Recent progresses in advanced Actinide recycling processes

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Advanced processes for Actinide recycling : French recent experiments and results

December 30, 1991 and June 28, 2006

OUTLINE

1 – (1995-2005) main achievements, in the frame of the 1991 Act

2 – (2006-2012) Recycling R and D program, in the frame of the 2006 Act

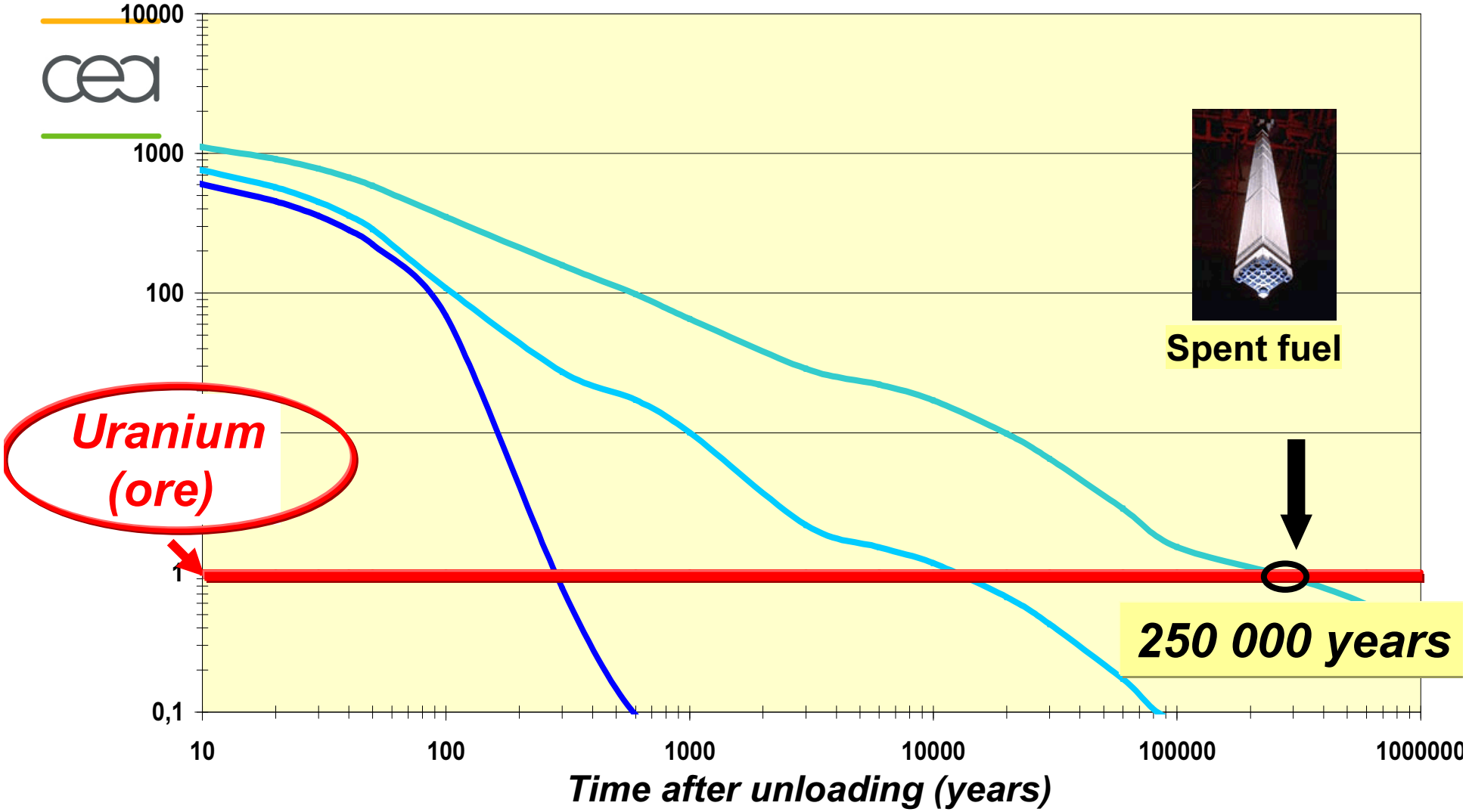
3 – Recent and on going R and D results for MAs recycling

4 – Industrial potentiality

5 – Conclusion

Atalante in Marcoule

Radiotoxicity of waste, to be disposed



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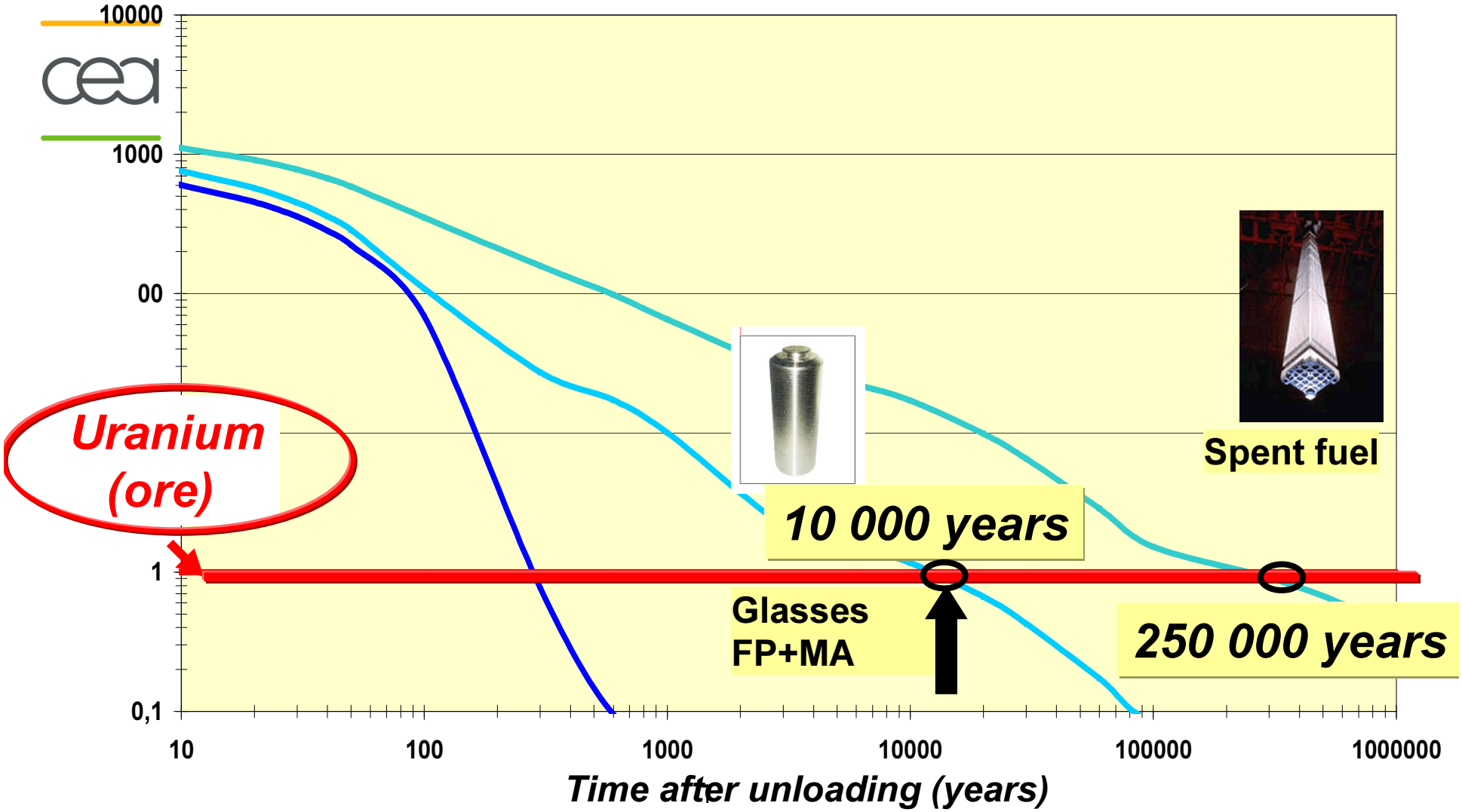

