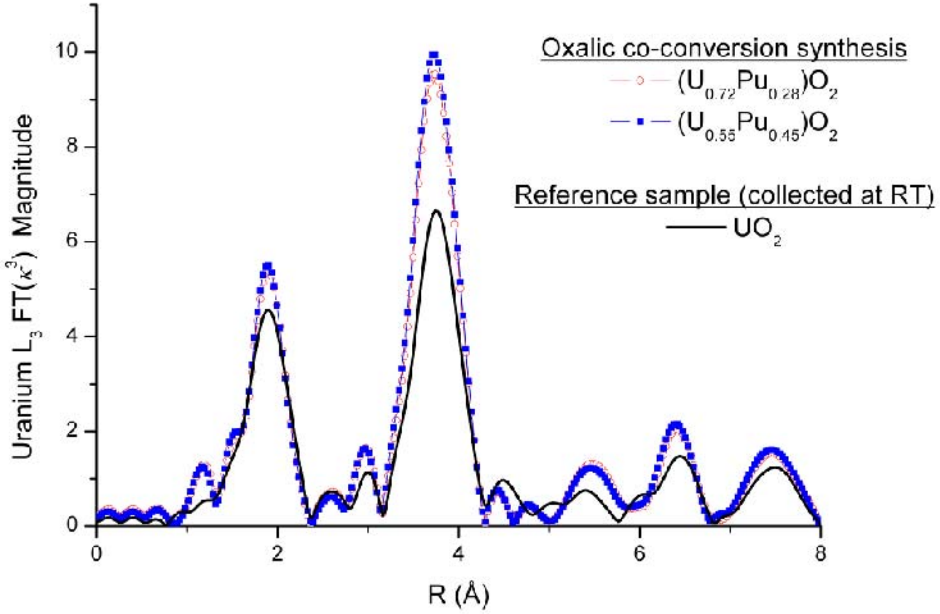
Focus on the oxalic (co)conversion process

- Calcination : structural data
  - XAS analysis of $(U,Pu)O_2$ obtained after oxalic co-conversion

$(T_{co\text{-}conversion} < T_{sintering})$ ROBL Beamline ESRF in coll. with Fuel Study Division – Cadarache, Ph. Martin

EXAFS U environment

Fourier Tranform at the Pu L_{III}edge.



Ph. Martin, S. Grandjean, C. Valot, G. Carlot, M. Ripert, P. Blanc, C. Hennig, “XAS study of $(U_{1-x}Pu_x)O_2$ solid solutions”, Journal of Alloys and Compounds, 444-445, 410-414, (2007).

Stoechiometric ideal solid solutions $(U_{1-x}Pu_x)O_2$

$0 < x \leq 0.5$

Focus on the methods developed in ATALANTE for MAs management

- **New challenges with MAs:**
  - High Activity
    - Am (alpha + gamma) and especially Cm (alpha + neutron)

  ⇨ Dilution (mixing with another element - no ~~AmO_{2-x}~~ or ~~CmO_{2-x}~~ handling - and homogeneity of the mixing in the solid phases), dustfree solids, robust process (remote operation + automation), innovative micro- and macrostructures of the fuel precursor.
  - Heat and Radiolysis Effects
    - Especially Cm

  ⇨ Simple and robust process, simple reagents (radiolysis, waste, by-product treatment and recycling), dilution (mixing with diluent)
  - Criticality
    - Example : ^{245}Cm

  ⇨ Dilution (mixing with diluent), controlled homogeneity of the mixing in solution and in the solid phases (no segregation)
  - MAs Chemistry ≠ Th,U or Pu Chemistry
    - Example : moderate reactivity of An(III) and An(V) as regards hydrolysis (≠ Th, U(VI) and Pu(IV))

  ⇨ Prior analytical chemistry in solution + solid chemistry with actinides, complete databases (phase diagrams)

Focus on the methods developed in ATALANTE for MAs management

- Oxalic coprecipitation with MAs **(e.g. in a vortex reactor)**
  - Evolutionary recycling of actinides

Oxalic precipitation of

Pu(IV)

(PUREX)

Oxalic co-precipitation of

U(IV) and Pu(III)

(COEX^{TM})

Oxalic co-precipitation of

U(IV) and/or Pu(III) + MAs(III,IV)

(transmutation in hom./het. mode)

U(IV)Pu(III)Np(IV)

U(IV)Pu(III)Np(IV)Am(III)

U(IV)M(III) (M=Am,Cm)

S. Grandjean, A. Beres, J. Rousselle C. Maillard, French patent n° FR/04 510058, (2004), European Patent, n° EP1756839(A1), (2007), International patent WO 2005/119699.

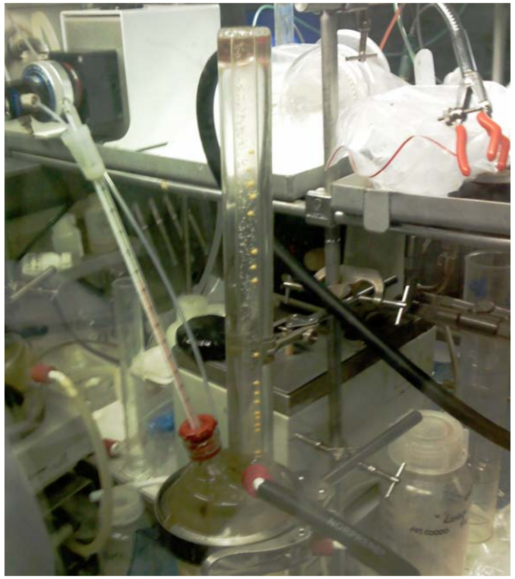
Focus on the methods developed in ATALANTE for MAs management

- Sol-gel (internal cogelation of U and TRUs)
  - Experiments performed in ATALANTE :
    - U(VI) and U(IV) from 2002
    - **U(VI)/Pu(IV)** and **U(IV)/Pu(III)**

⇨ **Few applications for only U+Pu processing within a LWR and/or Na-FR fuel cycle (relative complexity of the process *vs* other methods).**

⇨ Potential interest: Dustless fuel precursors containing MAs for FR.

⇨ recently optimized for U(VI)/Am(III) (90/10):



ATALANTE / L15

Focus on the methods developed in ATALANTE for MAs management

- Fixation of U(VI)-An(III) on IER and calcining (oxide, carbide)
  - Experiments performed in ATALANTE:
    - U(VI), M(III) – from 2003
    - **Microspheres of U(VI)-An(III) oxides**

⇨ **Few applications for only U+Pu processing within a LWR and/or Na-FR fuel cycle (relative complexity of the process *vs* other methods).**

⇨ Potential interest: Dustless fuel precursors containing MAs for FR.

⇨ recently optimized for U(VI)/Am(III) 90/10:

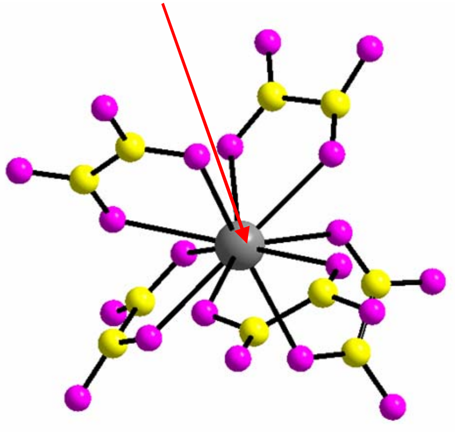
« Preparation of minor actinides targets or blankets by the mean of IER »
Sébastien Picart, Hamid Mokhtari, Isabelle Ramière, Isabelle. Jobelin, Global 2009

See:

Focus on the methods developed in ATALANTE for MAs management

- Controlling the composition and the homogeneity
  - Mixed oxalate An(IV)-An(III) structure

