Minor Actinides Incineration Partitioning and Transmutation

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Future Nuclear Energy Systems

CEA/Nuclear Energy Division/Division of Nuclear Development and Innovation

Which goals?

Goal 1 research : solutions that would allow partitioning and transmutation of long lived radionuclides present in HL LL waste and reduce the overall radwaste quantity.

Goal 2 research : feasibility of reversible or irreversible repository in deep geological formations.

Goal 3 research : conditioning processes and feasibility of long term storage, above-grade or below-grade.

P&T research : goals and strategy (1/2)

⇨ Comply with objectives and obligations set in law and produce results in time to meet the 2006 deadline.

⇨ *for partitioning :*

• investigate partitioning processes within the timeframe allowed by law :

- hydro-metallurgy consistent with current industrial practice,
- and also pyrochemistry (a more pioneering venture)

• primary focus on RN either contributing the most to HL waste LL radiotoxic inventory in current vitrified waste (americium, curium, neptunium) or considered highly mobile within a geological environment (iodine, caesium, technetium)

P&T research : goals and strategy (2/2)

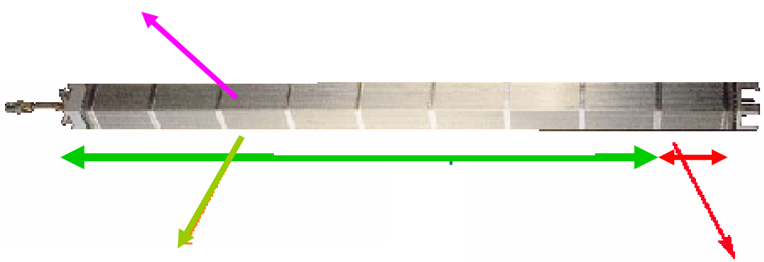
⇨ *for transmutation :*

- focus on transmutation of corresponding RN,
- increase knowledge and accuracy of nuclear database accordingly,
- look at fuel fabrication (homogeneous or concentrated) & ad-hoc materials for matrices,
- consider existing reactors (LWR and SFR Phénix) consistent with legal timeframe,
- consider future systems (ADS or new GEN IV FR),
- develop scenario studies of nuclear systems to shape out their relative transmutation potential in a national context.

Where does HL LL waste come from ?

Spent fuel reprocessing : what does it achieve?

Cladding & structural materials : medium level & long lived waste



Recyclable materials (95 %)

Uranium : 470 kg (94 %)

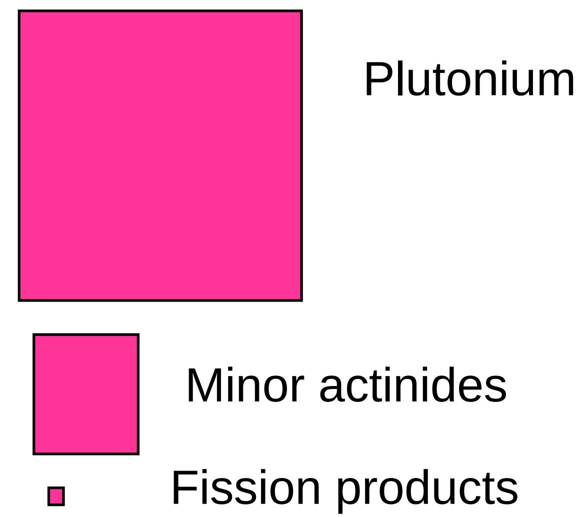
Plutonium : 5 kg (1 %)

Vitrified HL radwaste (5%)

Fission products : 25 kg (4 %)

Minor actinides : ~ 0,5 kg (0,1 %)

Current scheme for the PUREX process



Relative contribution to total radiotoxicity after 1 000 years

Goal 1 research

CEA alone, as research leader, has mobilised significant financial means : 708 M€ out of

810 M€ (to the end of 2004).

- **A large involvement of very many teams and a strong contribution from academic expert groups.**
- **Significant results obtained**.

- A continuous improvement process :

  ⇨ opening the scope of possible solutions.

Partitioning

Goal : separate HL LL nuclides,

- ⇨ **to allow for their recycling,**
- ⇨ **or reduce their half-life and therefore the associated long term risk.**

Partitioning : key nuclides

⇨ minor actinides : Am - Cm - Np

major contributors to long term radiotoxic inventory;

⇨ fission products : I - Cs - Tc

- long lived isotopes significantly present in spent fuel,
- with a potential for mobility within rock formations;

⇨ processes : hydro-metallurgy (consistent with PUREX) and pyro-chemistry (more innovative);

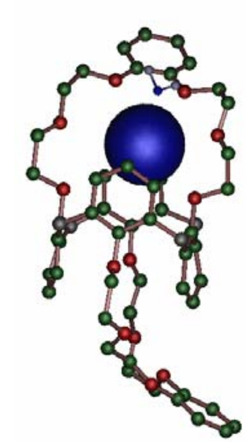
⇨ partitioning of nuclides present in the already vitrified HL waste would be rather difficult.

