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Recent progress in Advanced Actinide Recycling Processes

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Recent Progress in Advanced Actinide Recycling Processes

December 30, 1991 and June 28, 2006

OUTLINE

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

2 – (2006-2012) Partitioning 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, and next plan

5 – Conclusion

Atalante in Marcoule

LLRN Recycling for waste management

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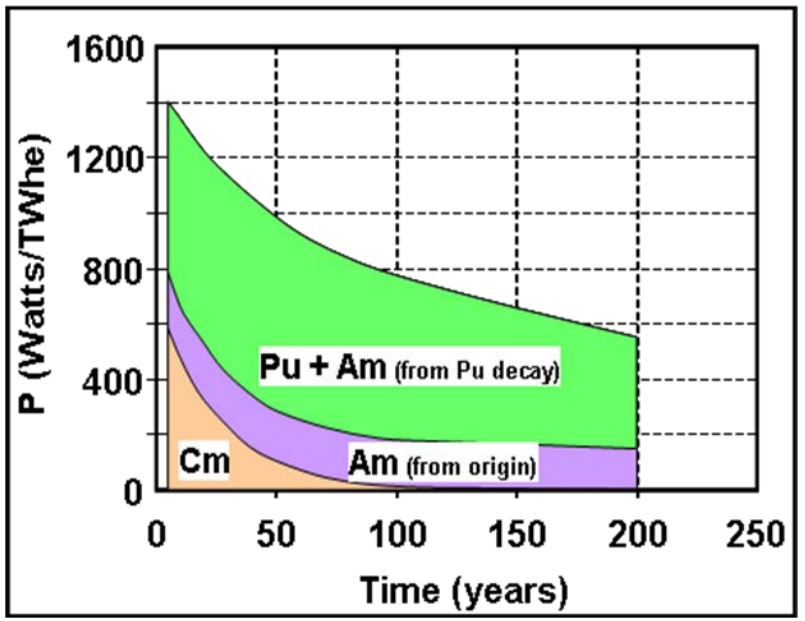
1st contributor : Pu

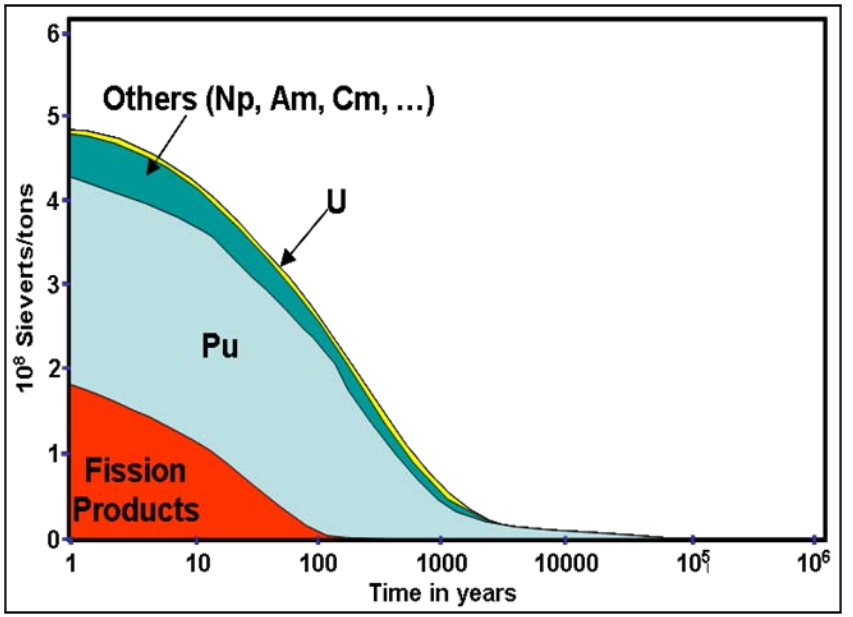
2nd contributor : Minor Actinides Np, Am, Cm

3rd contributor : Long Life Fission Products (LLFP)

Residual Heat load of Np, Pu, Am, Cm



Potential radiotoxicity

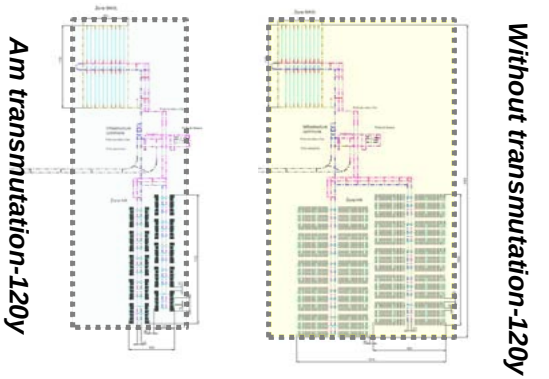


Processing and Recycling should minimize the repository space

LLRN Recycling strategy

- Aim of recycling : to minimize the **quantity, and residual heat** of long lived nuclear waste

- Potential gain : to decrease the volume and to ease the conditions of a deep geological repository



- Fundamental hypothesis : closed cycle, GEN IV fast neutron reactor implemented at mid or long term (whatever the cooling : Na, gaz,…)
- Reference strategy : LLRN (Am, Cm, Np) recovery, and their transmutation in reactors:
  - GEN IV SFR : Pu multi-recycling and MAs recycling, beginning after 2040
  - Homogeneous or heterogeneous recycling
  - Option of the « double strata » LWRs – Accelerator Driven Systems

- The research program for the different options : demonstrations of scientific and technical feasibilities,

  before pre industrial development of recycling expected after 2020

The enhanced Partitioning 2005 results

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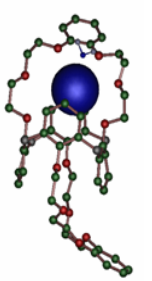
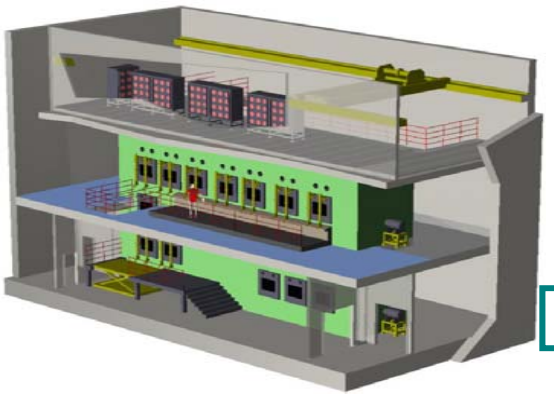
⇨ Applied research :