Mixed An(IV)–An(III) crystallographic site



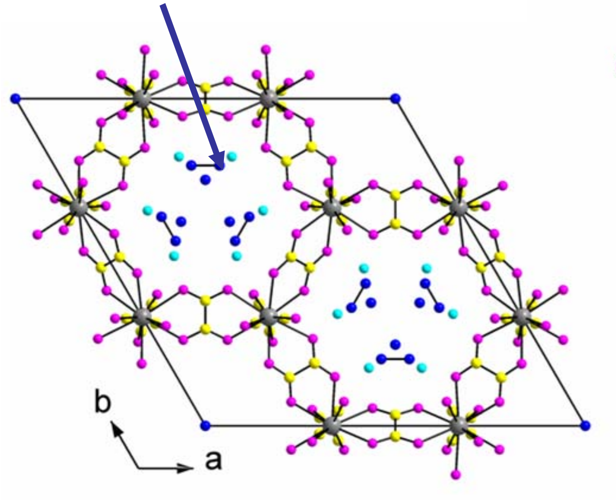
Hexagonal crystalline structure

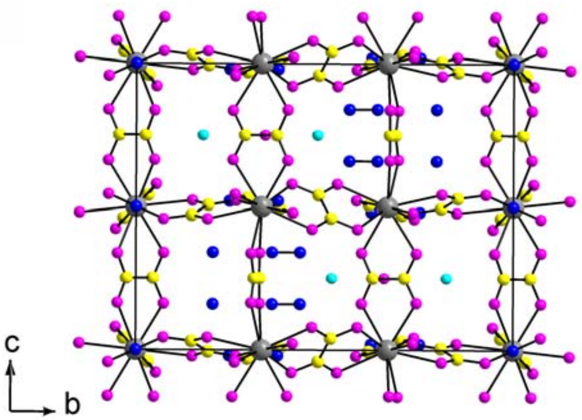
$(M^+)_{2+x}An^{IV}_{2-x}An^{III}_x(C_2O_4)_5.nH_2O$

An = U or Np, An = Am or Cm

M = monocharged cation in the second coordination shell

Monocharged cations (+) and non bonded water molecules





⇨ The oxalate An(IV)-An(III) mixing in the coprecipitate is homogenous at a molecular scale (solid solution from An(III)/(U+An(III)) = 0 to 50% at/at).

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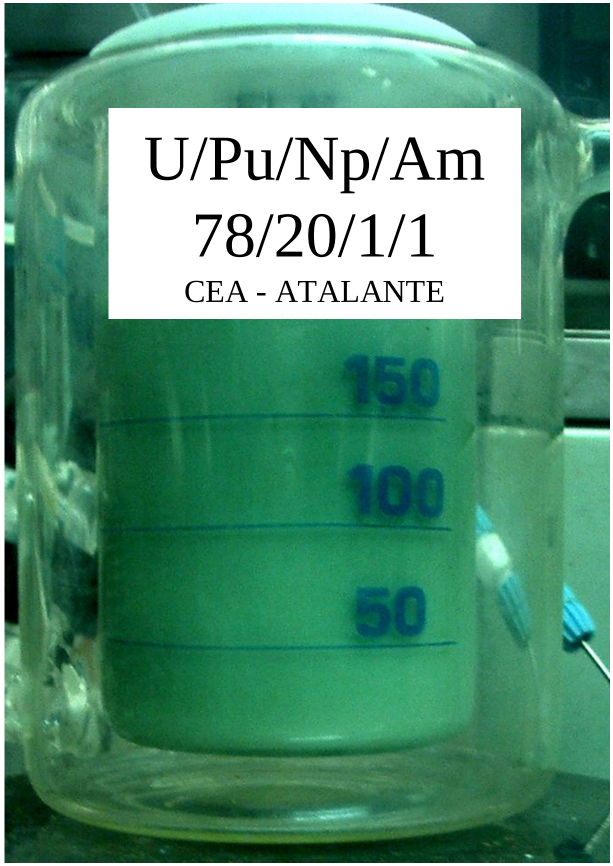
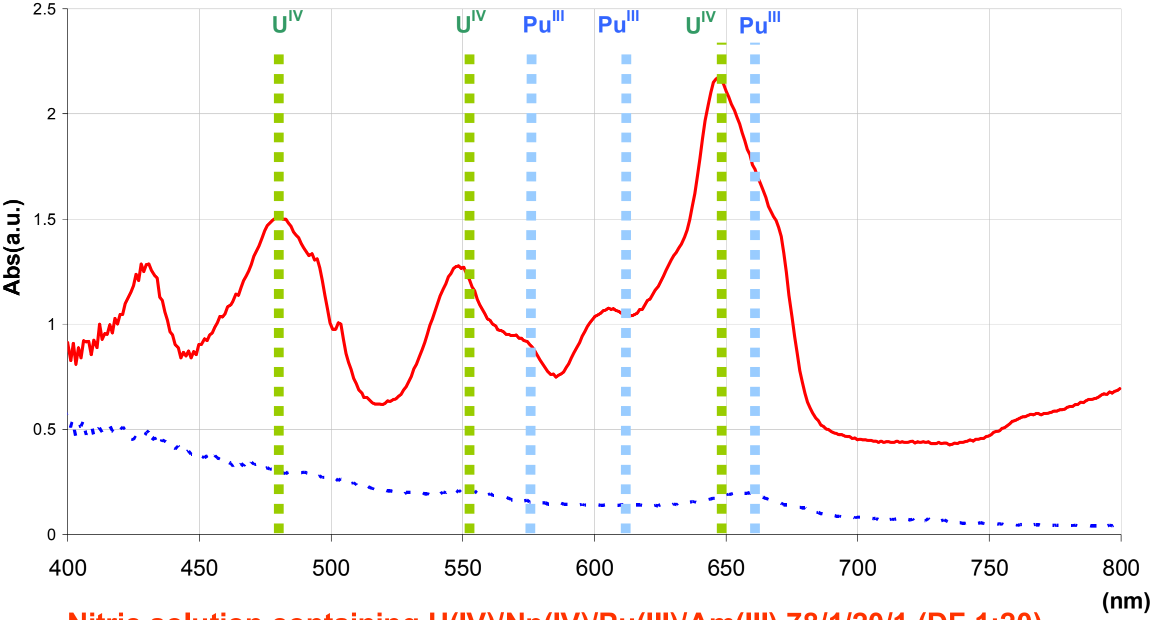
Focus on the methods developed in ATALANTE for MAs management

- Oxalic co-precipitation:
  - U +Pu + MAs co-precipitation (⇨ transmutation : homogeneous)



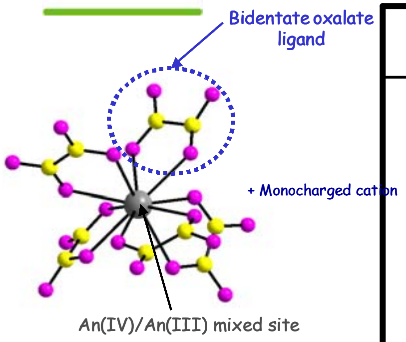


Nitric solution containing U(IV)/Np(IV)/Pu(III)/Am(III) 78/1/20/1 (DF 1:20)

Nitric supernatant after co-precipitating the latter actinides by $H_2C_2O_4$ (DF 1:1)

Focus on the methods developed in ATALANTE for MAs management

- Oxalic co-precipitation:
  - U +Pu + MAs co-conversion (⇨ transmutation : homogeneous)