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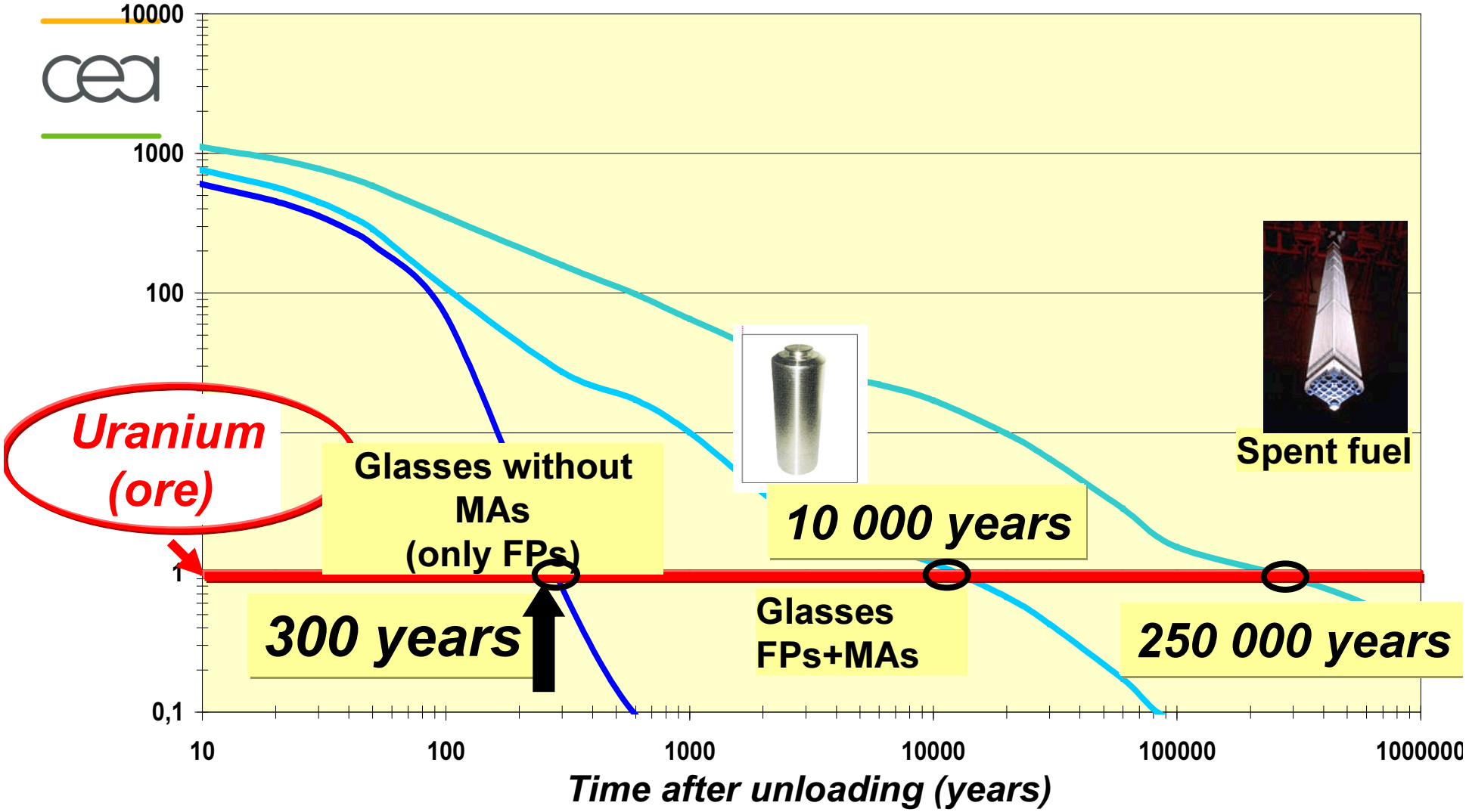

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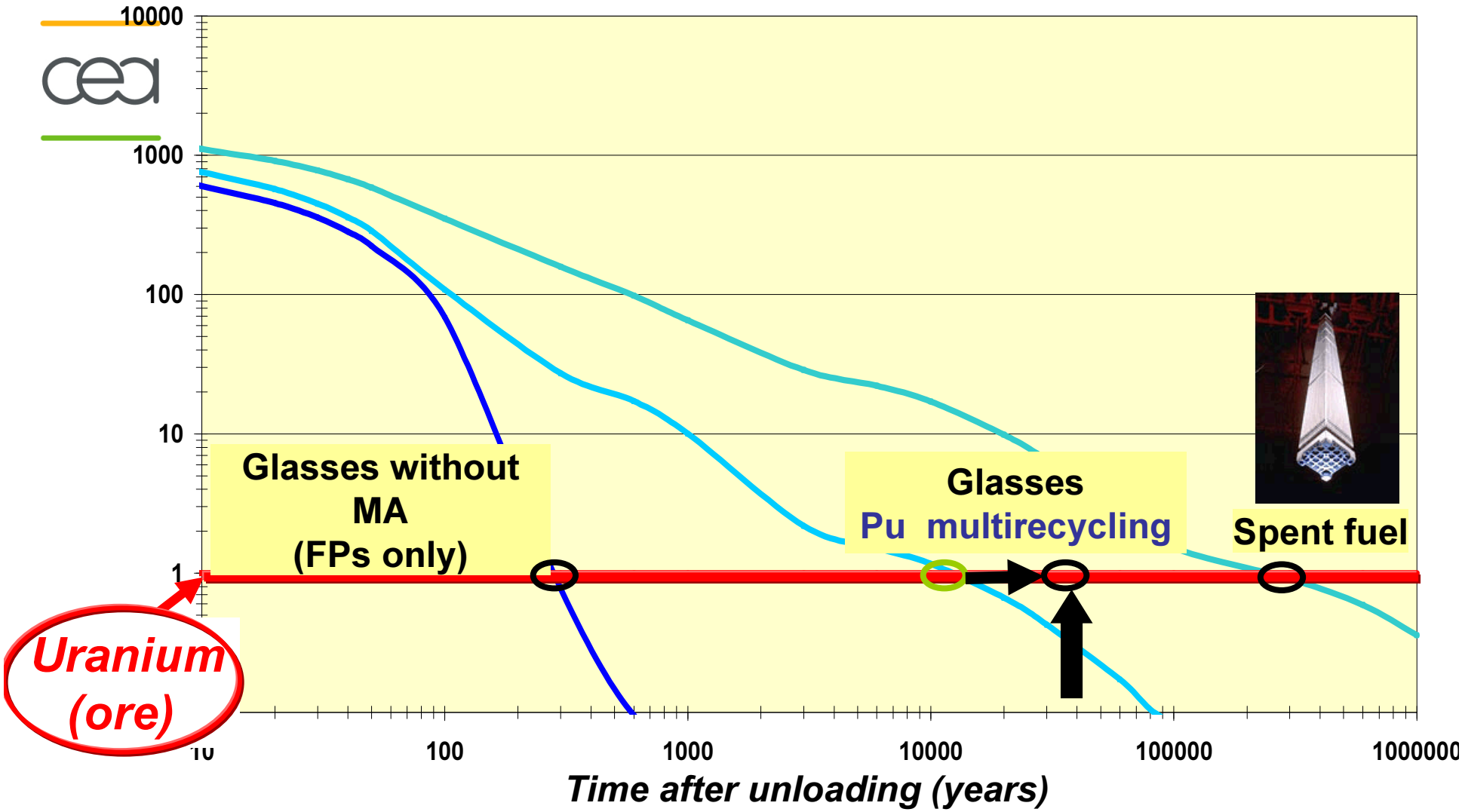
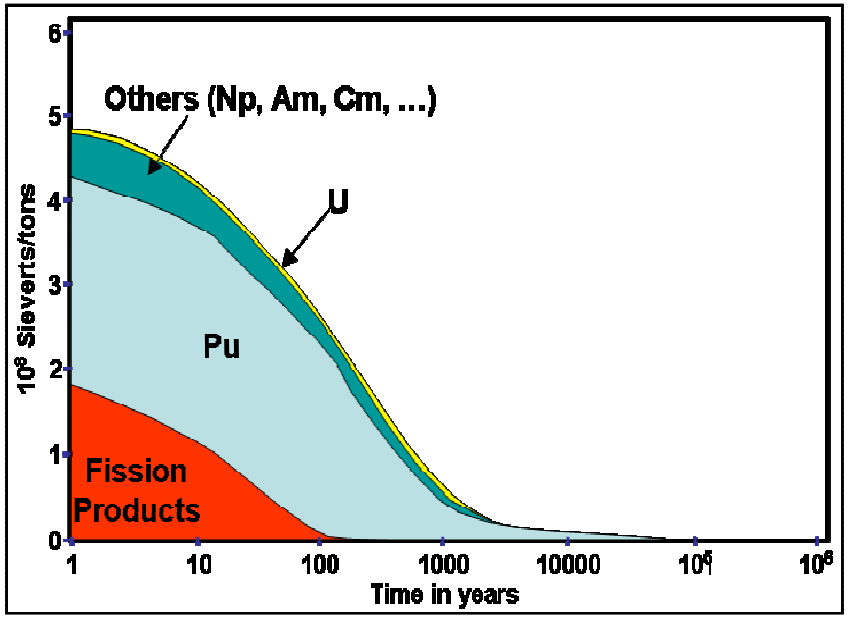