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

A few hundreds of new molecules



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

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

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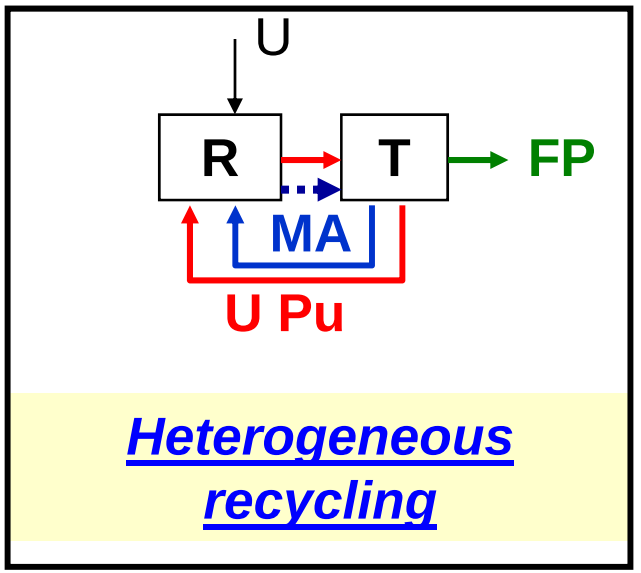
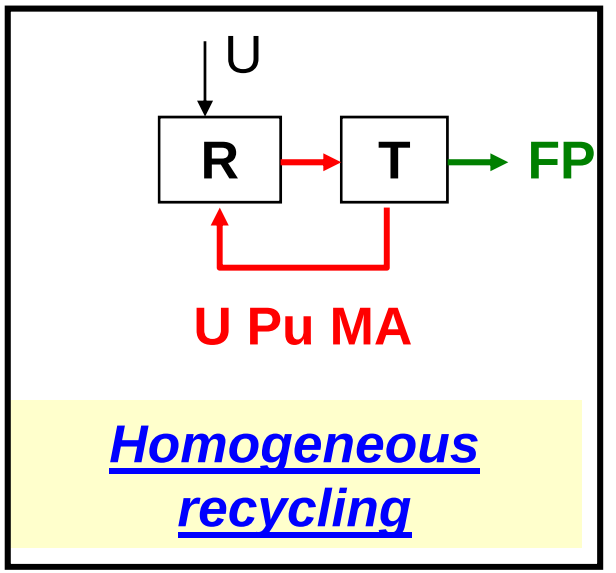
- Transmutation of Fission Products (I, Cs, Tc) is either not feasible or unrealistic ; it should be abandoned
- In LWR, MAs transmutation is not realistic
- For FR, transmutation experiments, at pin scale (Superfact, Ecrix,…), have been carried out for americium and neptunium in a power reactor, such as Phénix, which demonstrates the feasibility of their transmutation in SFR
- 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