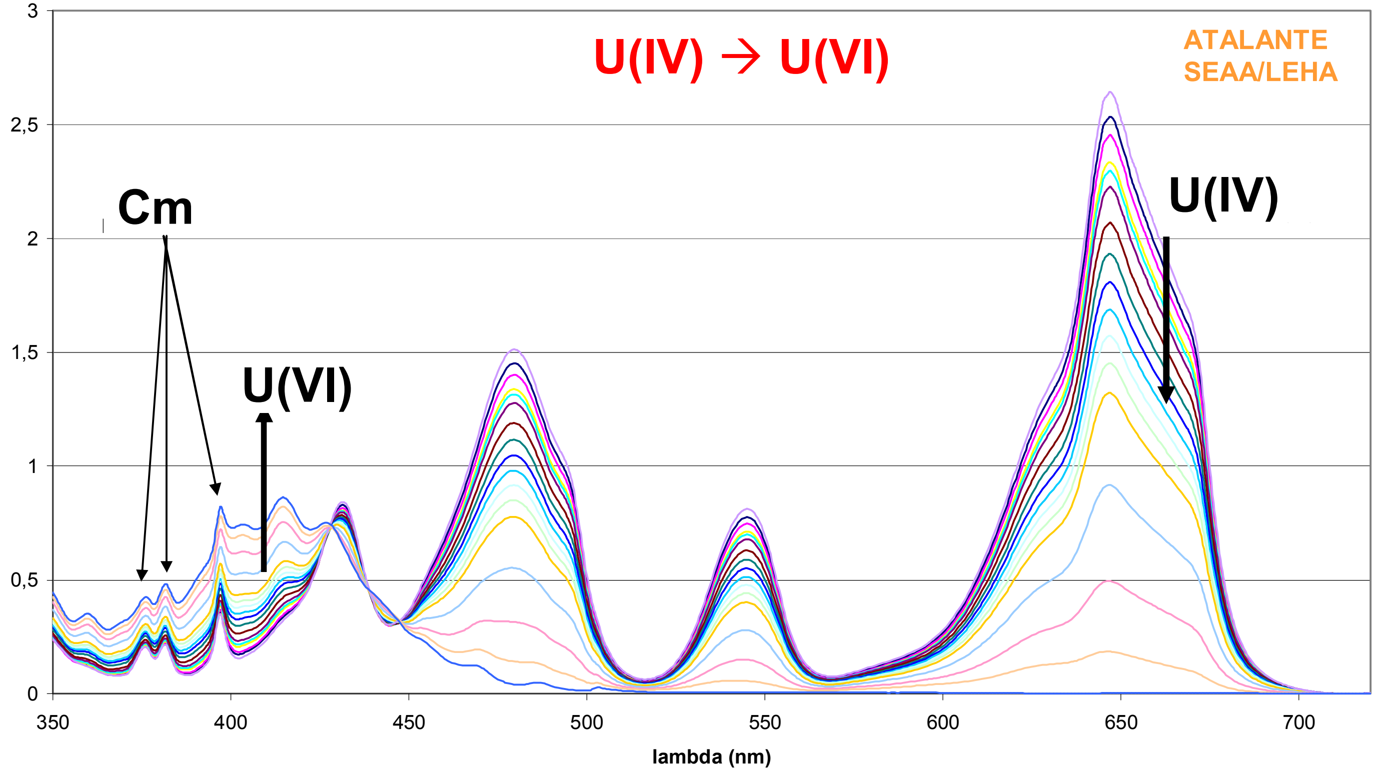


Intermediate co-precipitation step	
U(IV)-Np(IV)-Pu(III)-Am(III) oxalate co-precipitate 78/1/20/1	Single-phase **Crystal system : hexagonal** **Bravais lattice : P** **a(Å) = 19.112(5) ; c(Å) = 12.690(5)**
Co-conversion product	
$(U,Np,Pu,Am)O_2$ 78/1/20/1	Solid solution : **Crystal system : cubic** **Bravais lattice : F centered** **a(Å) = 5.454(3)** Specific area (BET): **7 ± 1 m^2/g** Mean particle diameter: **10 ± 1 µm**

- **⇨ Homogeneous mixing of the involved actinides in the solid phases (ΣAns mixed oxalate and ΣAns mixed oxide).**

Focus on the methods developed in ATALANTE for MAs management

- Mitigating the effects of radiolysis and thermal constraints
  - Example: impact of heavy α-emitters
    - ➢ Before co-conversion : **stability study of a U(IV)-Cm(III) (90/10) solution**



⇨ **Implies to anticipate a limited stability of the solution with strong α emitters. e.g. by on line mixing of U(IV) and MA(III).**

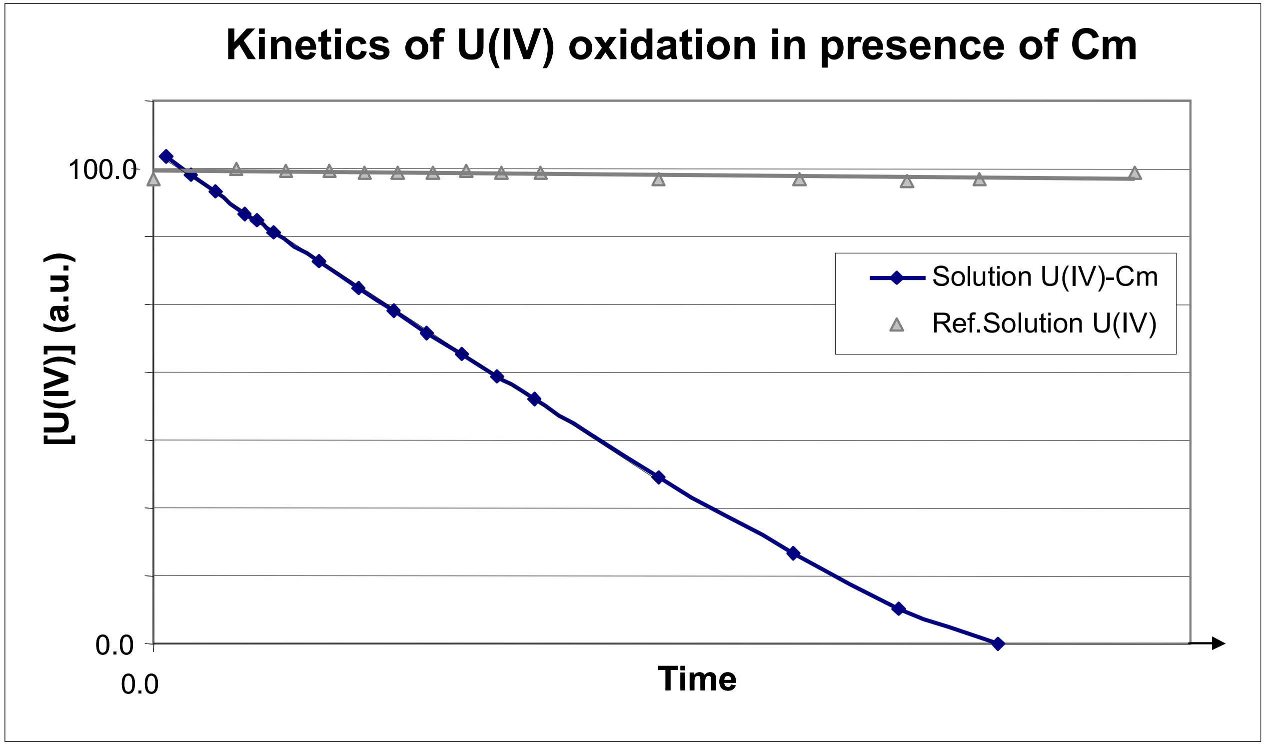
Robustness of the oxalic process related to residual U(VI) presence was assessed.

Focus on the methods developed in ATALANTE for MAs management

- Mitigating the effects of radiolysis and thermal constraints
  - U + MAs co-precipitation (⇨ transmutation : heterogeneous)

    Higher MAs contents in the solid phase (e.g. MAs > 10 mol% HM)

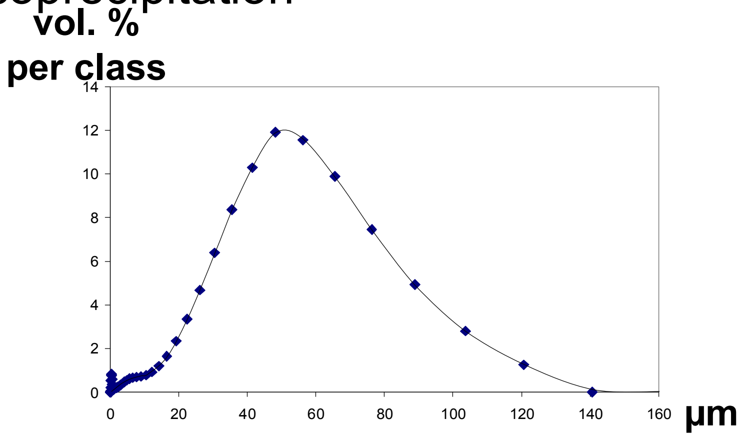
    ↗ Heat and ↗ Radiolysis effects ⇨ Process optimizations