LLRN Recycling for waste management

1st contributor : Pu

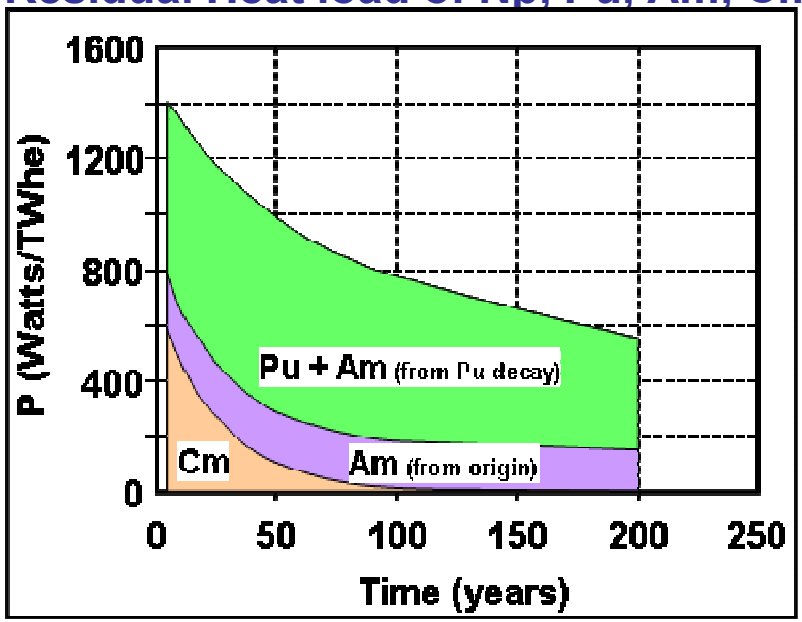
2nd contributor : Minor Actinides Np, Am, Cm

3rd contributor : Long-Lived Fission Products (LLFP)



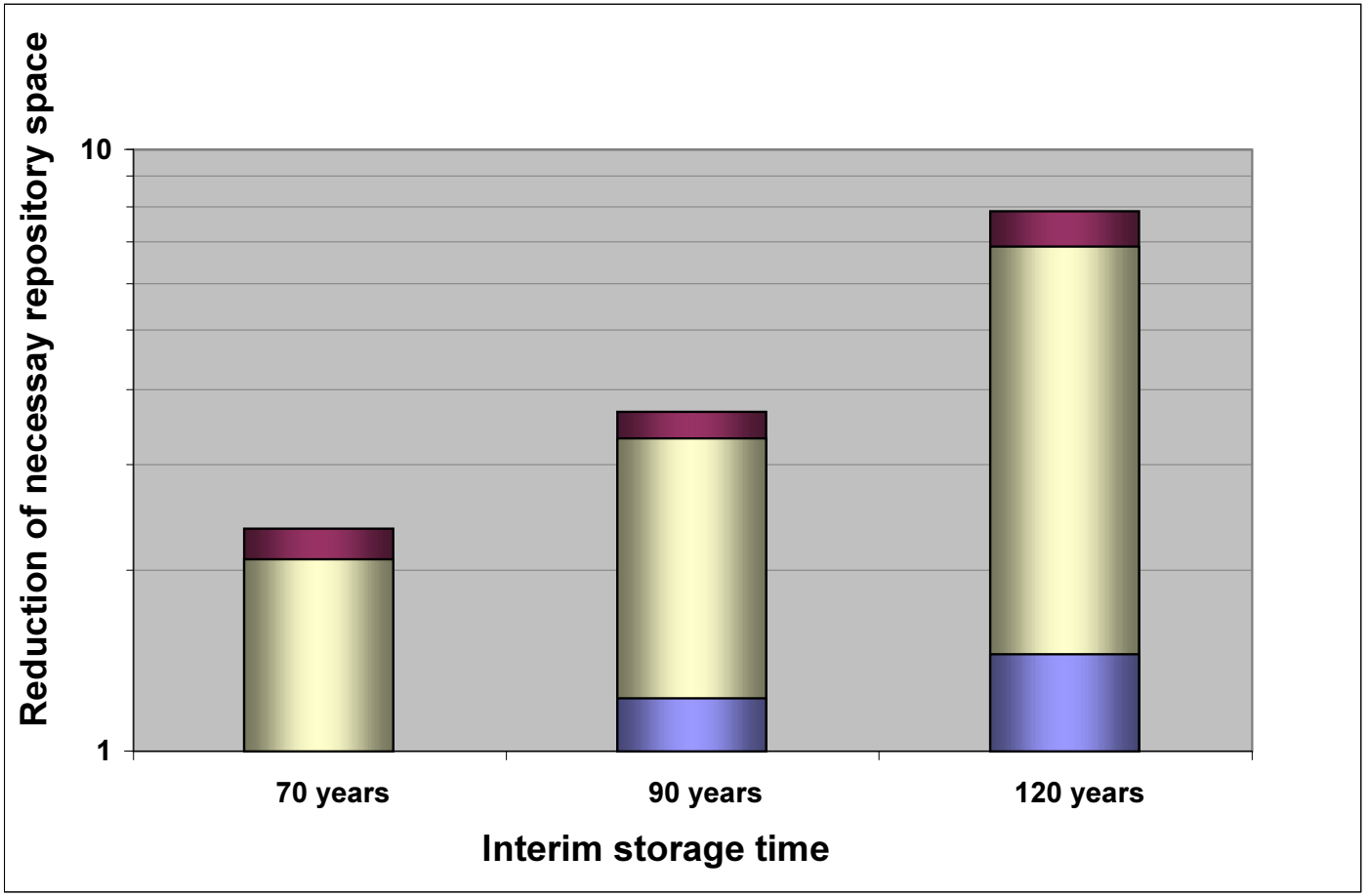
Potential radiotoxicity

Residual Heat load of Np, Pu, Am, Cm

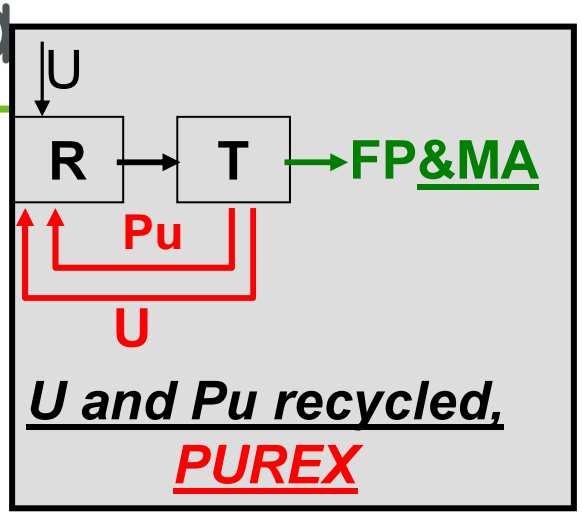


Processing and Recycling should minimize both the needed repository space and environmental impact

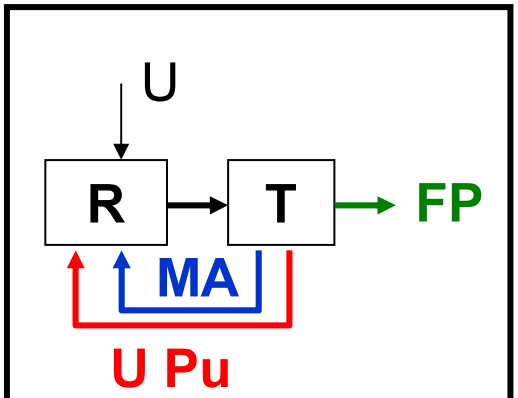
Needed waste repository space, versus strategy



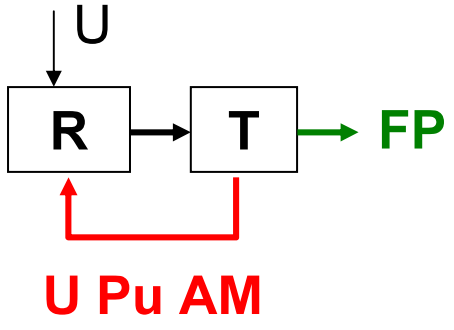
Actinide recycling : what fuel cycle option ?