The P and T MA recycling options



Heterogeneous recycling

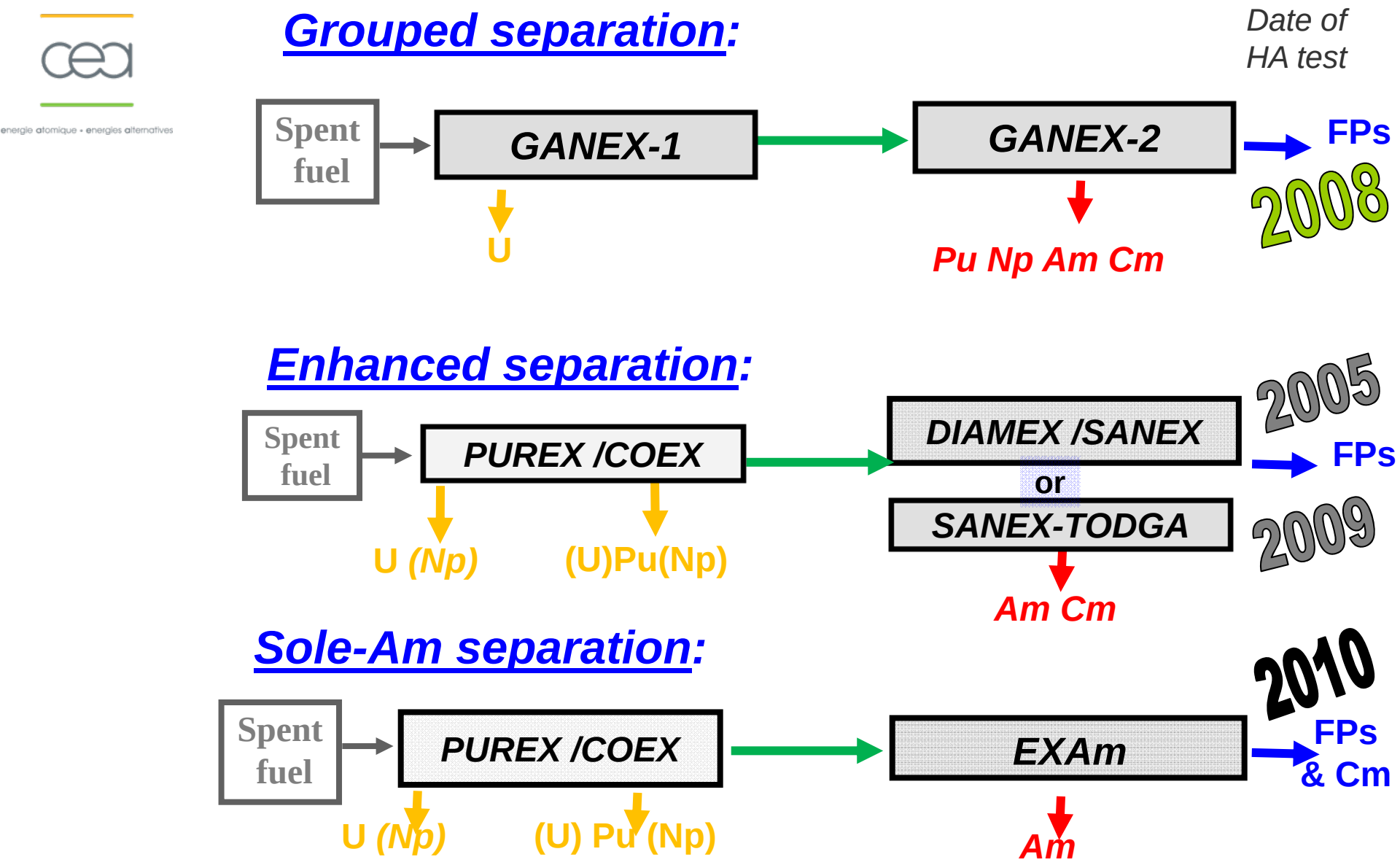
UPu: COEX
MA: SANEX-TODGA
Am: EXAm



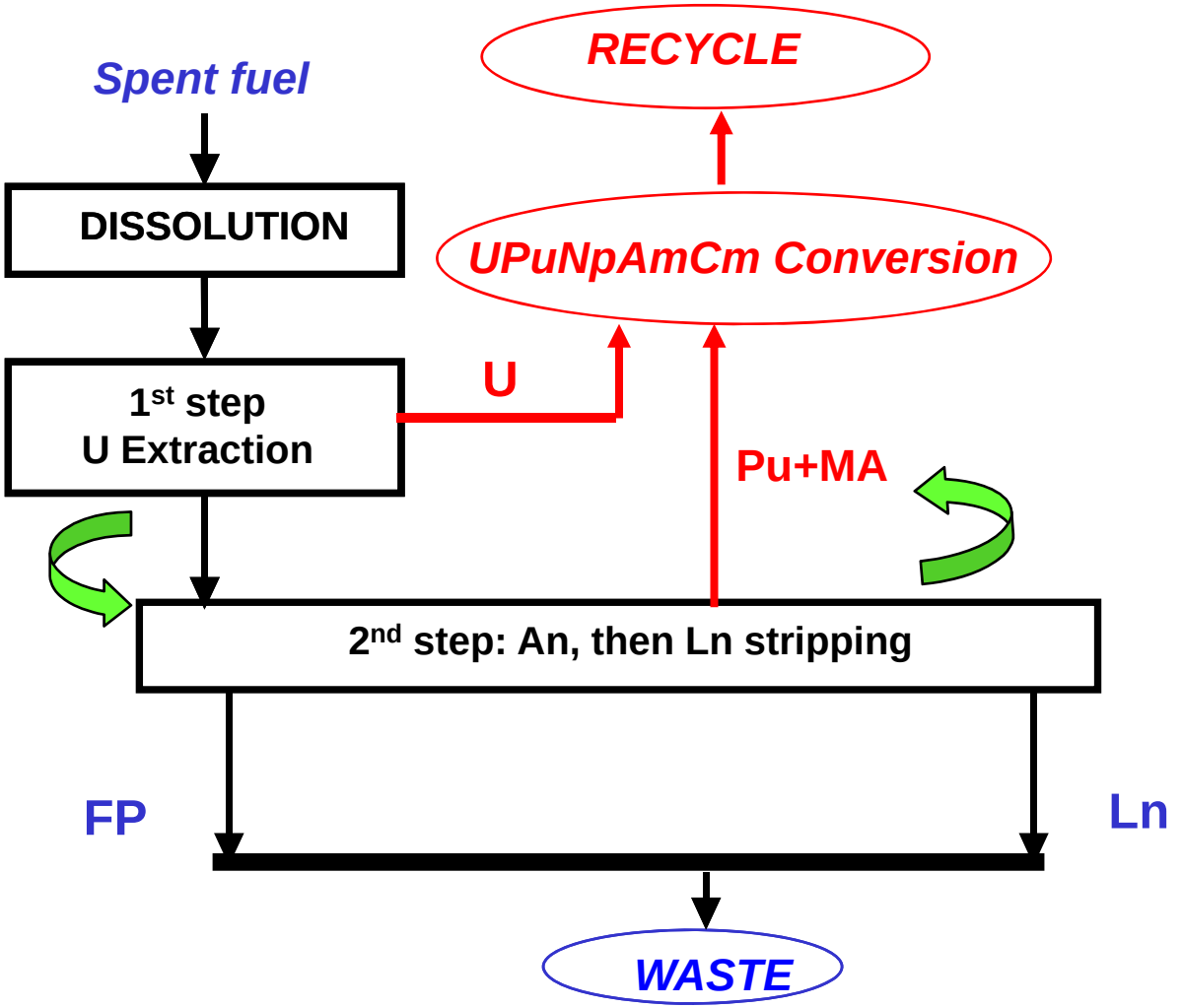
Homogeneous recycling

PuAmCmNp: GANEX

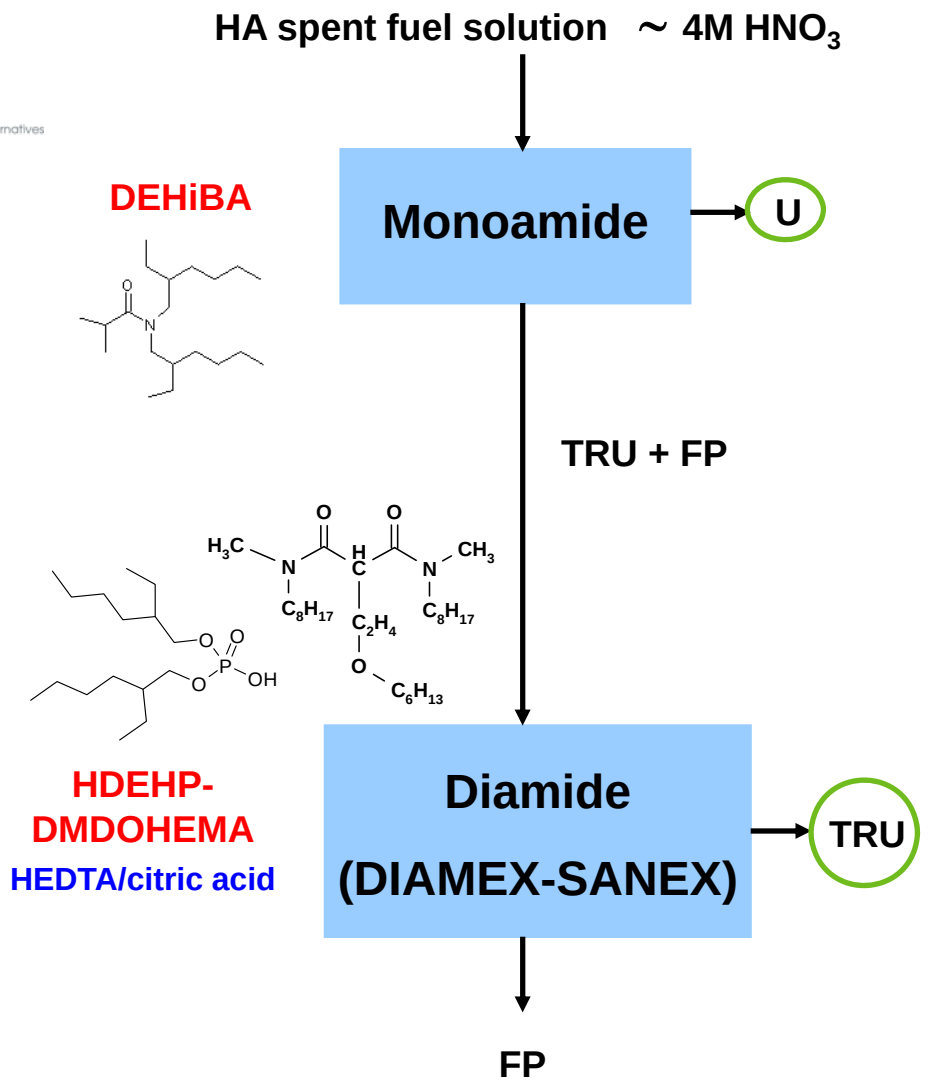
The MA partitioning options



The grouped actinide GANEX concept



Homogeneous recycling: the GANEX process



1st step: excellent compromise between complete Uranium extraction, and good U(VI)/Pu(IV) selectivity

2nd step: adaptation of the process already demonstrated in 2005

Homogeneous recycling: GANEX 2008 demonstrative hot runs