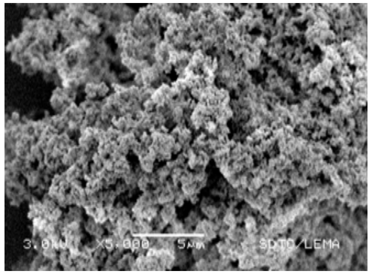


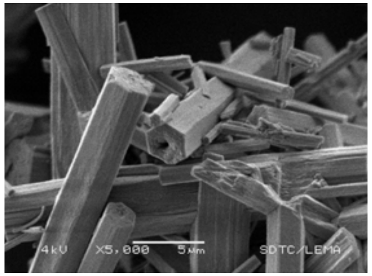
⇨ **Process+Technology development in hot cells, at laboratory then pilot scale, (still) required before industrial implementation.**

Focus on the methods developed in ATALANTE for MAs management

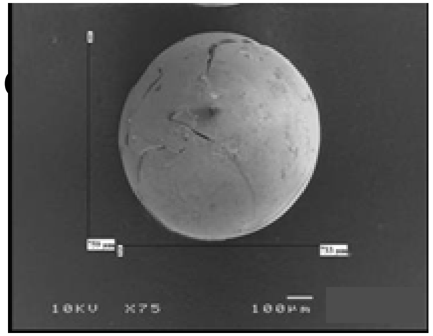
- Checking the suitability of the end-product as starting material for fuel manufacturing (e.g. oxalic co-conversion end-product)
  - Oxalic coprecipitation







  - Sol-gel



⇨ **MA-coconversion product ⇔ Fuel fabrication processes.**

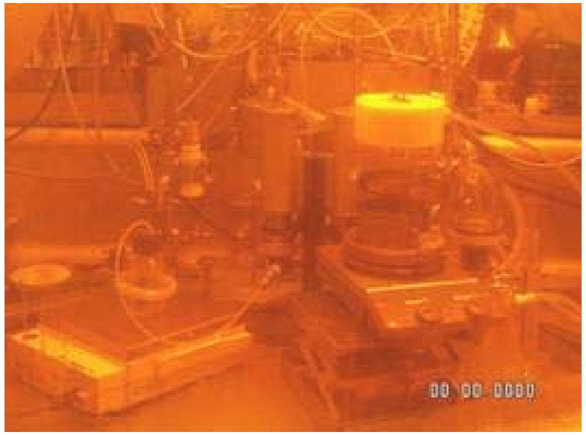
Focus on the methods developed in ATALANTE for MAs management

- Implementing the process/technologies in hot cells
  - 1^{st} applications of oxalic An(IV)-An(III) coprecipitation for the synthesis of mixed actinides oxide solid solutions in hot cells
    - Pu(IV)-Am(III) coconversion into oxide (**ATALANTE**) (FUTURIX)

      $Pu_{1-x}\mathbf{Am}_xO_2$ (x = 0,2 – 0,5 – 0,8)

      L. Donnet, F. Jorion, C. Leorier, N. Drin, « The FUTURIX-FTA Experiment in PHENIX. Status of the oxide fuels and pins fabrication », GLOBAL 2007 Advanced Fuel Cycles and Systems, Boise (US – Idaho), Sept. 9-14, 2007.

ATALANTE LEMA



- U(IV)-Cm(III) coconversion into oxide (**ATALANTE**)

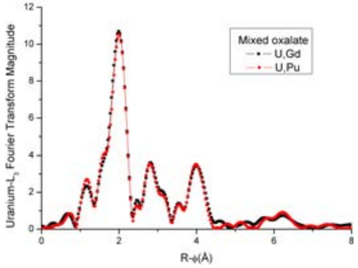
  $U_{1-x}\mathbf{Cm}_xO_2$ (x = 0,10)

  Cm content of 10% of mass U+Cm in order to exacerbate the thermal and radiolytic effects.

  S. Grandjean, N. Herlet, J-Ph Dancausse, « Results on U(IV)/An(III) co-precipitation studies for MA bearing solid solution synthesis », 10th IEMPT, Mito (Japan), 6-10 October, 2008.

⇨ Implementation of optimized coconversion processes at MA multigram-scale in progress, e.g. for technological assessment

Future R and D on conversion



- Further R and D in CEA/Atalante
  - Oxalic coprecipitation
    - Basic studies:
      - investigations by XAS of oxalate and oxide solid solutions,
      - syntheses involving less-investigated Np compounds.
    - Process development:
      - adapting the microstructures of the fuel precursor,
      - assessing the robustness with MAs (thermal, radiolysis constraints),
      - adapting the technology to hot cells environment.
    - (U,Pu,MAs) oxide powder production for in pile test fuels:
      - new production unit at MAs multigram scale (ALFA: Atalante Laboratory For Actinides Bearing Fuel Manufacturing), and at pin scale.
  - Other methods* (Sol-gel, IER…)
    - Basic studies
    - Process development (less experience with MAs):
      - simplifying the successive steps for continuous and safe processing,
      - characterizing and adapting the microstructures of the fuel precursor,
      - assessing the robustness with MAs (thermal, radiolysis constraints),
      - developing the technology in hot cells environment.

* collaborative work (FP'7 ACSEPT, …)

What characterize the MA bearing fuels

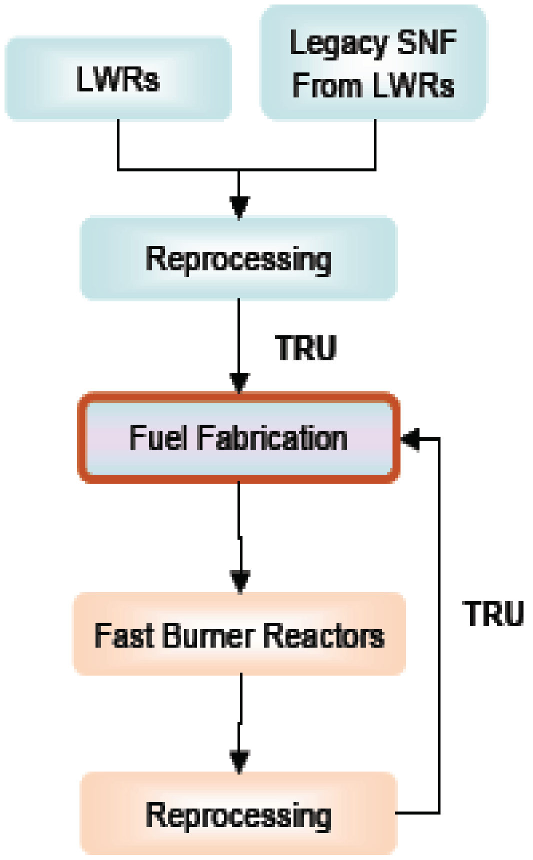
Transmutation fuel development is considerably more challenging than conventional fuels :

- Multiple elements in the fuel

  U, Pu, Np, Am, Cm
- Varying thermodynamic properties

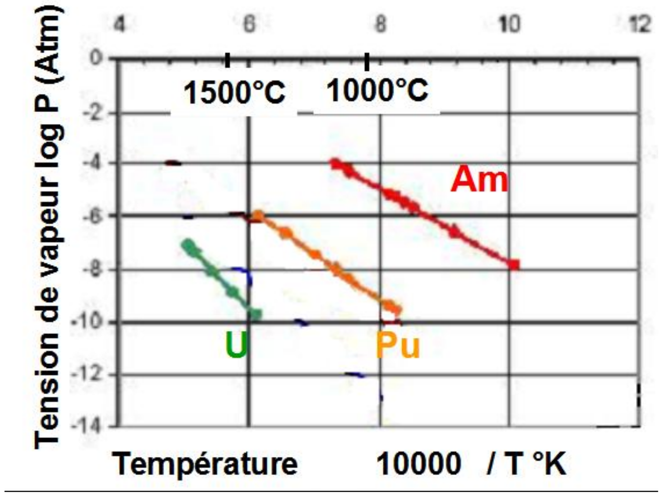
  e.g. High vapor pressure of Am
- Impurities from separation process