Research steps

⇨ **Fundamental research : a wide co-operative framework**

- **exploration : new extracting systems ;**
- **fundamentals : in-depth study of mechanisms at work ;**

A few hundreds of new molecules



⇨ **Applied research :**

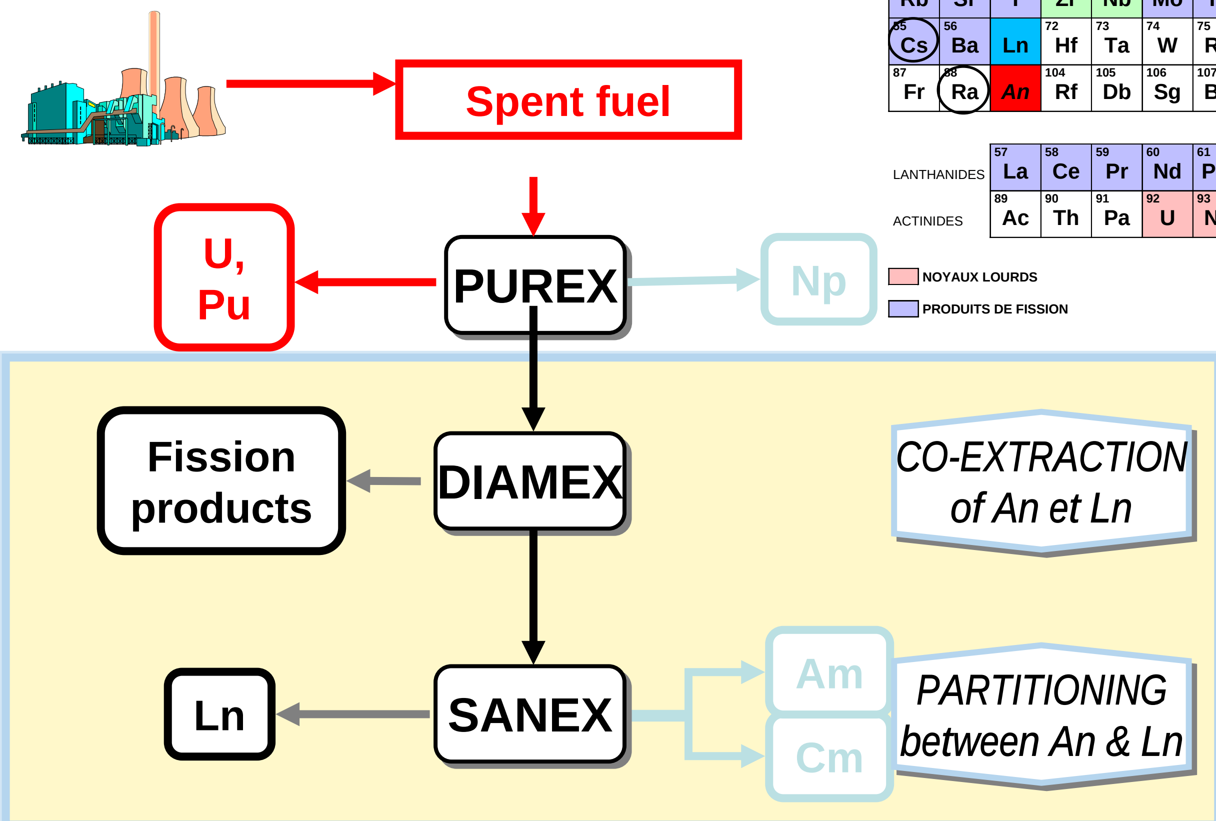
- **process design ;**
- **lab experiments on actual spent fuel material ;**
- **« demonstration » experiments : integration, representativeness, long-lasting performance ;**