U and Pu recycled, PUREX



MA heterogeneous recycling



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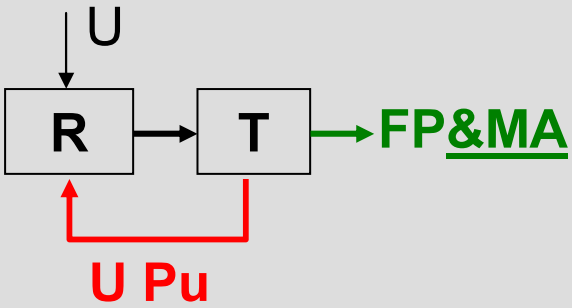
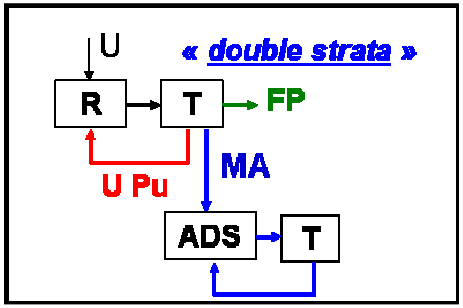


U and Pu recycled together, COEX

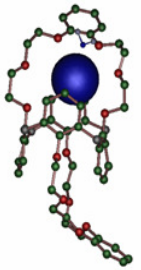
MA : Np, Am, Cm



The enhanced Partitioning 2005 results

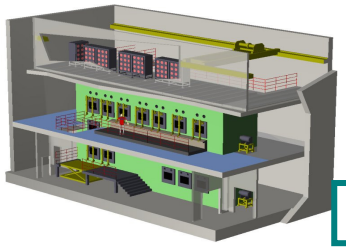
- **A true challenge : a sophisticated partitioning chemistry under highly radioactive conditions ; fundamental and applied research :**
  - exploration : new extracting molecules and systems
  - fundamentals : in-depth study of mechanisms at work

A few hundreds of new molecules



- **Applied research :**
  - process design
  - lab experiments on actual spent fuel material
  - « demonstration » experiments : integration, representativeness long-lasting performance, secondary waste

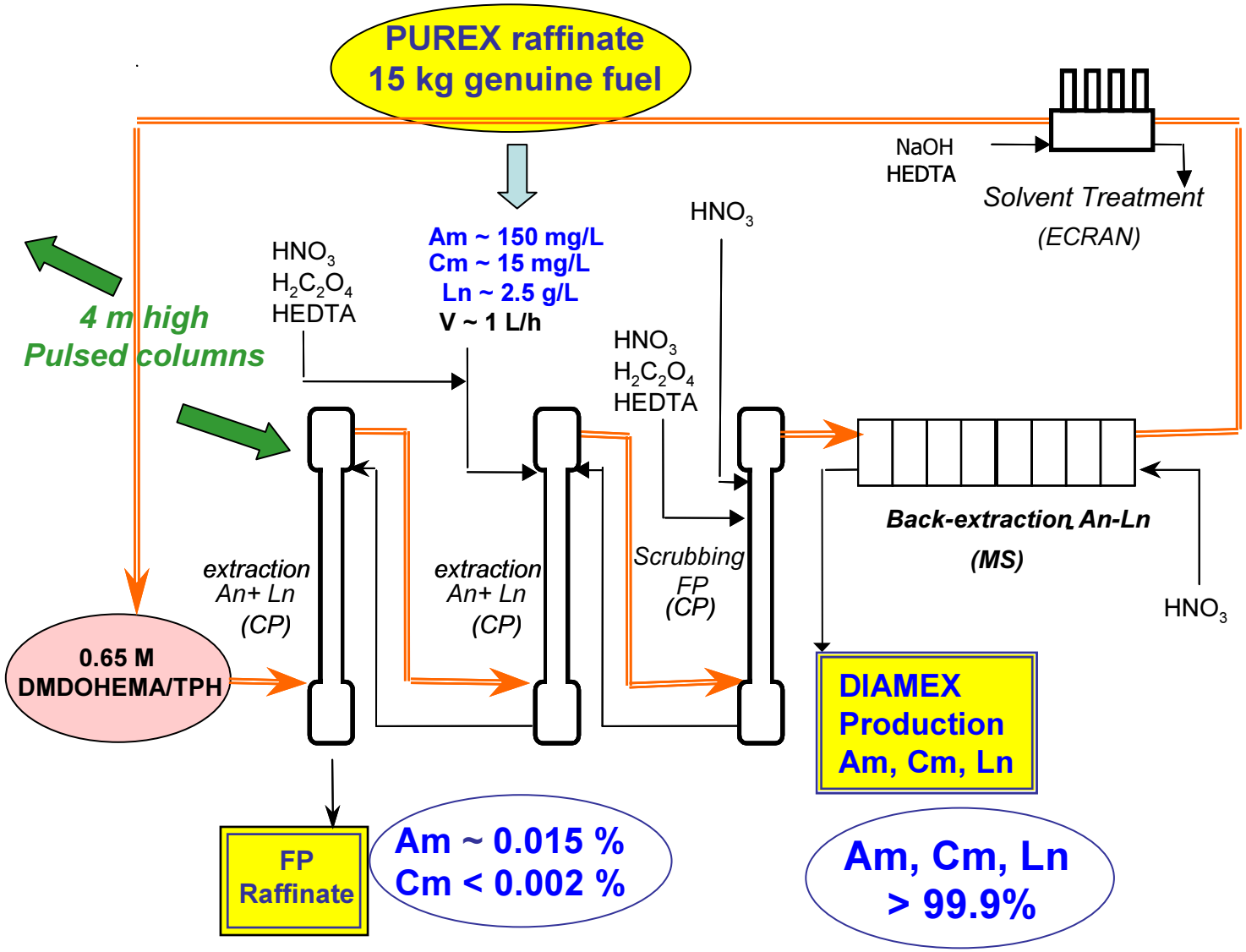
Scale : 1/10000



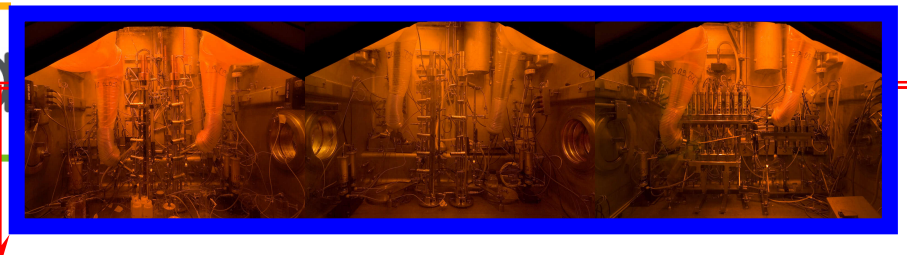
Scale : 1/100 to 1/1000

- **Neptunium: recovery ratio up to 99%, with modified La Hague PUREX**
- **Americium and Curium: recovery ratio up to 99.9%, with new DIAMEX-SANEX process**
- **Technetium: recovery ratio from 45 à 90%**
- **Iodine: - recovery ratio > 97% with PUREX**
  - additional recovery up to ~ 99% possible
- **Cesium: recovery ratio > 99.8%, with the use of the calixarene extractant**

DIAMEX demonstrative hot run, November 2005



SANEX demonstrative hot run, December 2005



Solvent
DMDOHEMA 0.5 M
+ HDEHP 0.3 M
TPH

DIAMEX
Production
Am, Cm, Ln
~ 45 mL/h

extraction-scrubbing

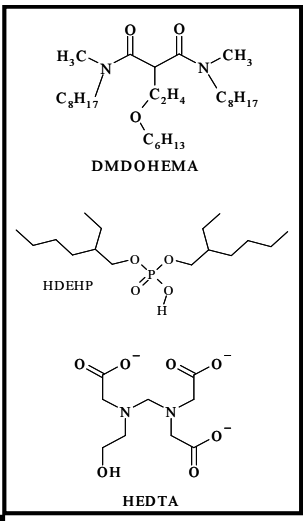
HNO_3

Raffinate

Am, Cm < 0.01%

HNO_3

NaOH
HEDTA



Ln
scrubbing

An stripping

Ln stripping

An

Am, Cm > 99.9%
F.D.Ln ~ 80

HEDTA
Citric ac.