  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 MOX fuels

- Thermal, physico-chemical properties MA-$UPuO_2$ / $UPuO_2$
  - Melting point : lower
  - Thermal conductivity : lower
  - Volatility : higher
  - Oxygen potential : higher



- Thermal and radiological constraints
  - Helium production : higher
  - Neutron emissivity : higher
  - Thermal power : higher
    - Pth MA blanket = ~10 x Pth LWR MOX
    - Neutron emission blanket = ~2000 x LWR MOX

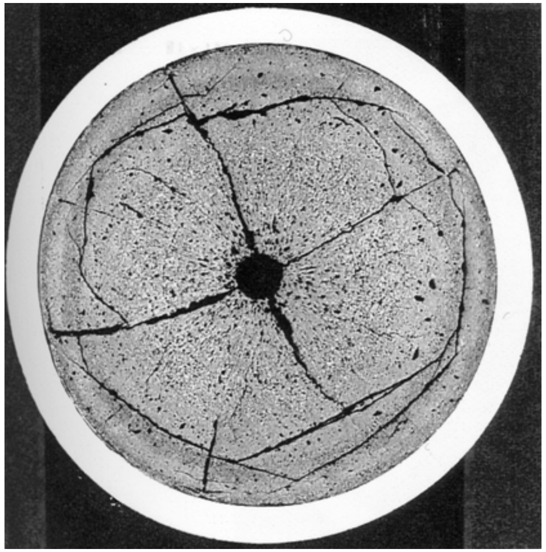
⇒ MA recycling impacts :

In-core fuel behavior

Fabrication conditions

Homogeneous recycling mode in SFR : acquired knowledge

- SUPERFACT homogeneous in Phénix, **1986 to 1988**
  - $U_{0.74}Pu_{0.24}Am_{0.02}O_2$ and $U_{0.74}Pu_{0.24Np}Np_{0.02}O_2$
  - **382 efpd, P_{LIN} = 380 to 325 W/cm**
  - **Burn up = 6.7 at%**



- Fuel restructuration is similar than for $UPuO_2$

- U, Pu, Am and Np radial distributions are very flat

→ No actinide specific redistributions

- Transmutation ratio at the reactor middle plane :

≈ 28 % for 241Am and ≈ 30 % for 237Np

- This satisfactory behavior has also been confirmed with the TRABANT experiment (annular pellets) in the HFR under thermal flux

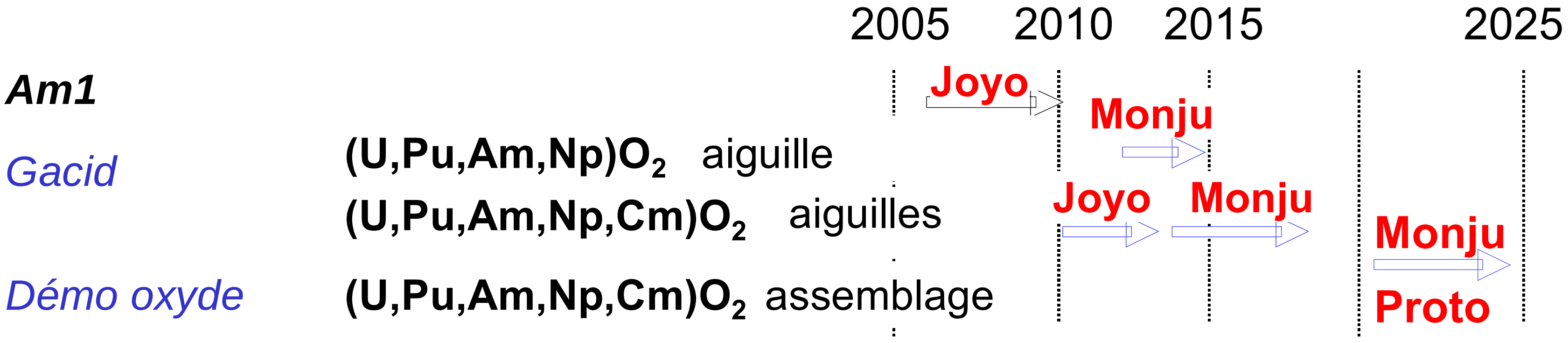
⇒ For low linear power, no real influence of the low MA amount, up to a BU equal to 6.7 at%, except for the He release of the Am fuel (4 times greater than standard UPuO2 pins release)

Homogeneous recycling mode in SFR : perspectives

- **Main goals for MA SFR fuel development : behaviour under irradiation**
  - enhanced objective burn up (fission gas release, swelling,…)
  - optimisation of clad material : ODS steel up to 160 dpa
  - still enhanced safety

⇒ Necessity of irradiation tests :

- **French Phénix shut down is in 2009**
- **Russian BOR60 under important renewal works,…**
- **The collaboration framework GACID between DOE, CEA and JAEA is currently defined using Joyo and Monju in order to implement a common irradiation program, particularly on SFR fuels, pins and sub assembly, Am and Cm**



Am1

Gacid **(U,Pu,Am,Np)O_2** aiguille

(U,Pu,Am,Np,Cm)O_2 aiguilles

Démo oxyde **(U,Pu,Am,Np,Cm)O_2** assemblage

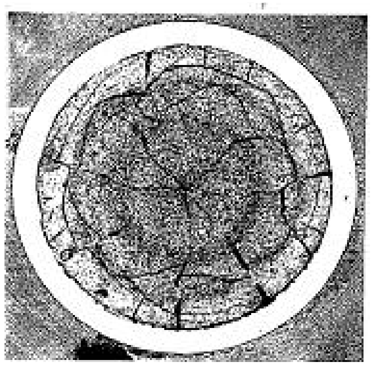
Heterogeneous recycling mode in SFR : acquired knowledge

- **Dilution of MA in UO_2 matrix, MA : 10-20 %, periphery of the core : "blanket"**

- **SUPERFACT heterogeneous in Phénix :**
  - **$U_{0.6}Am_{0.20}Np_{0.20}O_2$**
  - **382 efpd, P_{lin} = 174 to 273 W/cm**
  - **Burn up = 4.1 at%**

- **Limited fuel restructuring:**
  - **Fuel-clad interaction,**
  - **Cold fuel (no central hole),**
  - **High He retention for $UAmNpO_2$**

No central hole

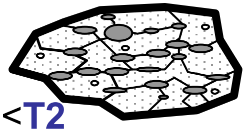


He Production

mm³/g fuel	Xe Kr	He
Standard $UPuO_2$ 6at%	1220	40
$UAmNpO_2$, 4at%	764	2970

Heterogeneous recycling mode in SFR : perspectives

- In a « cold » fuel, the **He behavior in the UO_2 matrix** has to be carefully studied (potential cladding failure under operation)



T1<T<T2

- *Potential significant fuel swelling*

⇒ **Conditions of swelling-release has to be known quantitatively , in order to design an heterogeneous MA transmutation pin :**

- On going experimental program (in the French Osiris thermal reactor, with UO2 matrix + Am concentrations, open/closed porosity, Cm?) in order to obtain experimental data in support of fuel design
- A second experimental phase for optimizing the microstructure
- Third phase : high burn up experiments of representative fuel elements under operating conditions in order to finally qualify the concept

Heterogeneous recycling mode in SFR : what inert matrix material ?