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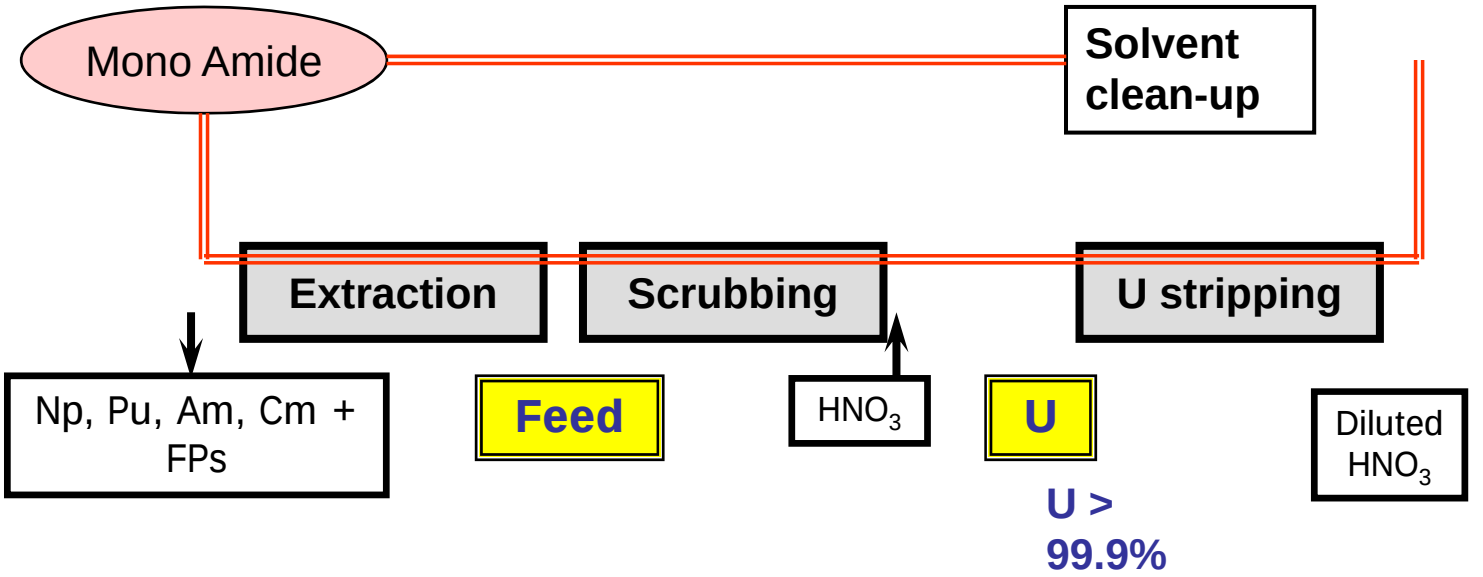
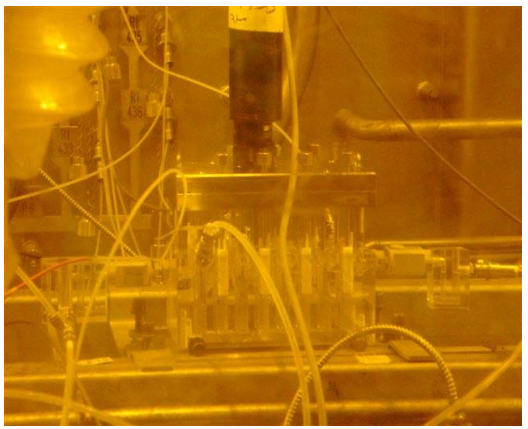
1st step : U selective extraction *(performed successfully in June 2008)*





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

(performed successfully in November 2008)

Heterogeneous recycling: the simplified SANEX-TODGA process

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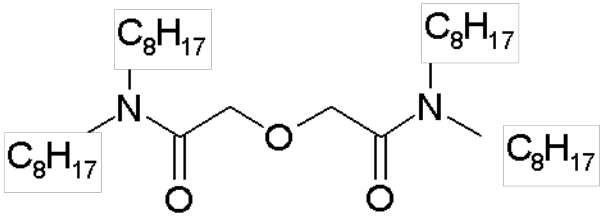
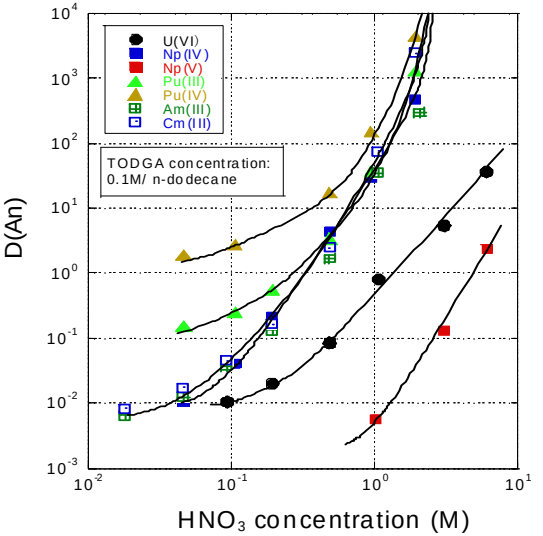
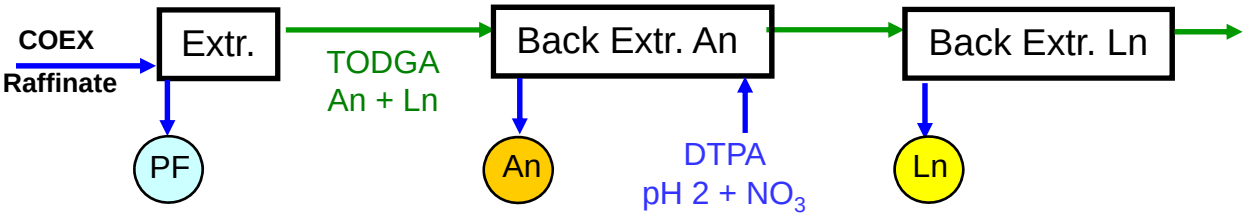
- **Co-extraction An (III) and Ln (III) with TODGA, using HNO_3 4N**