Ln

Am < 0.06 %
Cm < 0.03 %

HNO_3

Future fuel cycle options in the 20006 Act : the 2012 milestone

1. 2005 conclusions :

- **Transmutation of Fission Products (I, Cs, Tc) is either not feasible or unrealistic ; it should be abandoned**

- **MAs transmutation is not realistic in LWR; on the contrary, for FR, transmutation calculations and experiments at pin scale have been carried out for americium and neptunium in a power reactor, such as Phénix, which demonstrates the feasibility of their transmutation in SFR**

2. 2006-2012 objectives

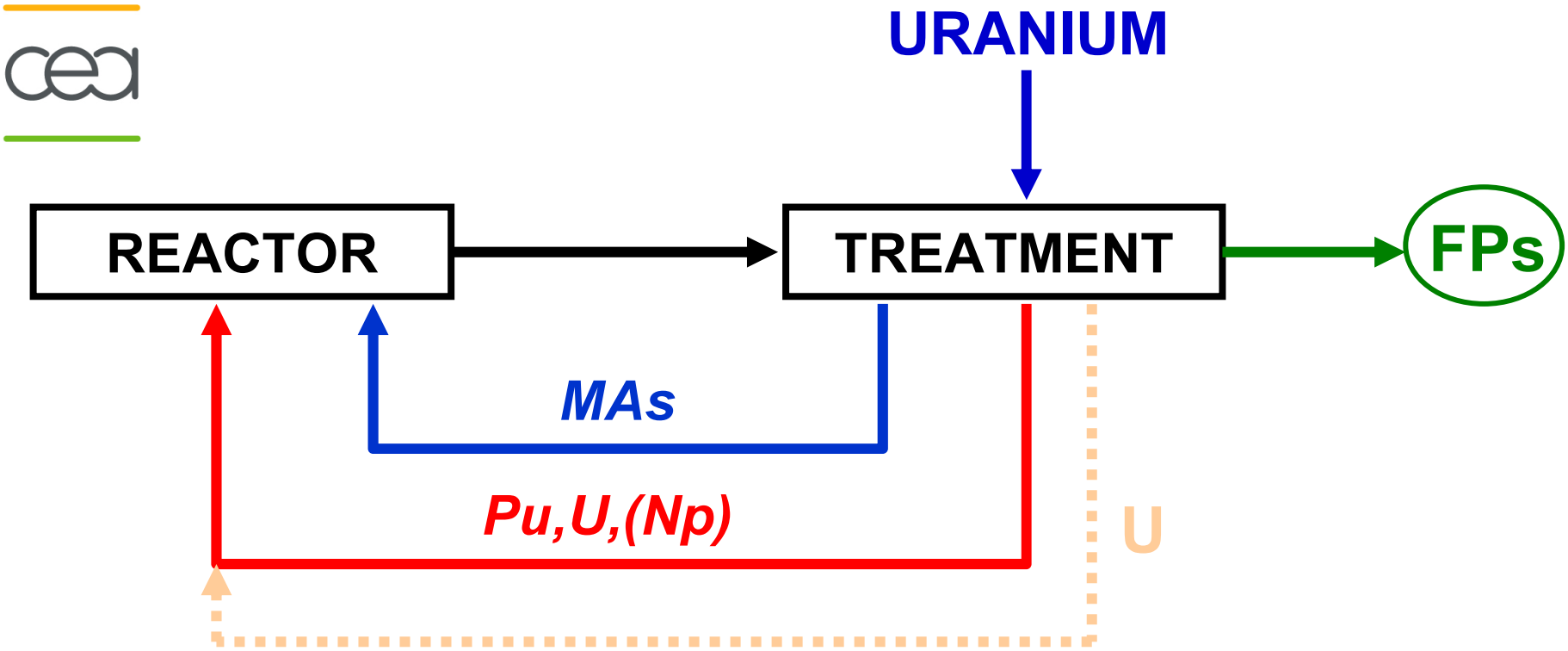
- **Define the several recycling options of interest, which could be successively deployed (heterogeneous, homogeneous, all-actinide, Americium only,…)**

- **Assess benefits /costs ratio for the several recycling options, considering diverse criteria and "densification" of the final storage**

- **Design / Optimize separation processes, transmutation fuels and their fabrication processes**

- **and gather technical elements for industrial operation evaluation**

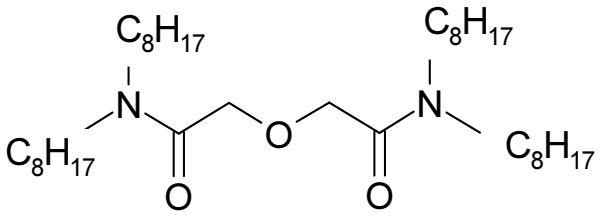
Fuel cycle, the MA heterogeneous recycling option

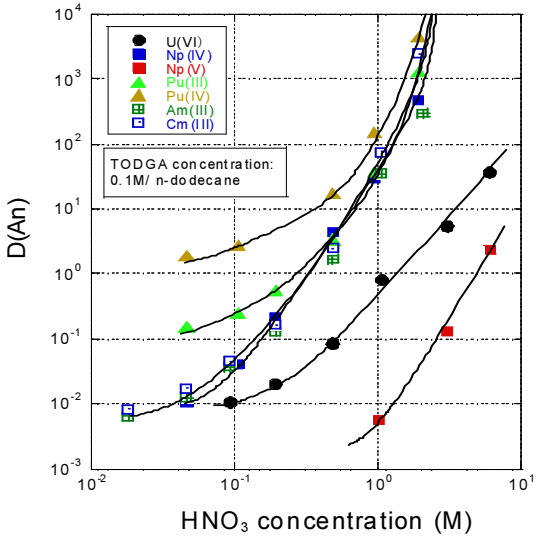


- U, Pu, Np by COEX™
- Am (and Cm) separation : simplified DIAMEX-SANEX,…
- Am (and Cm) recycled on dedicated « targets-blankets »

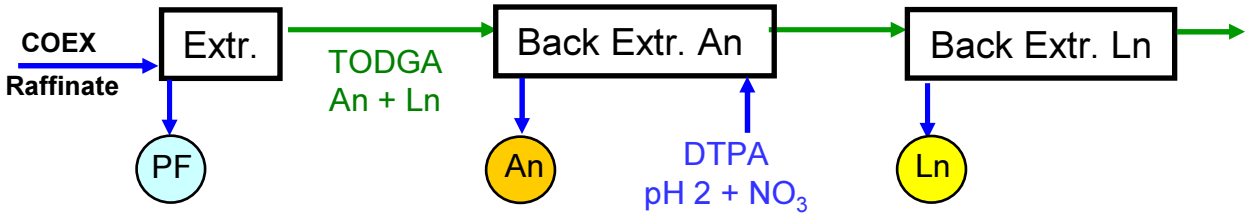
Simplified SANEX-TODGA process

- **Co-extraction An (III) and Ln (III) with TODGA, using HNO_3 4N**





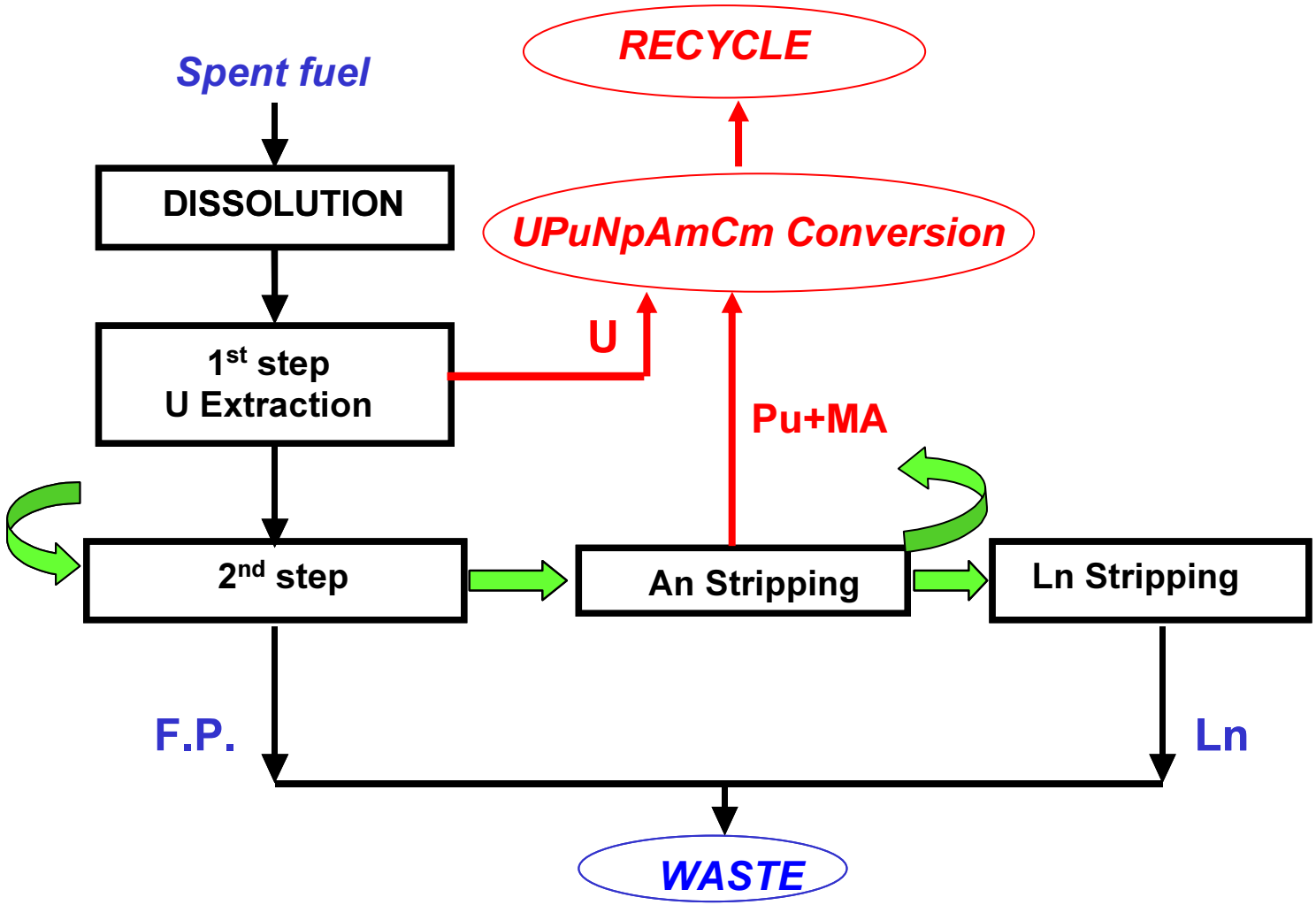
- **Selective back-extraction of An (III)**
  - **With polyamino-carboxylic hydrophile complexing agent**