- Dilution of MA into inert material matrix, MA: 1.5 to 2.5 g/cm^3 = "target"

- Main issues to be studied :
  - Materials: actinides compound and inert matrix
  - Damages due to FP recoil
  - Limit excessive swelling due to He production

- Needed properties of the inert matrix material :
  - Limit the maximum temperature into the target
    - High Thermal conductivity after material damaging
    - Low swelling due to neutrons and FP
  - Allow high temperature
    - High melting point
  - No chemical interaction with actinide compounds, clad and coolant

First selection (bibliography, out of pile measurements):

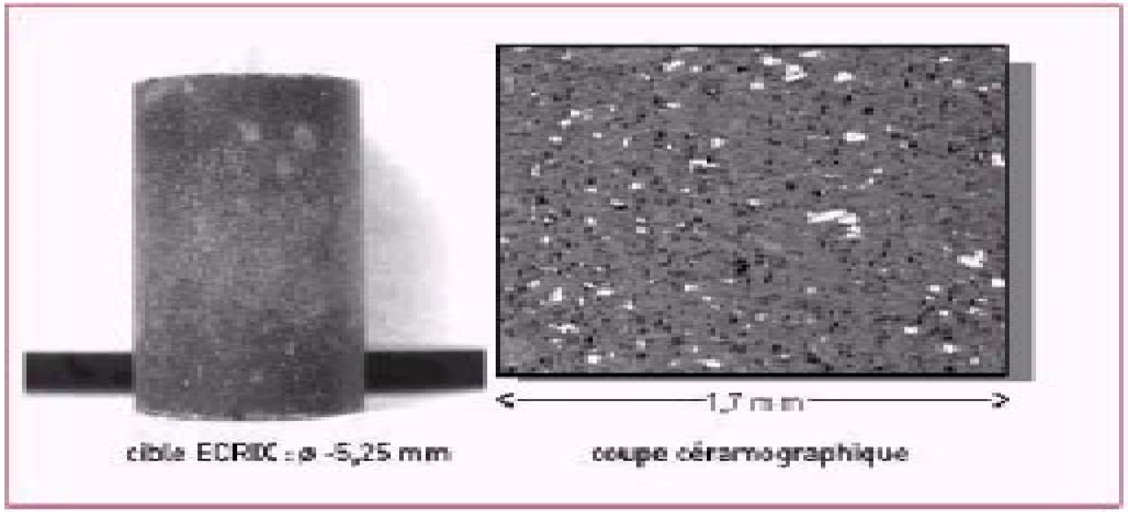
- compatible with Na: **MgO, Al_2O_3, $MgAl_2O_4$, $Y_3Al_5O_{12}$**
  ZrN, TiN, W, Nb, V, Cr, Mo

- compatible with water: **St-ZrO_2, CeO_2**

Heterogeneous recycling mode in SFR : what matrix material ?

- ECRIX irradiation
    - in Phénix, from March 2003 to March 2006
    - $AmO_{1.6}$ micro dispersed into MgO
    - Am = 0.7 $g.cm^{-3}$ (2.75 g of Am in 200 mm height column)
    - Objective Fission rate > 30 at% (90% transmutation rate)
    - non destructive PIE results indicate a satisfactory behavior of the target

- Results of on going complete PIE will allow to increase the performance of magnesia targets : Am amount, and transmutation rate



Heterogeneous recycling mode in SFR : what matrix material ?

- T4 bis irradiation
  - in HFR Petten, AmO_2 microdispersed in $MgAl_2O_4$
  - 171 GWd/m3, ΔV/V~28%, Am fission 72 %

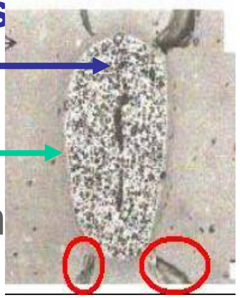
- MATINA 2-3 irradiation
  - in Phénix from July 2006, for 360 efpd (fast fluence:12 10^{26} m^{-2})
  - inert matrices MgO, $ZrYO_2$
  - MgO+UO_2 macro 100 μm or micro 1 μm particles (damage)
  - irradiation temperatures: 1000°C and 1400°C

- CAMIX-COCHIX irradiation
  - in Phénix from April 2007, for 240 efpd (fast fluence: 1.9 10^{26} m^{-2})
  - $AmZrYO_2$ solid solution, 2200 °C
  - MgO+ $AmZrYO_2$ with $AmZrYO_2$ 100 μm and 50 μm particles, 1400 °C
  - Objective Fission rate > 23 at%

- HELIOS irradiation
  - in HFR Petten, from 2008
  - MgO + $AmZr_2O_7$, with MgO matrix including a tailored open porosity to “facilitate” the He release

Heterogeneous recycling mode in SFR : perspectives

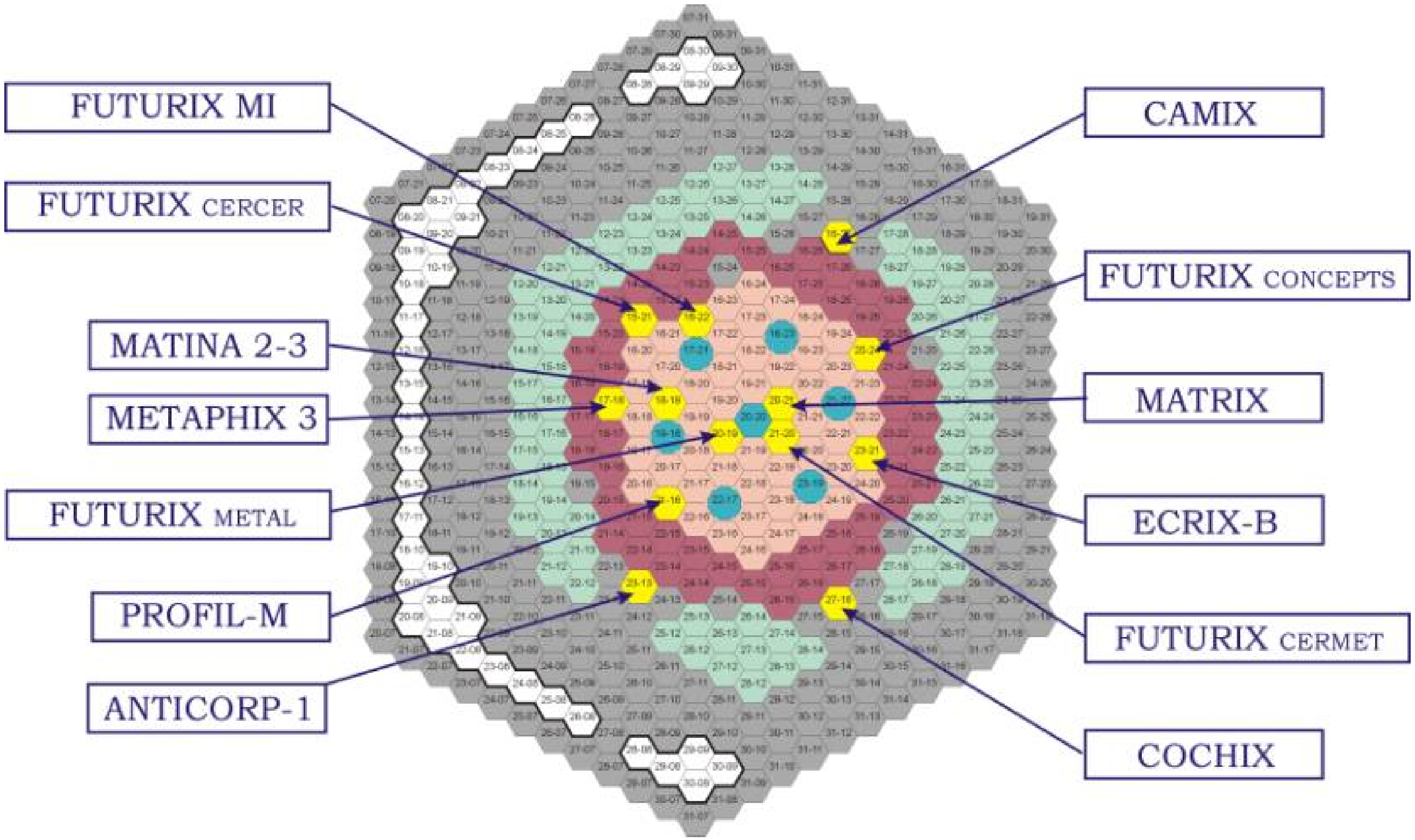
- The irradiation experiments lead to some **first conclusions**
  - **Composite swelling due to He retention**
  - Fissile compound-inert matrix interaction
  - For some matrices, matrix swelling (amorphisation



⇒ **Optimization of inert matrix targets as a function of the irradiation conditions : on going and future experiments, in order to still increase MA concentrations (MA from 0.7 to 4 g/cm3) and burn up**

⇒ **Curium… issue?**

The Phénix core on going transmutation experiments



Oxide versus metal, carbide or nitride ?