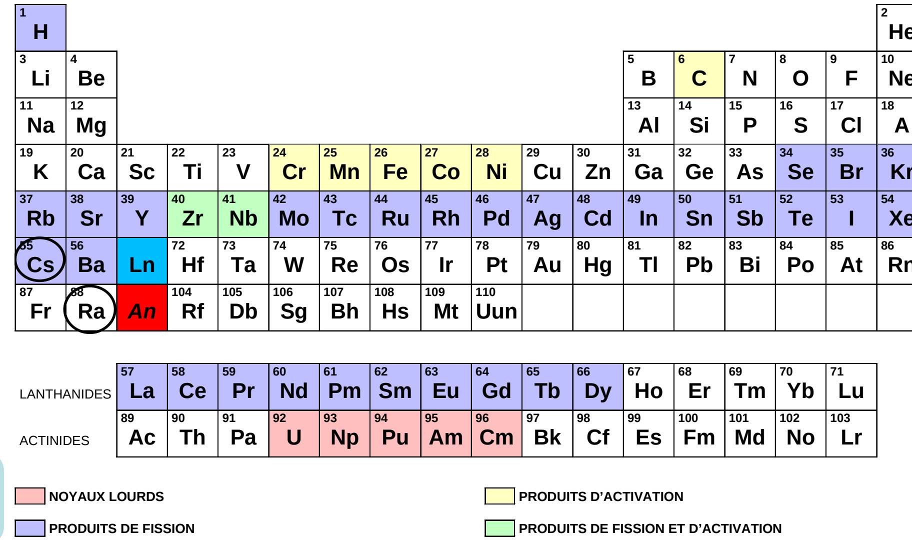
Scale : 1/10000

Scale : 1/100 à 1/1000

⇨ **A true challenge : a sophisticated partitioning chemistry under highly radioactive conditions.**

Actinides partitioning : research path used





R&D in the ATALANTE facility

From fundamental research to demonstration experiments …

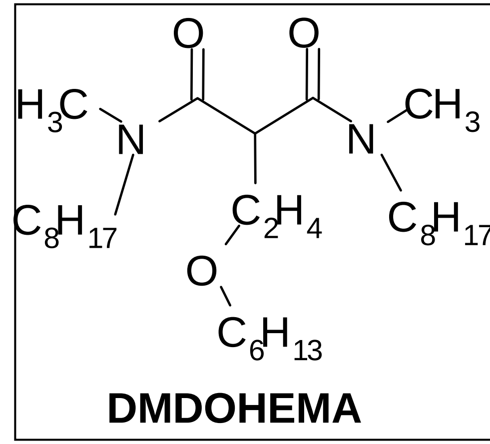
… up to making use of a few kilograms of actual spent fuel material.

Am, Cm, Np, Tc, I et Cs partitioning : lab scale results

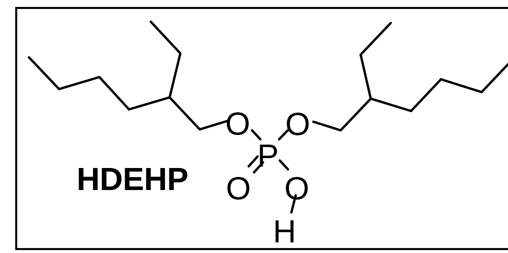
Americium & curium : recovery ratio up to 99,9% for Am & 99,9% for Cm ;

⇨ long-lasting performance tested (« accelerated » simulation within an irradiation loop) ;

⇨ confirmation test with the use of « industrial » technology scheduled in 2005 ;



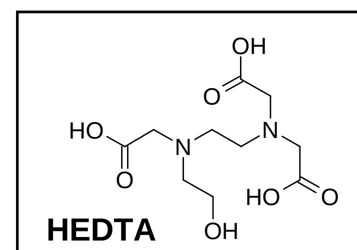
Neptunium : recovery ratio up to 99% ;



Technetium : recovery ratio from 45 à 90%;

Iodine :

- recovery ratio > 97% with PUREX;
- additional recovery up to ~ 99% possible ;



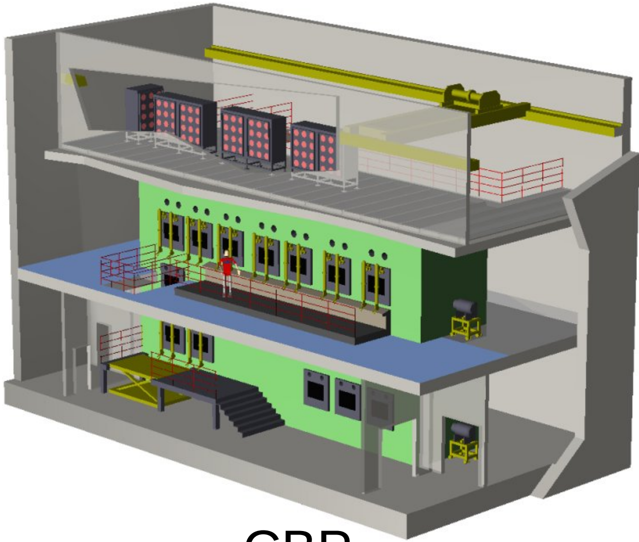
Caesium : recovery ratio > 99,8%, with the use of the calixarene extractant.

Conclusion …. for partitioning R&D

⇨ A large scope for research : *from fundamentals to applications;*

- the actinides: the key target;
- a scope wide open : *(numerous options explored and co-operations);*
- feasibility of pyro-chemical processes not yet demonstrated;
- molecules and partitioning processes successfully tested at the lab scale;
- a further step scheduled in 2005 within the Atalante CBP hot cell *(to replicate an industrial-like set-up);*
- a research booster for partitioning and actinides chemistry;
- further steps will be required prior to any industrial implementation.