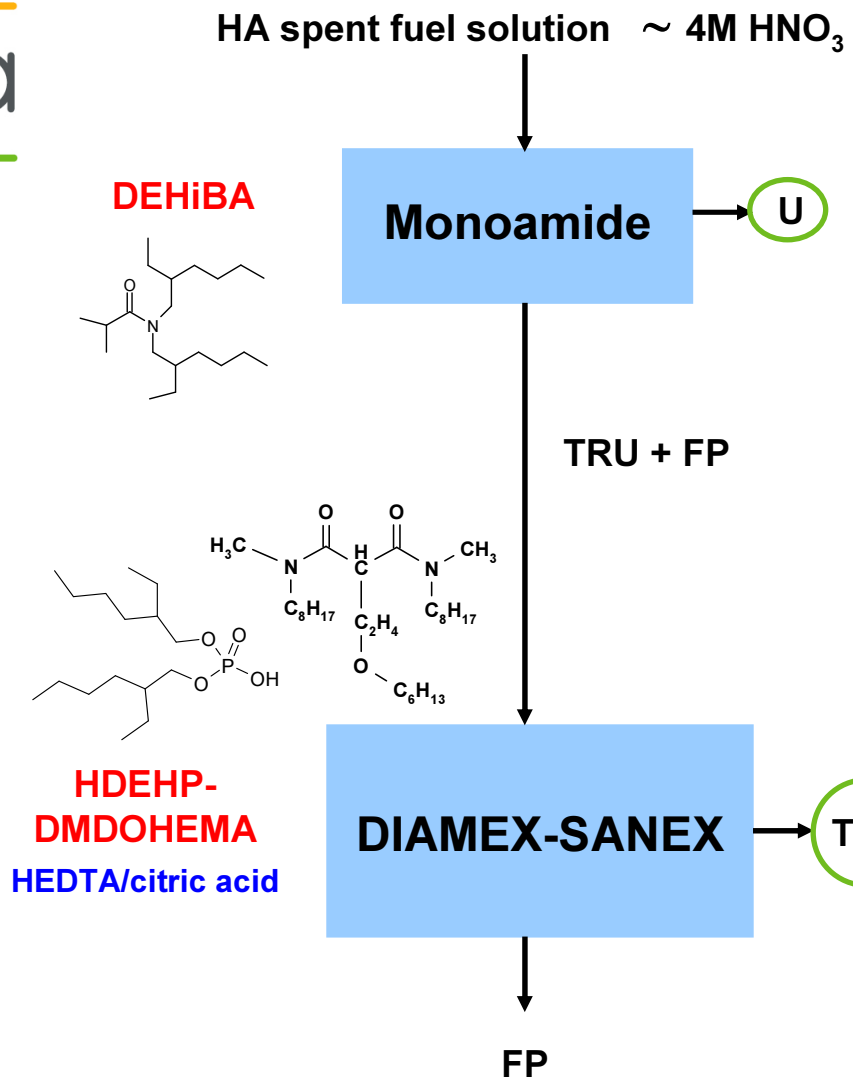
- **<u>Advantages</u> : simple scheme, TODGA synthesis low cost**
- **<u>Drawbacks</u> : high sensitivity of the Am-Cm back extraction step to pH and temperature**

Fuel cycle, the MA homogeneous recycling option

The grouped actinide GANEX concept



The GANEX process



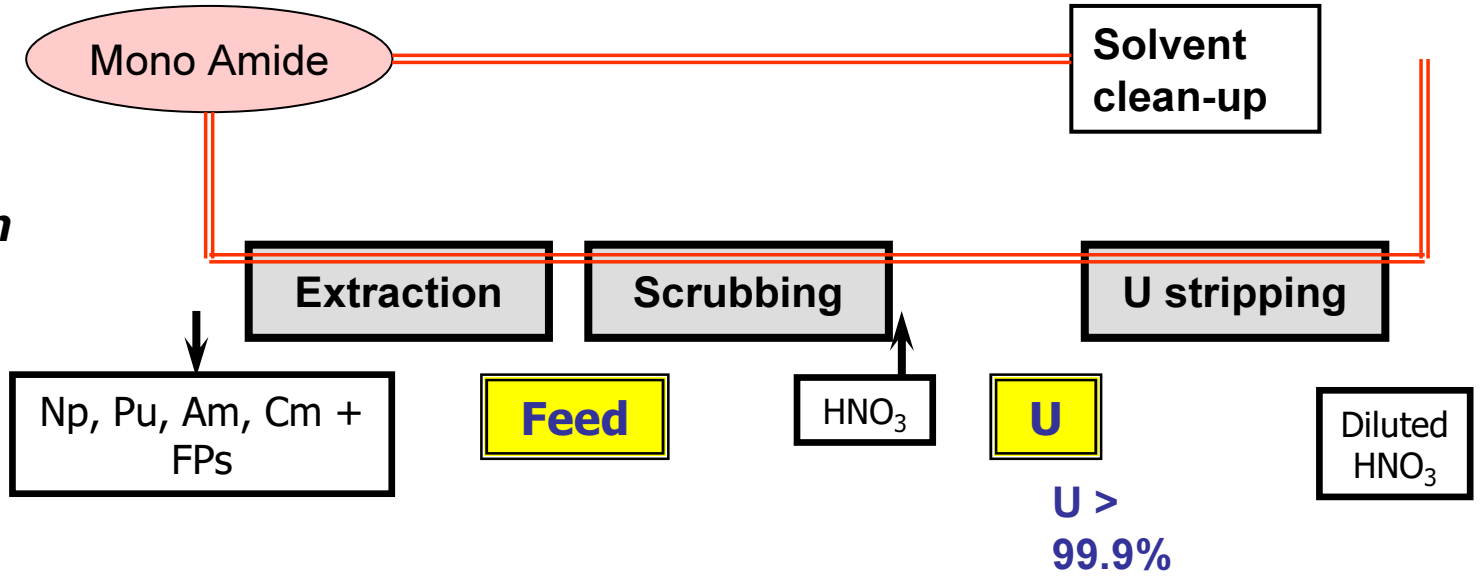
Excellent compromise between complete Uranium extraction, and good U(VI)/Pu(IV) selectivity

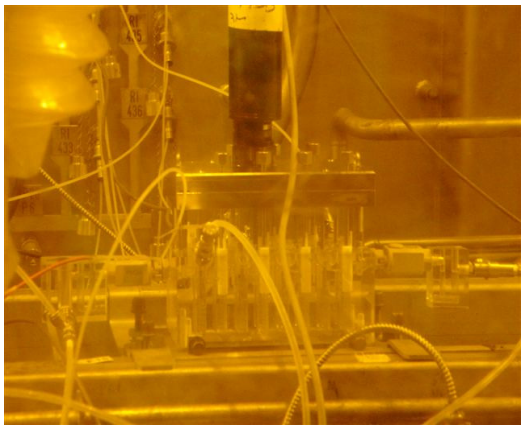
Adaptation of the process already demonstrated in 2005

GANEX demonstrative hot runs, 2008

1st step : U selective extraction

(performed successfully in June 2008)