- Metal
  - a good option for SFR fuel, in the US
  - a large experimental program in the US National Labs
  - on going MA experiments in the Idaho ATR (UPuAmZr)
  - the on going Futurix experiment in Phénix (UPuAmZr and PuAmZr)

The choice must depend on

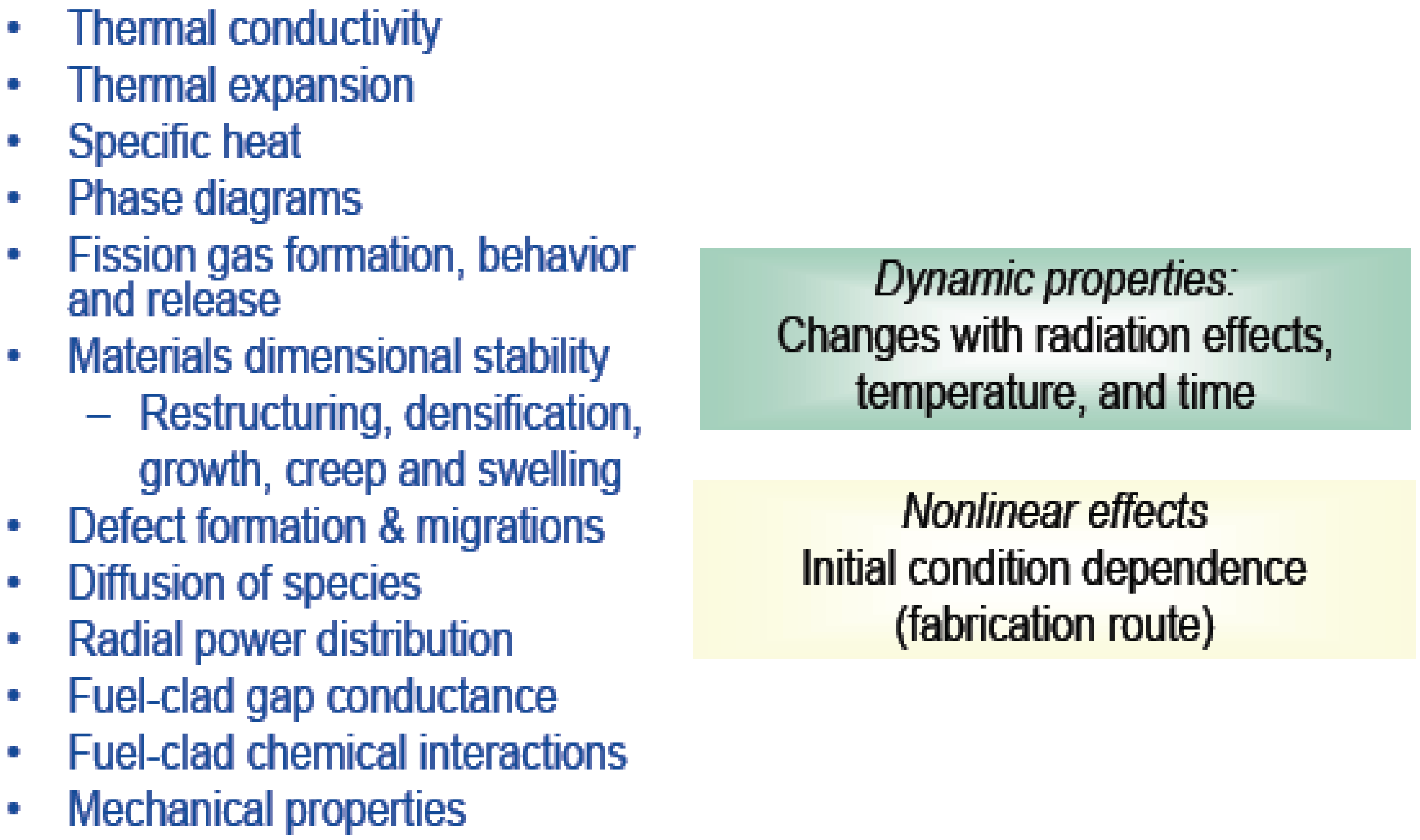
- Design and safety philosophy for the reactor
- Availability and cost of fabrication facilities
- Reprocessing and recycling strategy and technology for the driver fuel

- Carbide
  - a good option for SFR fuel, "almost" reference option for GFR fuel
  - France (limited) and India (large) experience on UPuC
  - no MA-UPuC experiment so far ?
  - pyrophoricity of divided material, fabrication and reprocessing in safe industrial conditions ?

- Nitride
  - a good experience in Japan, France and US
  - a complicated fabrication process, N15 enrichment
  - stability of Am nitride compounds as function of temperature ?
  - the on going Futurix experiment in Phénix (PuAmZrN and PuAmN)

(Transmutation) Fuel Modeling

Fuel performance prediction requires integral 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

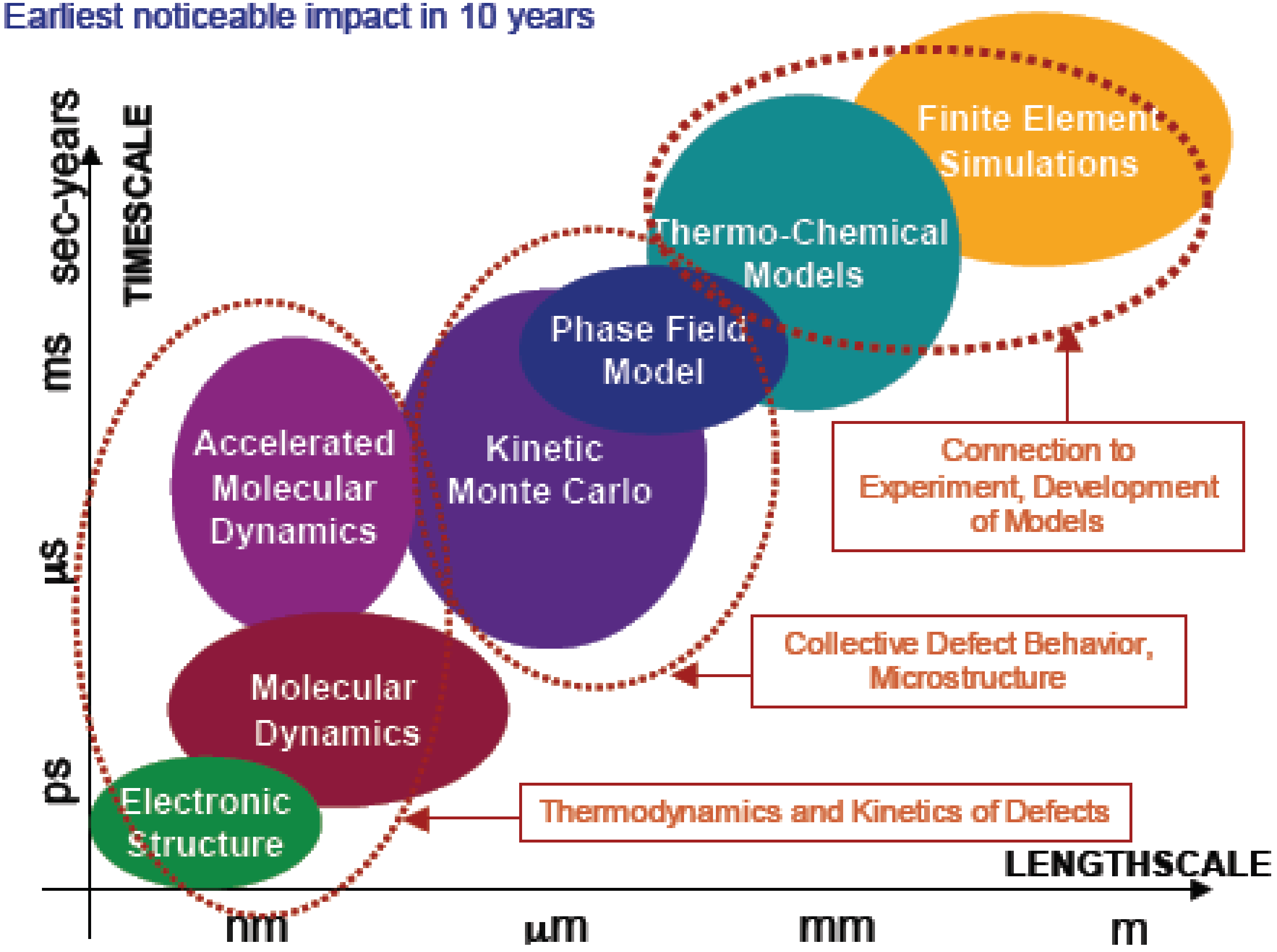
Dynamic properties:
Changes with radiation effects, temperature, and time

Nonlinear effects
Initial condition dependence (fabrication route)

(Transmutation) Fuel Modeling

Multi-scale modeling approach is being used to develop fuel performance suite of codes :

Incremental improvements in reducing the number of tests and qualification duration
Earliest noticeable impact in 10 years



Connection to Experiment, Development of Models

Collective Defect Behavior, Microstructure

Thermodynamics and Kinetics of Defects

Next workshops for Actinide fuel fabrication ?