Atalante in Marcoule



CBP

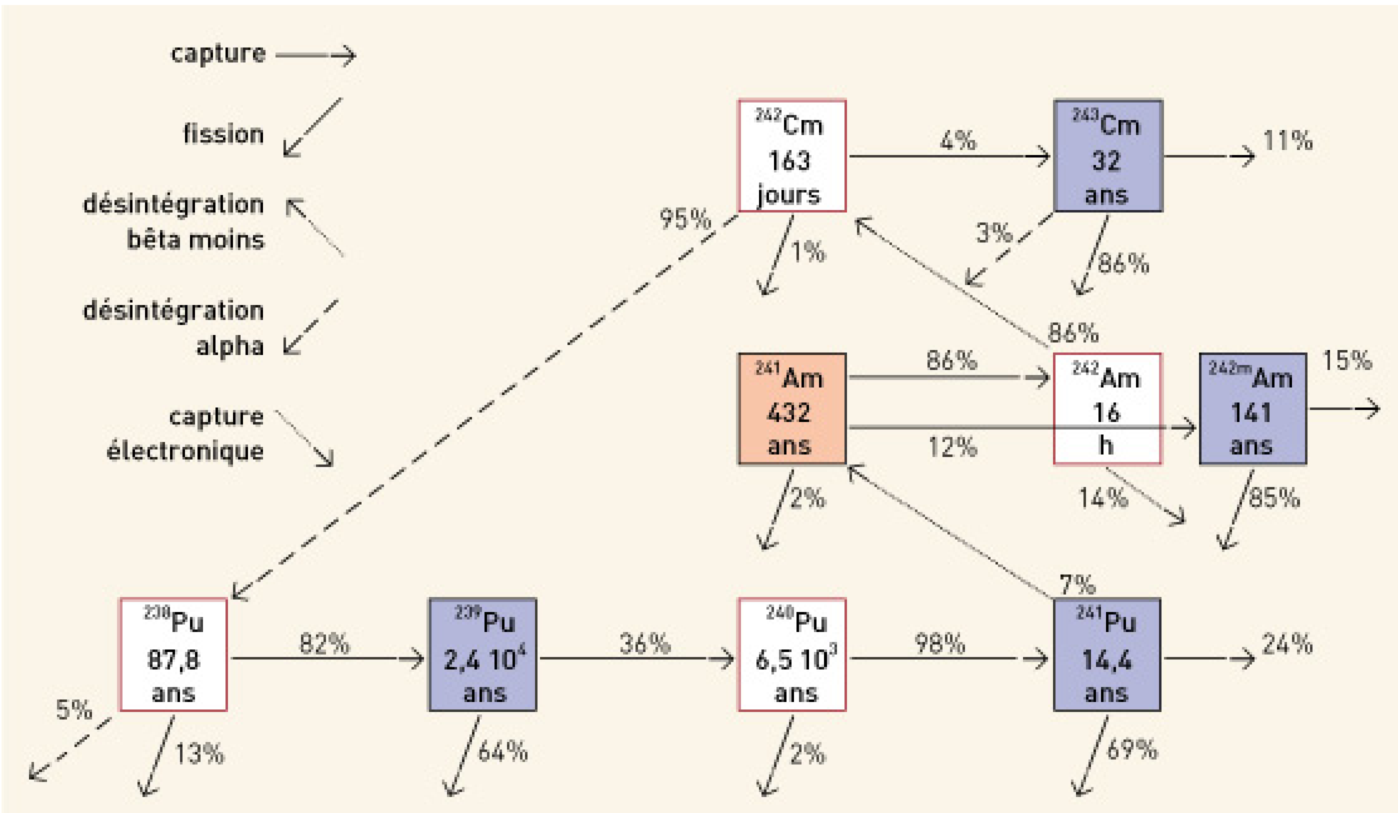
Transmutation

Goal : turn some of the nuclides included in radwaste qualified today as ultimate waste (vitrified) into fuel material to morrow.

- ⇨ Capture & fission;
- ⇨ Systems for transmutation;
- ⇨ Nuclear fuels for transmutation.

Capture & fission

⇨ Making use of neutron flux (within a reactor) to transform LL radionuclides into stable or short-lived isotopes through neutron capture or fission reactions.



For actinides, fission reaction, not neutron capture, must be considered.

For fission products, neutron capture reactions are observed resulting in stable or short-lived nuclides.