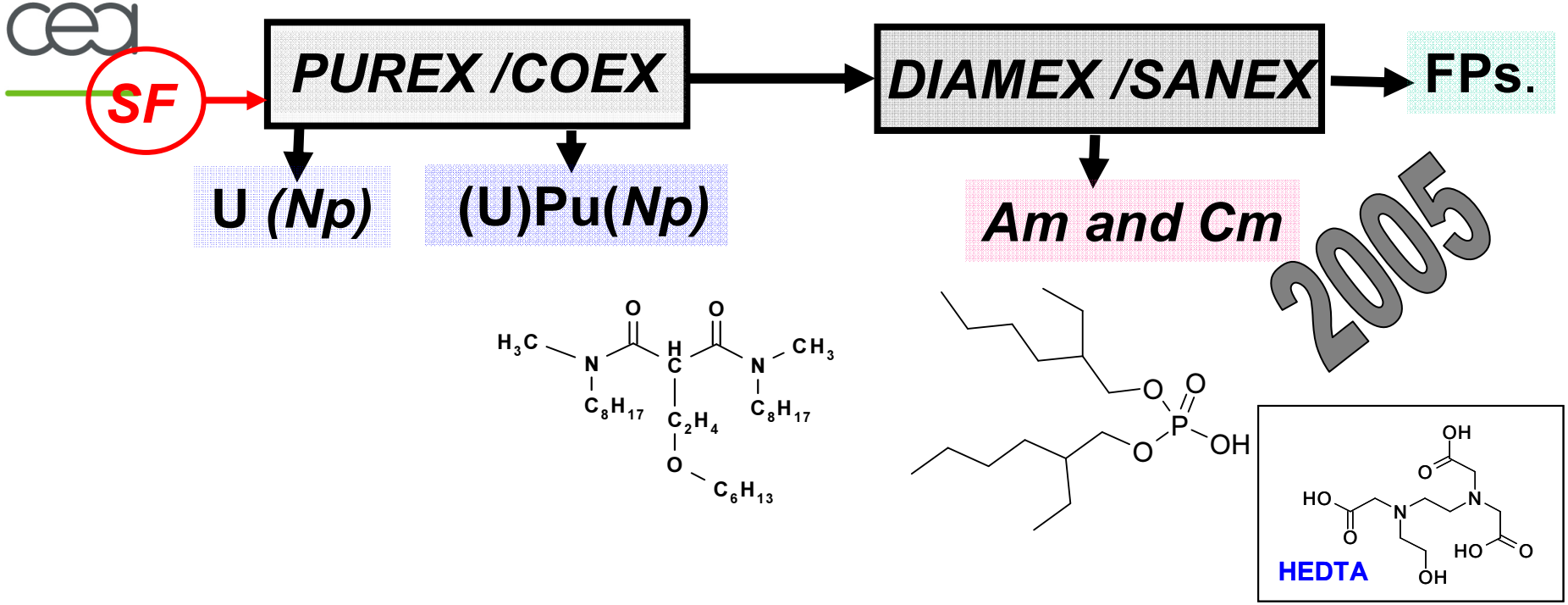


2nd step : Pu-Np-Am-Cm co-recovery *(DIAMEX-SANEX diamide-based process)*

(performed successfully in November 2008)

Partitioning : concepts and results

- ***Enhanced separation :***



Partitioning : concepts and results

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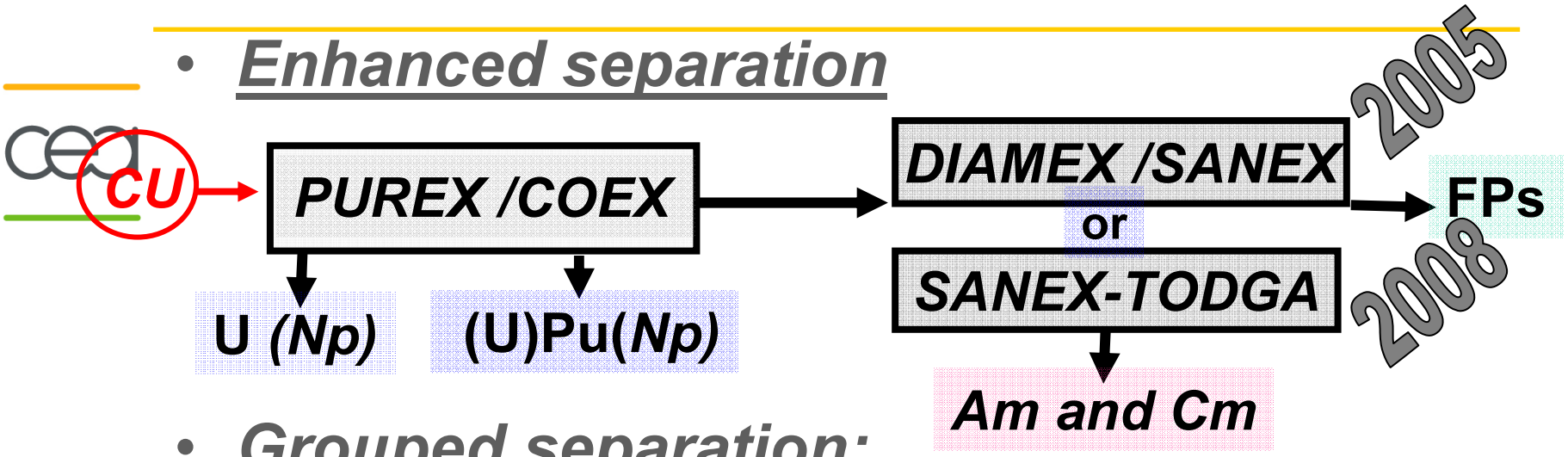
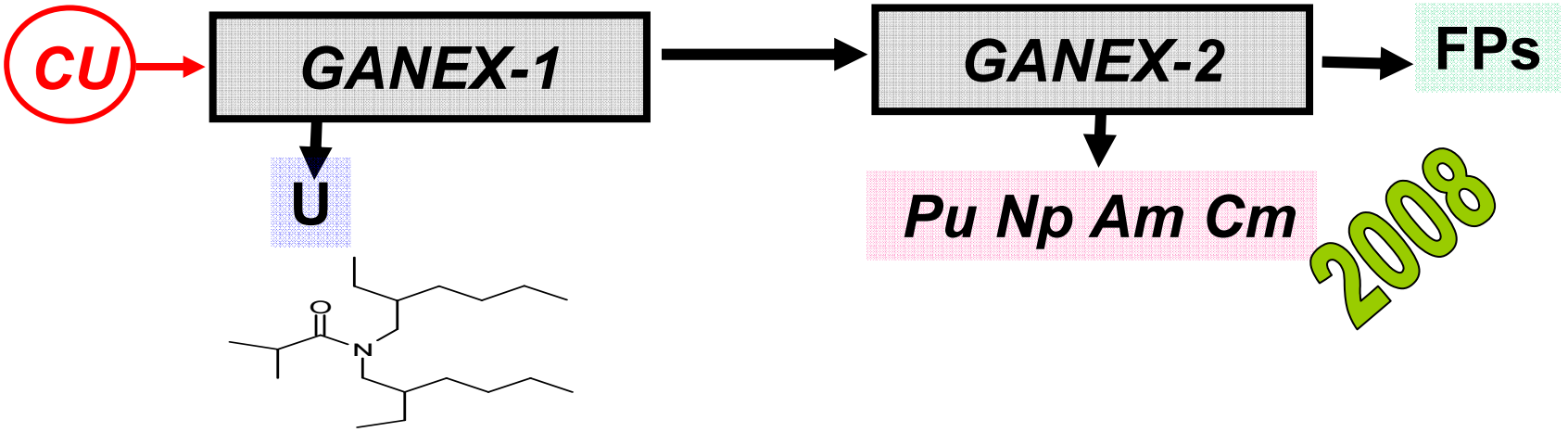