SFR / MOX

FUEL FABRICATION

(the core of the new

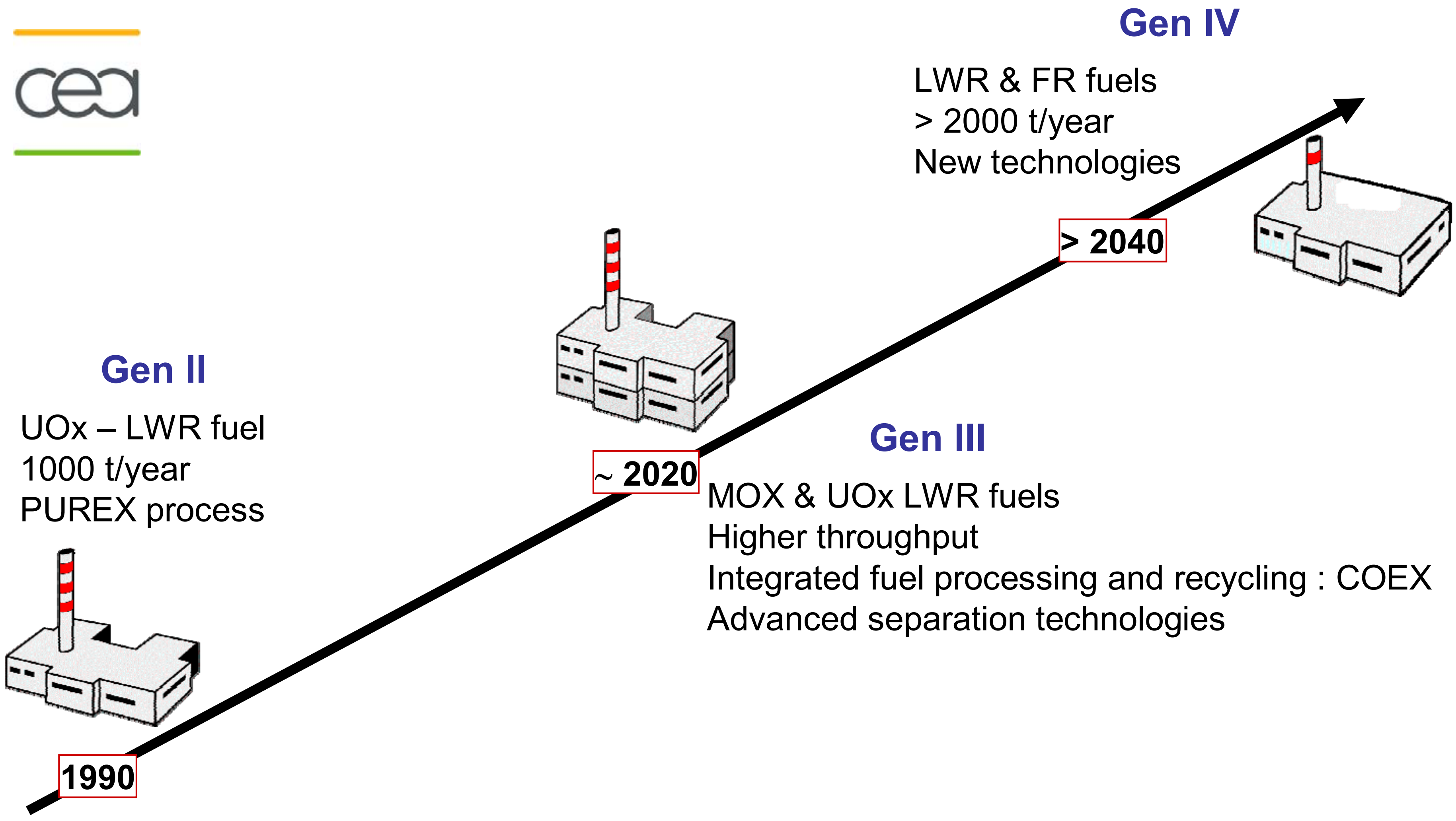
SFR ASTRID proto, tons)

MINOR ACTINIDES

PILOT at MARCOULE

(experimental pins, kg)

The next generation of reprocessing facilities



Gen II

UOx – LWR fuel
1000 t/year
PUREX process

Gen III

MOX & UOx LWR fuels
Higher throughput
Integrated fuel processing and recycling : COEX
Advanced separation technologies

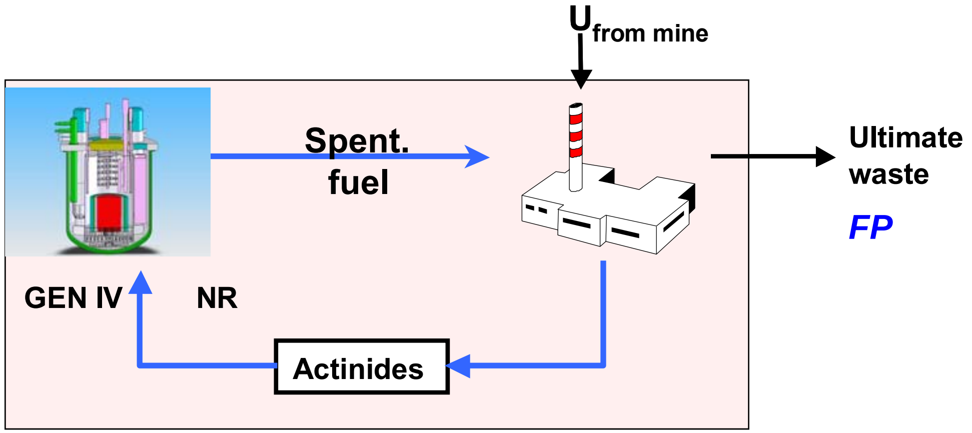
Gen IV

LWR & FR fuels
> 2000 t/year
New technologies

Looking for a long term management strategy?

⇨ **Results already in use :**
- improved industrial facilities and processes
- since 1991, significant waste volume reduction (by a factor of 6)
- feasibility of disposal

⇨ **A continuous improvement process to be continued:**
- for opening the scope of possible solutions
- and defining future electricity generating systems



- **Minimising ultimate waste**
- **Optimal use of energy-bearing natural resources**
- **Proliferation resistance**

General conclusion: in France, towards the 2012 milestone

- Recycling options, for sustainable FR systems
- Some options still open (what, and how), assess benefits/cost ratio by 2012 : a progressive step by step approach (from U and Pu first, Am to MAs recycling?)
- A need for flexible processes?
- - A specific new and important program on reprocessing modelling
- - A consolidation program for industrial potentiality by 2012
- - From separated MA solutions to Am and MA-bearing experimental fuels: to be tested at pin scale in the ASTRID SFR after 2020 …

The two mainstays of the future nuclear energy

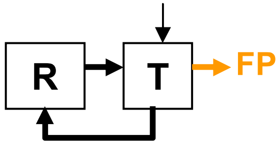
Nuclear energy for the 21st century

RENAISSANCE

avoid spent fuel accumulation!

3rd generation reactors with advanced recycling proven technologies

SUSTAINABILITY



4th generation reactors with appropriate fuel cycle options