Transmutation output according to neutron spectrums used

R&D on basic nuclear data :

⇨**Nuclear data obtained on actinides cross-sections is used to rank the transmutation potential of various systems.**

Results obtained using the homogeneous mode

	PWR MOX Flux 2,5 10^{14} n/cm^2.s Ts : 1500 JEPN	SFR Flux 3,4 10^{15} n/cm^2.s Ts : 1700 JEPN
Isotopes	Fission	Fission
^{237}Np	4%	24%
^{241}Am	10%	24%
^{243}Am	6%	15%
^{244}Cm	16%	27%

- **Two recycling modes have been considered : homogeneous mix within the nuclear fuel or heterogeneous in targets at higher concentrations;**

- **There is obviously a net advantage in using a fast neutron flux to get higher fission rate, however increasing the fission rate requires multiple recycling ;**

⇨**Fast neutron flux and spectrum are more efficient to transmute actinides through fission reaction.**

Transmutation (Present and Future Technologies)

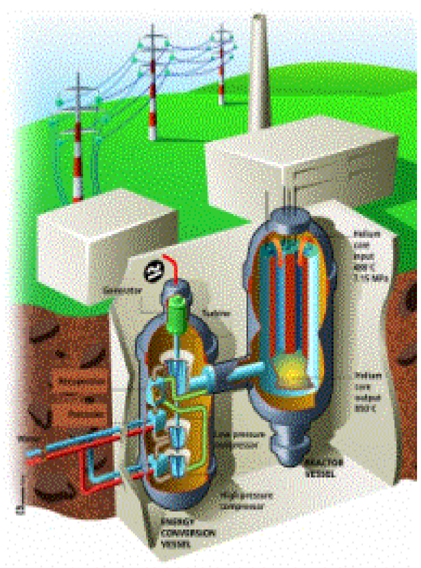
Fast neutrons reactors

Light water reactors

New perspectives……….. …………….

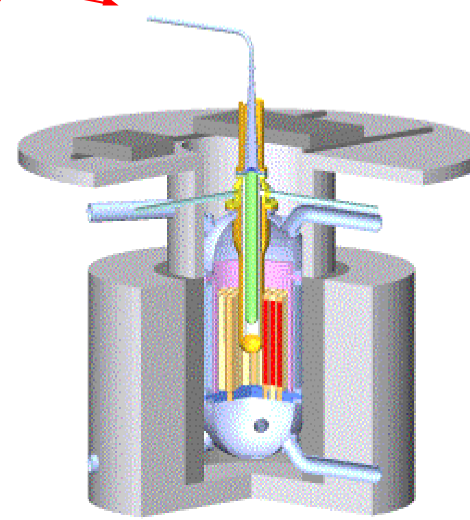
Advanced gas cooled reactors (fast neutrons)

Generation IV



Hybrid reactors

Proton beam



Transmutation systems

1. In PWR : the feasibility of americium transmutation is recognised, however:
   - with an increased consumption of enriched uranium;
   - and the production of a large quantity of curium, not recyclable in PWR.
2. In fast neutron reactors : the feasibility of minor actinides transmutation is recognised without impeding the usual reactor operating mode :
   - homogeneous mode in SFR : concentration of minor actinides up to a max of 2,5 % (safety,…);
   - homogeneous mode in GFR (first trend) : concentration of minor actinides up to a max of 5 % (behaviour of fuel under irradiation is the limiting factor);

⇨ Fast neutron reactor can help eliminate minor actinides produced by the PWR fleet and manage those that they produce ;

⇨ ADS : 5ème and 6ème EC FP, collaboration between CEA and CNRS :
   - significant R&D work has been conducted or is on-going,
   - the ADS concept is quite complex, however no stumbling stones that would preclude the demonstration of the feasibility of one experimental reactor have been identified so far,
   - yet, challenging aspects are not resolved and have been pointed out.

Accelerator Driven Systems (ADS)

- **Sub-critical core physics** *(MUSE in MASURCA with the CNRS)*
- **Spallation source** *(MEGAPIE at PSI)*
- **Accelerator/reactor coupling** *(TRADE → ?)*
- **System studies** *(PDS-XADS in the European 6th Fwk Pgm)*
  (XADS Pb-Bi 80 MWth, Myrrha Pb-Bi 50 MWth et XADS He 80 MWth)
- **TRU Fuel and targets** *(20-50 % MA)*
  Irradiation tests in Phenix *(Camix, Cochix, Futurix/FTA…)*

Milestones:

2008 - Synthesis report on the technical and financial viability of power ADS for industrial transmutation *(EUROTRANS)*

Collaborations :