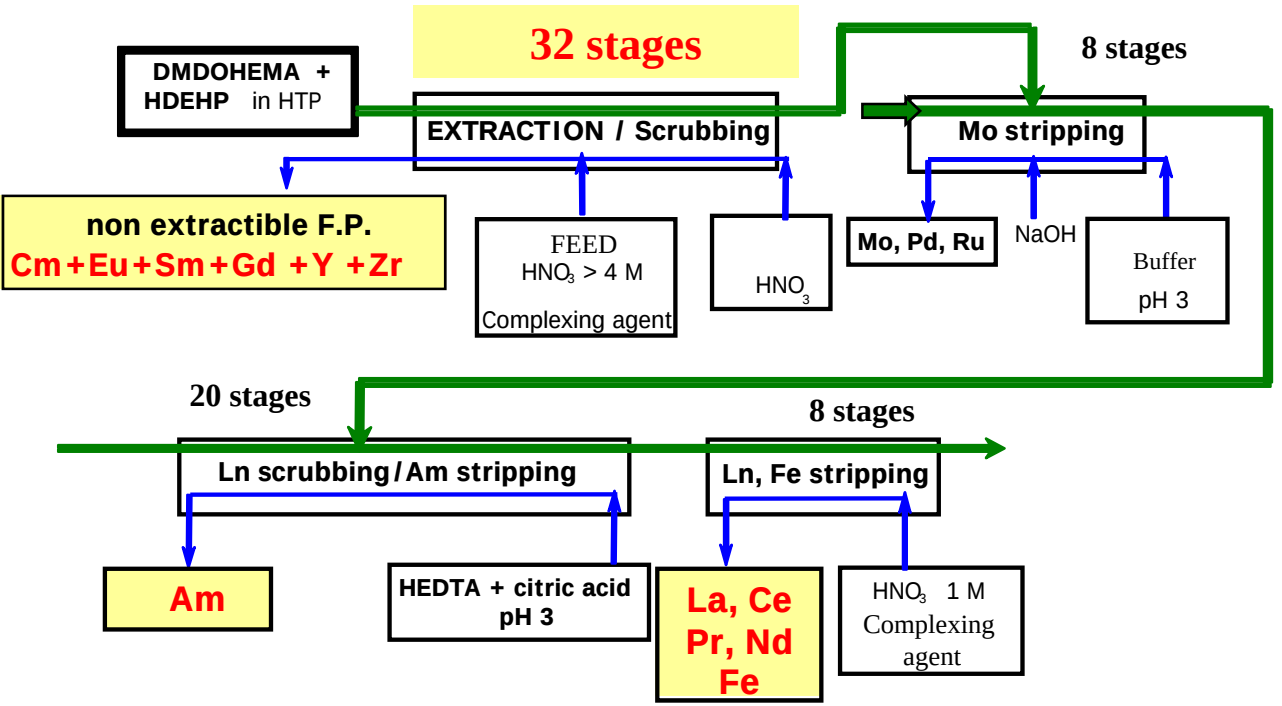
- **Selective back-extraction of An (III)**
  - **With polyamino-carboxylic hydrophile complexing agent**

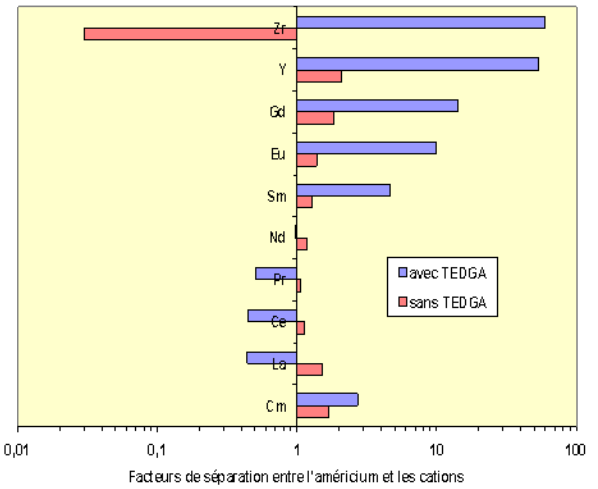


- **<u>Advantages</u> : simple scheme, TODGA synthesis low cost**
- **<u>Drawbacks</u> : high sensitivity of the Am-Cm back extraction step to pH and temperature**

Heterogeneous recycling: the EXAm process for Am-only recovery

- based on the DMDOHEMA diamide extraction properties, but Fs (Am/Cm)=1.6
  (patent FR 0955239, 07/2009)
- increase of the Am/Cm selectivity by the use of a complexing agent TEDGA in aqueous phase (48 stages down to 32 stages)
  (patent (FR 0955240, 07/ 2009)
- separation between Am and light Lns by the use of HEDTA complexing agent





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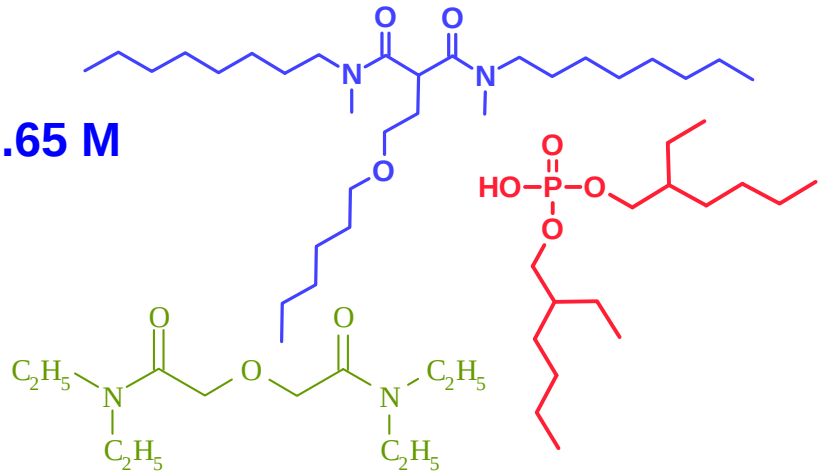
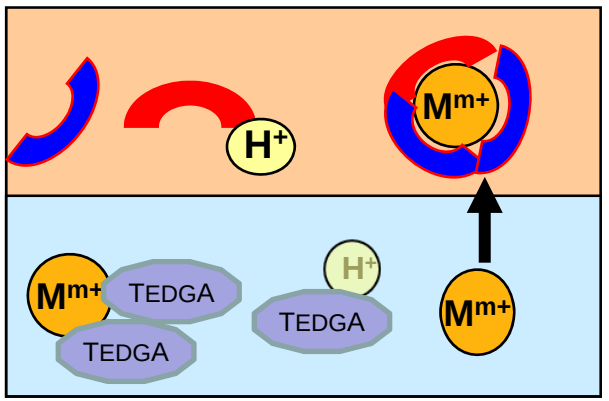
Complex chemical system :