- ***Grouped separation:***



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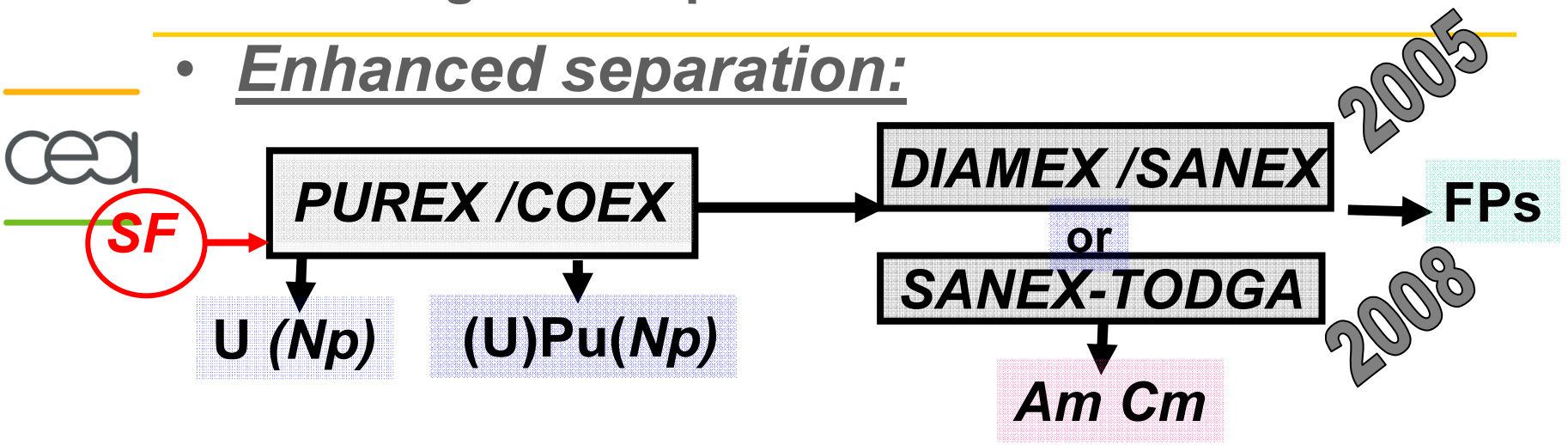

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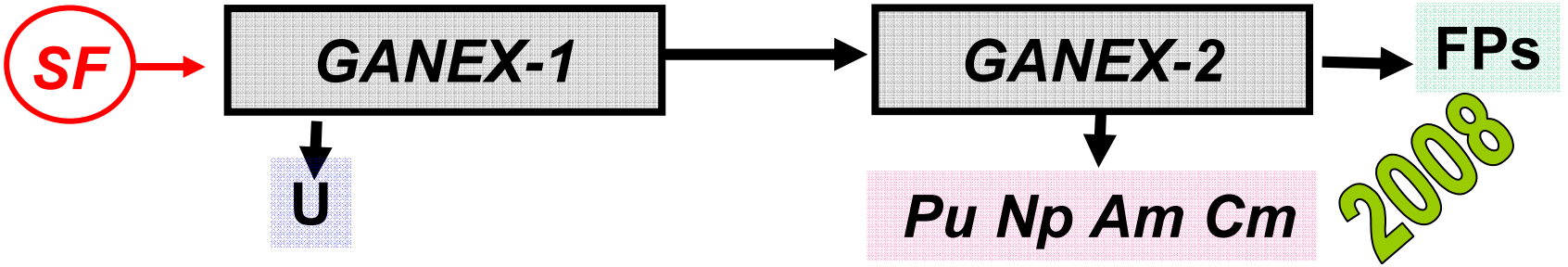
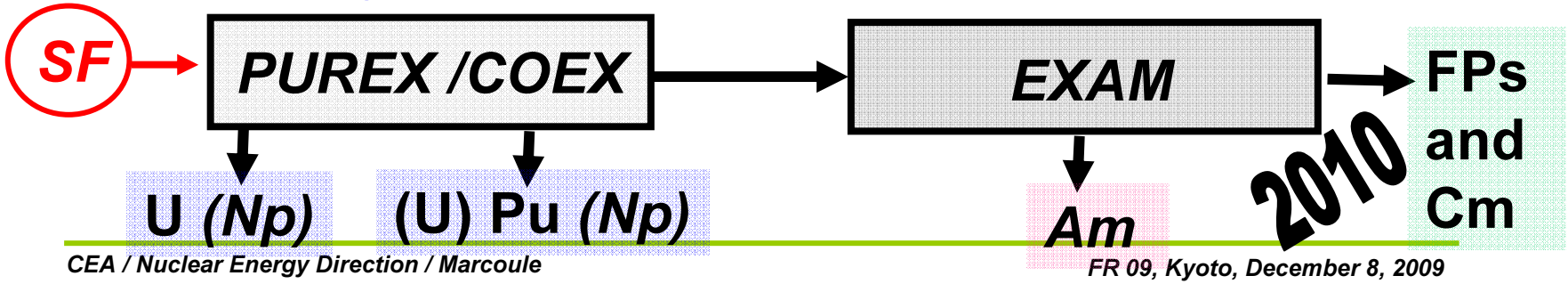
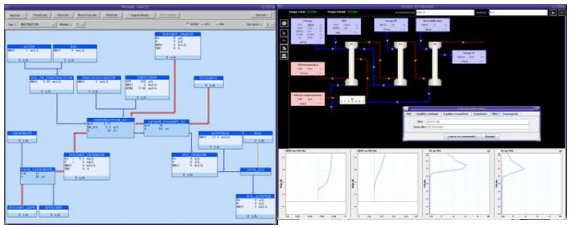


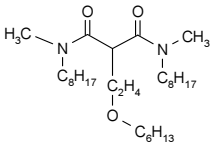
- ***Am only separation:***



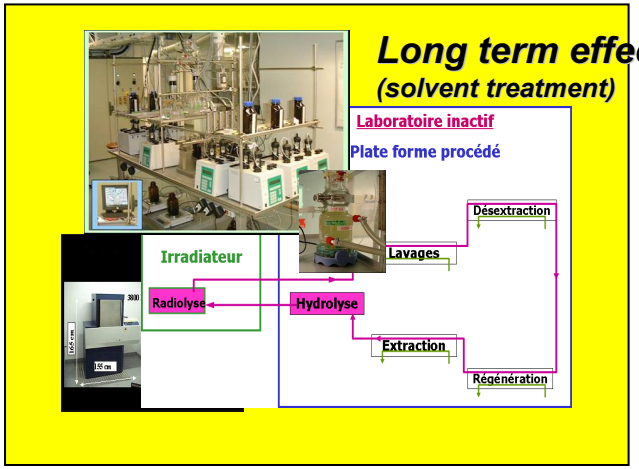
Separation process : towards industrialization



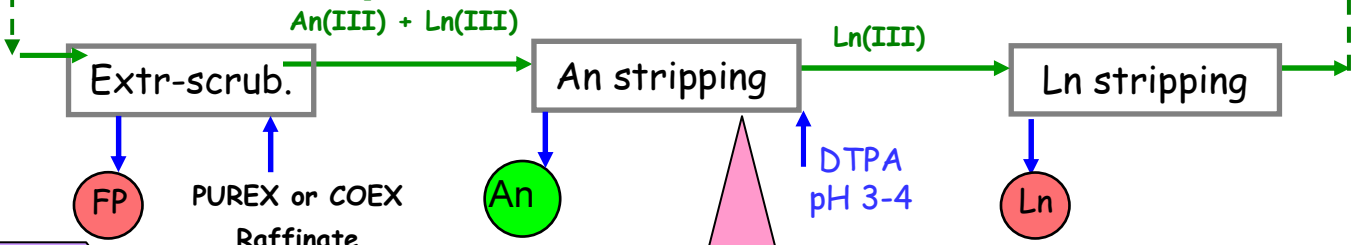
Modeling
(flowsheet, sensitivity, operation)



Extractant synthesis

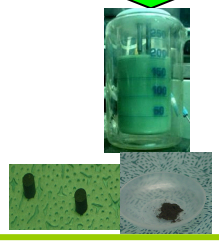


Core of the process demonstrated at lab scale

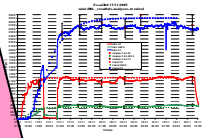


Equipments
(Implementation, extrapolation)

Interface
Co-conversion



In situ analysis
(cations, pH...)



FP solution adapted to vitrification
(Concent. Calcination)

MA bearing fuels : development of fabrication process

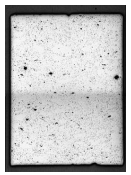
- **Synthesis of MA compound powders, starting from separated MA nitric solution (interface co-conversion)**
- **A promising process : the oxalic co-precipitation, calcination, then direct-powder or UO_2-diluted powder pelletizing**

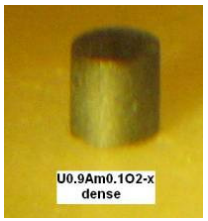
Pu

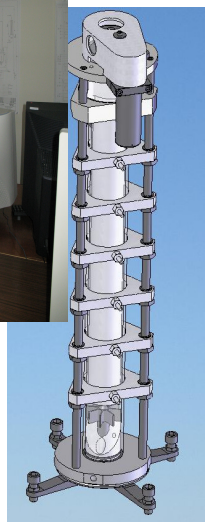
U and Pu

U, Pu, Np, Am,..Cm

COPIX (UPu)O2 irradiation test in Phenix, 2008-2009







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

Conclusion : towards 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?
- On-going research in the CEA Atalante facility, with international collaboration for optimizing separation process
  (many process options already explored, optimization, simplification)
- A specific new and important program on reprocessing modeling
- 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 …