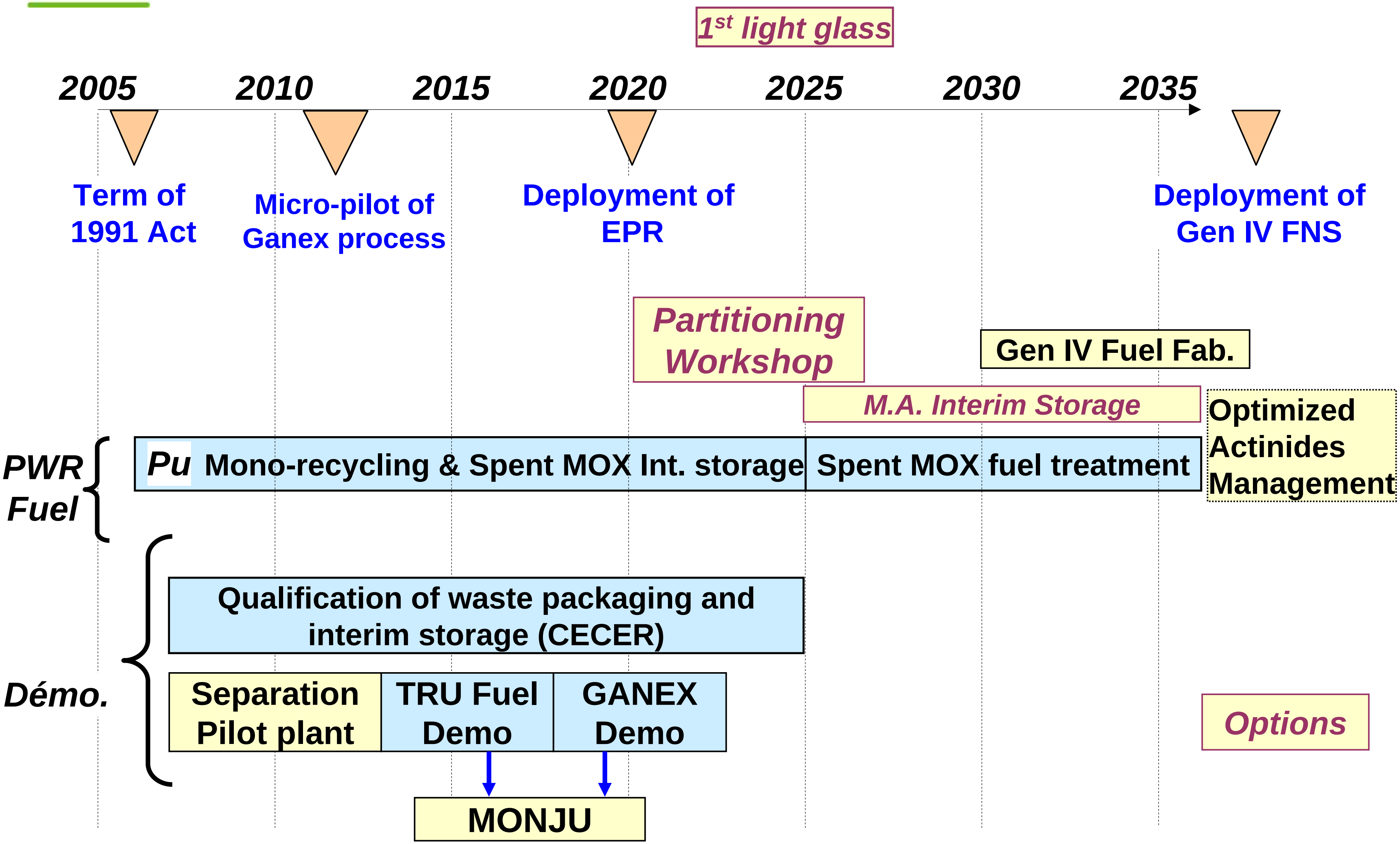
- **CNRS, Areva**
- **Europe** *(5th & 6th PCRD…)* **+ Japan (***JAERI),* **United States** *(DOE/AFCI)*

Minor actinides transmutation with fast neutrons

	SUPERPHENIX		PHENIX	
	Homogeneous	Heterogeneous	Homogeneous	Heterogeneous
Np	**NACRE** **UO_2,PuO_2 + 2% Np** **Fabricated** **Not irradiated**		**SUPERFACT** **pins** **UO_2 + 2% NpO2**	**SUPERFACT** **pins** **UO_2 + 40% NpO_2** **METAPHIX** **Np, Am, Cm**
Am **(Cm)**	**Few pins** **with 1% Am**		**SUPERFACT** **pins** **UO_2 + 2% AmO2**	**SUPERFACT** **pins** **UO_2 + 20% AmO_2** **ECRIX** **AmO_2 + MgO** **CAMIX/COCHIX** **FUTURIX FTA**

Ended | **On-going** | **In preparation**

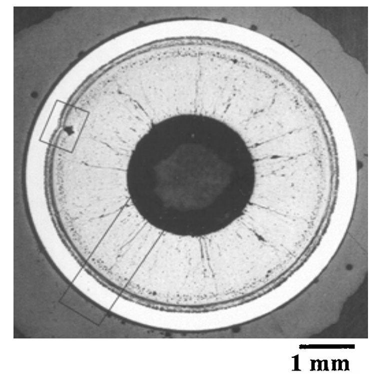
Main technology demonstrations



Nuclear fuel for transmutation : current R&D programmes

⇨ Reactors in use or considered :
- irradiation reactors : HFR,SILOE;
- fast neutron research reactors :
  - PHENIX,
  - JOYO, MONJU (Japan),
  - BOR 60 (Russia).