- Organic phase **DMDOHEMA 0.65 M**
  HDEHP 0.3M
  Alcane

- Aqueous phase **TEDGA 0,3M**
  HNO_3 4.5M



Extraction modeling by different equilibria :



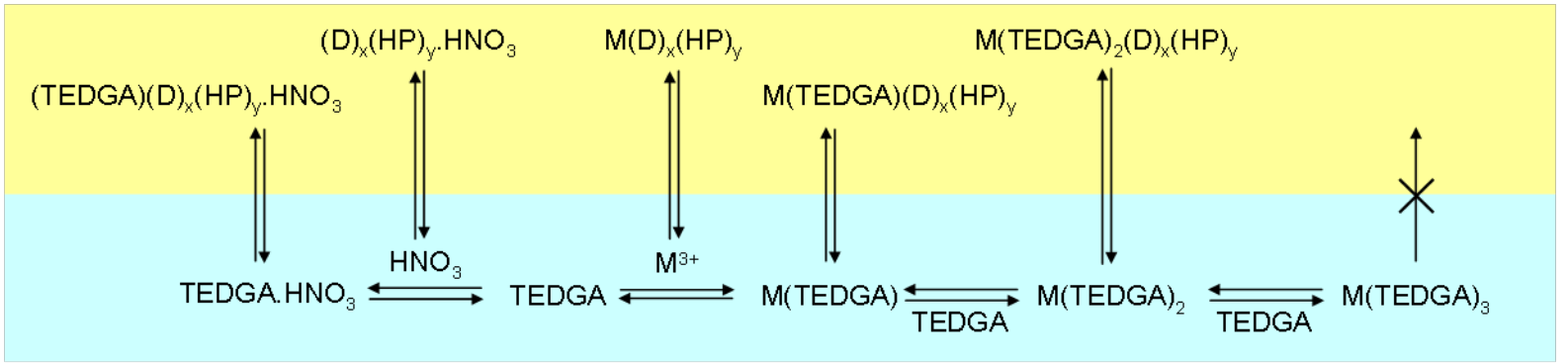
Extraction by diamide $(M(NO_3)_{\alpha1}(HNO_3)_{\alpha2}(D)_{\alpha3})$

Extraction by diamide + HDEHP $(M(NO_3)_{\alpha1}(D)_{\alpha2}(HA)_{\alpha3})$

Complexation by TEDGA $(M(NO_3)_{\alpha1}(T)_{\alpha2})$

Objective : modeling improvement by *better basic mechanism characterization and knowledge*

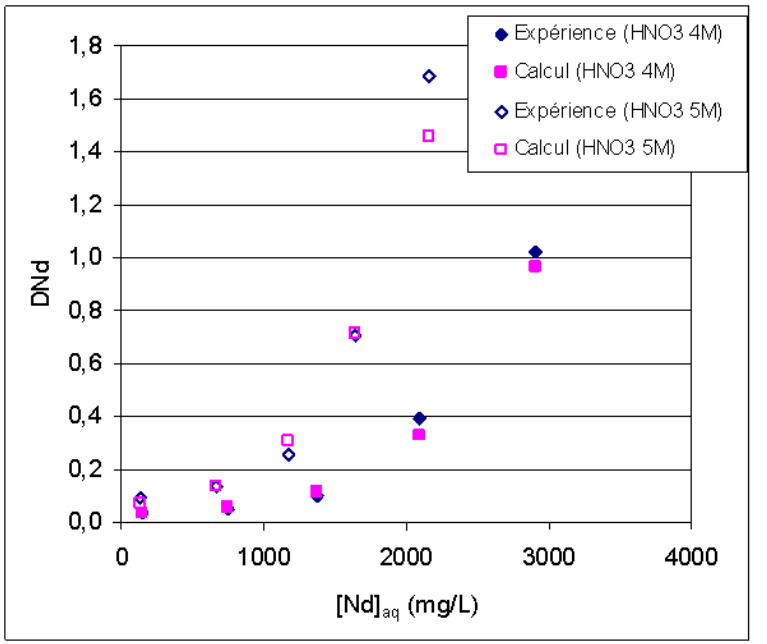
Modeling of extractant system with TEDGA



$$\left(M_j\right)^{\nu_j} + a_j\ NO_3^- + b_j\ TEDGA \Leftrightarrow \left(M_j\left(NO_3\right)_{a_j}\left(TEDGA\right)_{b_j}\right)^{\nu_j - a_j}$$

$$\beta_j = \frac{\left[\left(M_j\left(NO_3\right)_{a_j}\left(TEDGA\right)_{b_j}\right)^{\nu_j - a_j}\right]}{C_j^{libre} \cdot \left[NO_3^-\right]^{a_j} \cdot \left(C_n^{libre}\right)^{b_j}}$$

	La	Am	Nd	Cm	Eu
b_j	2,1	2,6	2,6	2,7	2,9
β_j	$9,7\ 10^3$	$8,9\ 10^5$	$8,9\ 10^5$	$1,1\ 10^7$	$2,3\ 10^7$



Modeling exemple for Nd

Development and validation of EXAm process

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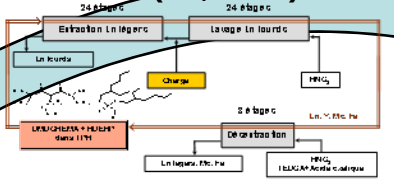
Lab studies
(2008-2009)

Modeling/ Flowsheet (T2, 2009)