⇨ Design and fabrication of experimental fuel, and associated safety case ;
- irradiation (minimum 5 years),
- non destructive and destructive examinations,
- 35 irradiation experiments have been conducted or are on-going.



Results validation :

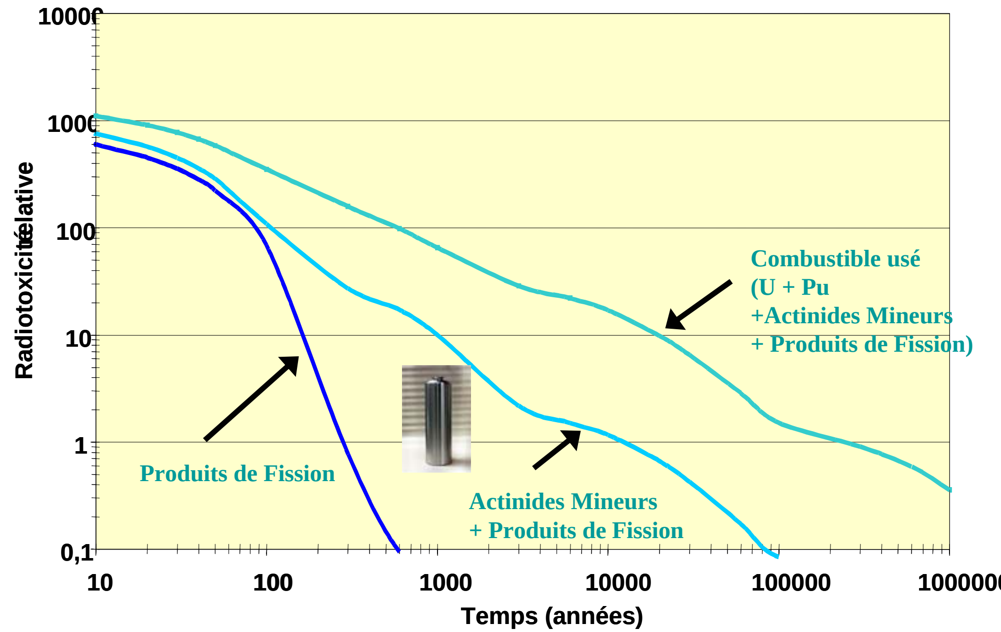
- actinides concentration and burn-ups;
- fission rate for one cycle :
  i.e. 70% of the americium contained in one target has undergone fission.

Conclusion … for transmutation (1/2)

⇨ the feasibility of transmuting actinides such as Am and Np is recognised in fast neutron reactors according to experiments run in SFR;

⇨ ADS systems are quite complex, and their technical and economical viability for actinides transmutation is yet to be proven;

⇨ transmutation of technetium 99 is feasible, but not efficient;
transmutation of caesium 135 is no longer pursued;
transmutation of iodine 129 is not currently feasible.



⇨ Reducing the ultimate waste radiotoxic inventory is possible with implementing a continuous improvement step-wise process;

⇨ Goal 1 cannot apply to current ultimate waste (vitrified).

⇨ An international expert peer-review report will be made available by the end of 2005

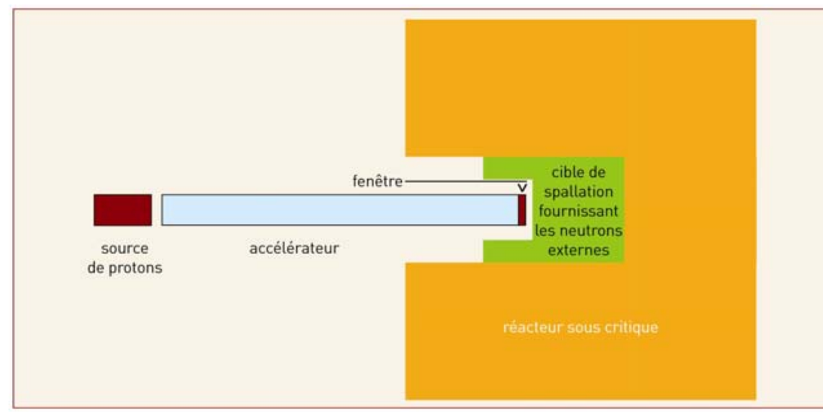
Conclusion … for transmutation (2/2)