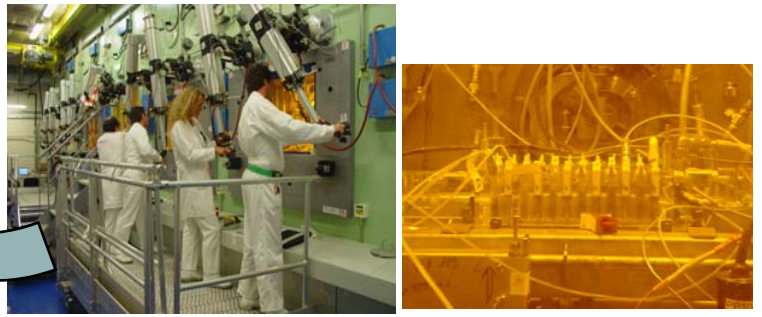


Inactive test (6/2009)

+ TEDGA

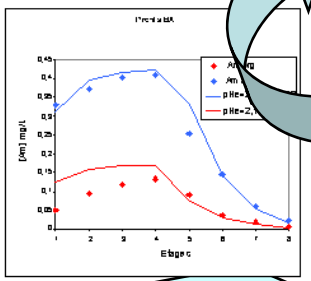
Lab studies
T3-4, 2009

Partial test (glove-box)
Simulated solution (12/2009)



Complete validation test

Genuine solution in CBP (04/2010)



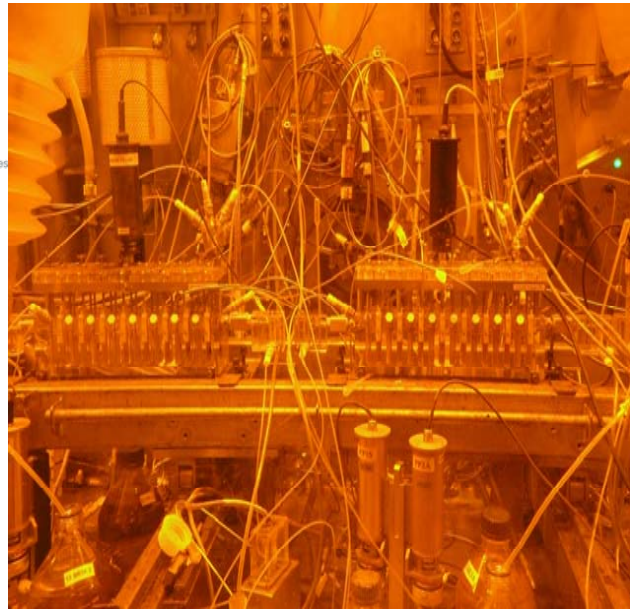
Modeling/flowsheet
Based on Lab and Batch tests
Test management (T1, 2010)

Assessment HA CBP test results
mid 2010

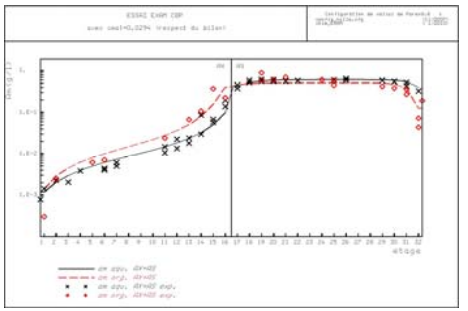
Validated process

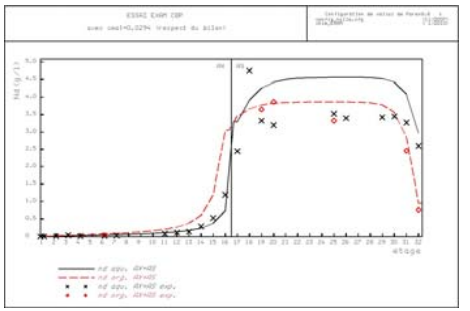
« Batch » test
Real genuine solution
Hot cell (01/2010)

HA EXAm Test in CBP-Atalante (04/2010)



- **~1 kg SNF dissolved**
- **~ 60 h test, with genuine solution**
- **spectrophotometer for on line Nd and Am measurements during the test ⇨ essential tool to follow the test operation**
- **~ several hundreds analytical determinations on samples for concentration profiles (to be compared to model)**





EXAm performances :

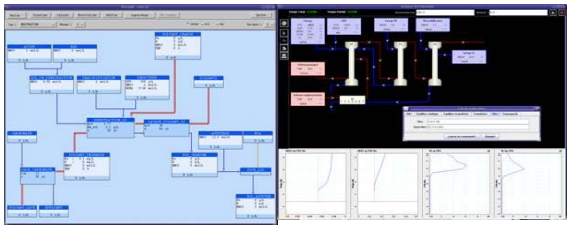
- **Decontamination Factor F_D (Cm/Am)** = $\dfrac{\left(Cm/Am\right)_{\text{Exam raffinate}}}{\left(Cm/Am\right)_{\text{Am Charge}}}$ = **1/505**

- **Am recovery ratio** = $\dfrac{(Am)_{\text{Charge}} - Amloss}{(Am)_{\text{Charge}}}$ = **98.5 %**

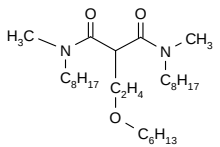
Process validated

- **Key steps : Am stripping ; Mo stripping to be improved**

Separation process : towards industrialization

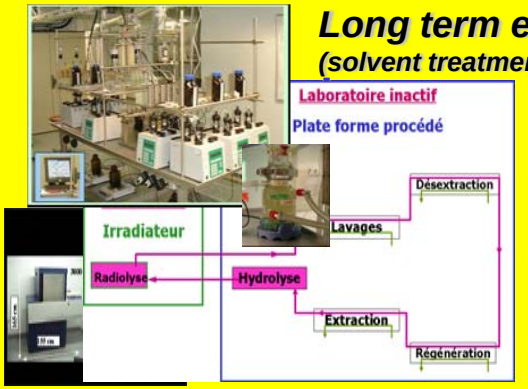


Modeling
(flowsheet, sensitivity, operation)

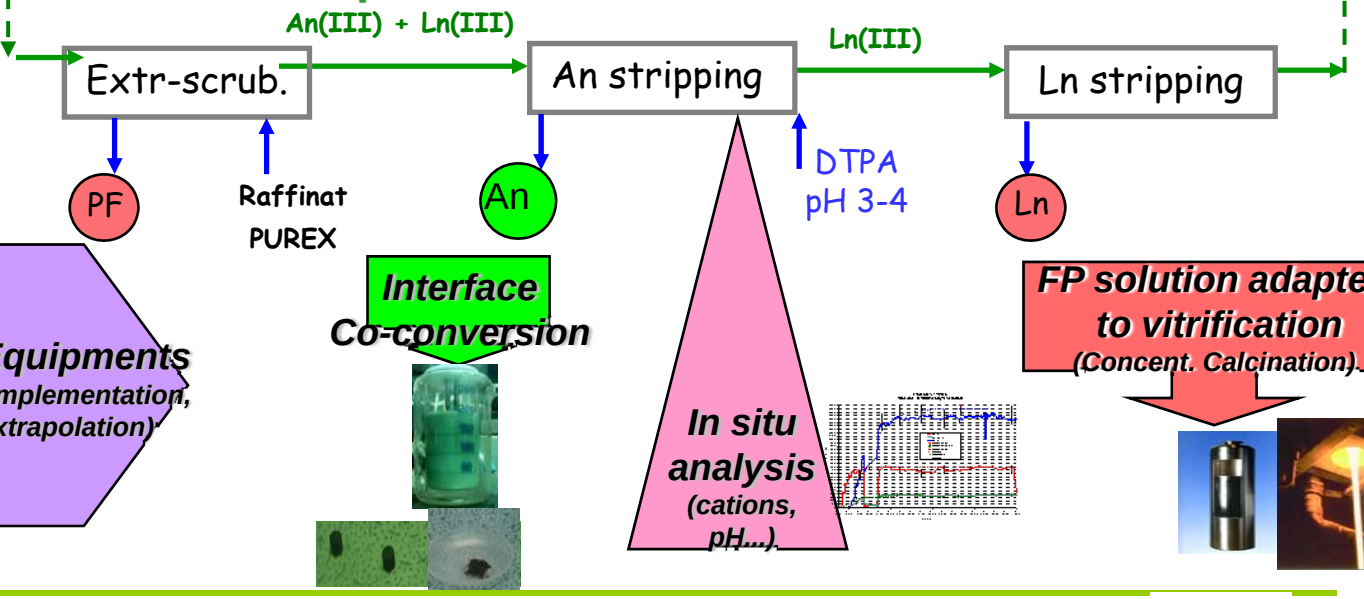


Extractant synthesis

Long term effects
(solvent treatment)



Core of the process demonstrated at lab scale



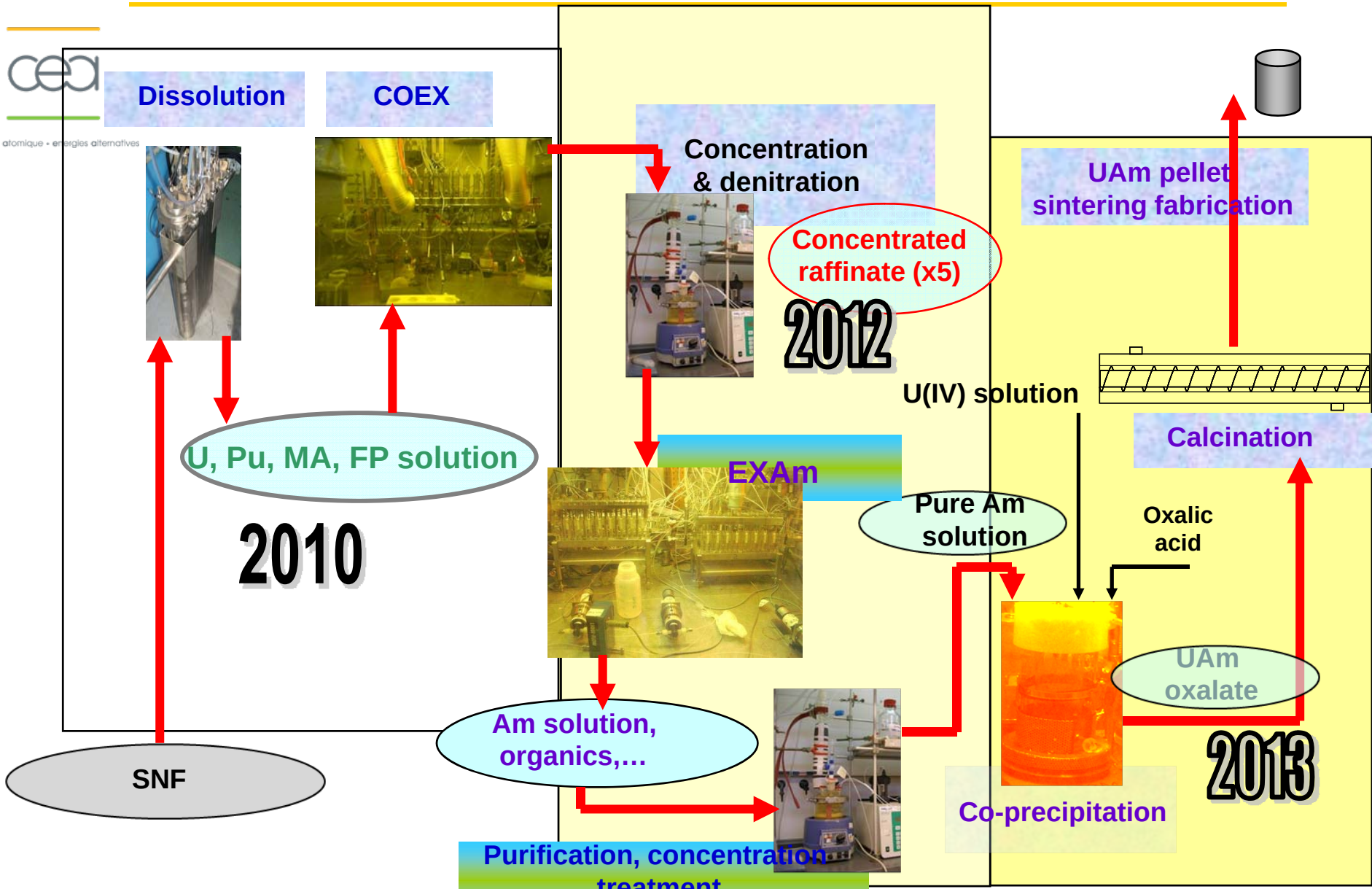
Equipments
(Implementation, extrapolation)

Interface
Co-conversion

In situ analysis
(cations, pH...)

FP solution adapted to vitrification
(Concent. Calcination)

Integral test of Am recycling: from SNF solution to UAm pellet



Towards 2012 milestone, and after

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- 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, with performances adapted to transmutation needs (purity,…)
- On-going research in the CEA Atalante facility, with international collaboration for optimizing separation process
  (EU ACSEPT, CEA-DOE-Japan GACID,...)
- 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 …