⇨ **The scientific feasibility of transmutation is recognised;**

⇨ **Fast neutron spectrum is best for transmutation.**

Transmuting systems producing energy

Transmuting systems supplementing producing energy systems

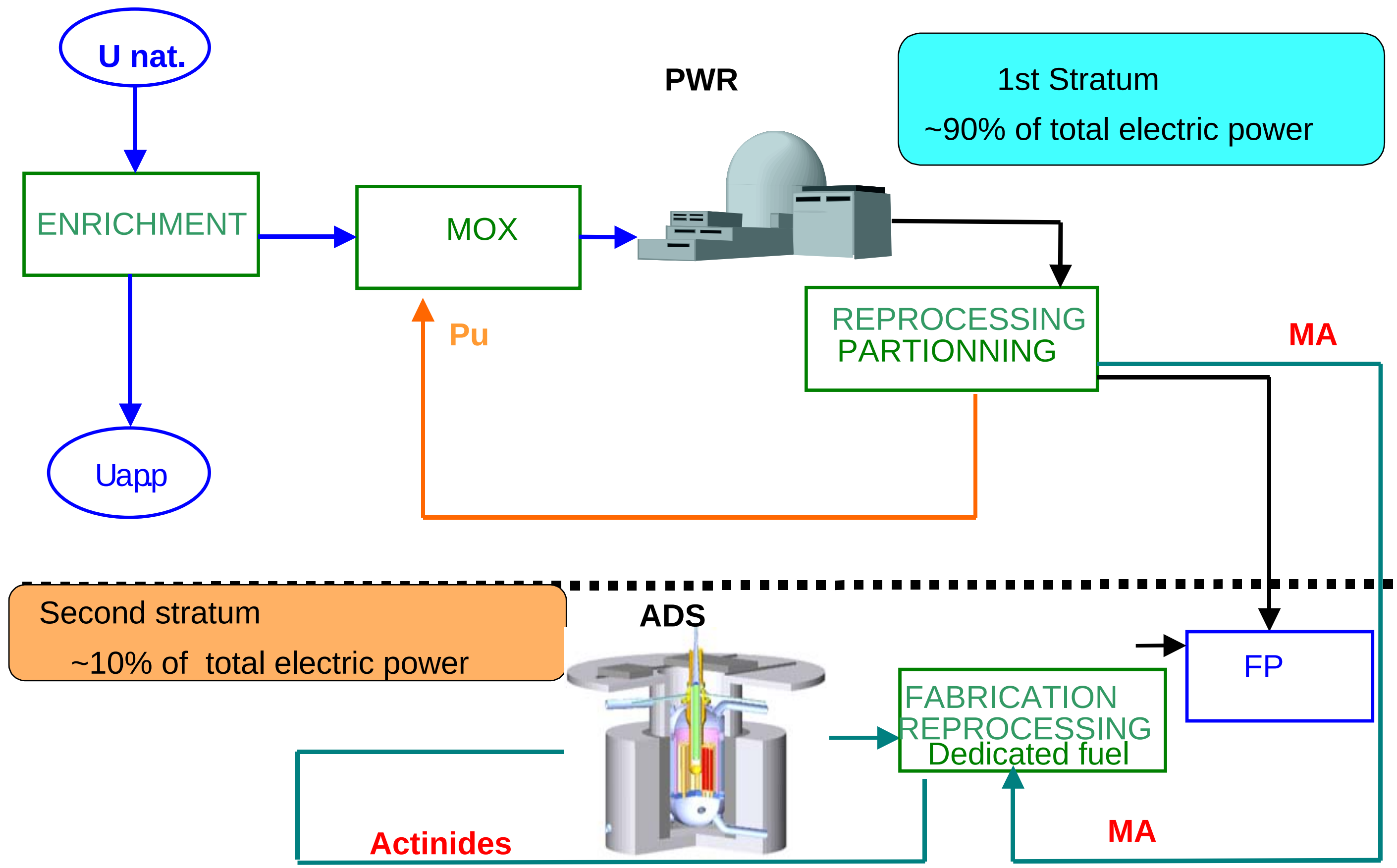


⇨ **Selection of the transmuting system by 2015.**

Scenarios : outlines

- Introduction
- Analysis of preliminary steady-state scenarios
- Transition Gen II – Gen III -> Gen IV systems (fuel cycle and reactors)
  - Main results
  - Analysis
- Conclusions

Double strata scenario with ADT



Preliminary scenarios evaluations **at steady state** : case of the partitioning / transmutation scenarios

P&T options :

- Comparison of different options
- Homogeneous / Heterogeneous
- Natural uranium ressources
- Annual mass flux in fuel cycle facilities
- M.A. inventories

Reactor	**PWR**			**FR**			**PWR + ADS**	**PWR + FR +ADS**
Fuel cycle type	***Once-through cycle***	***Pu recycling***	***Pu+MA recycling***	***Pu recycling***	***Pu+MA recycling***		***Pu + MA recyling***	
Pu recycling	*No*	*Yes*	*Yes*	*Yes*	*Yes*	*Yes*	*In PWR*	*In PWR and FR*
M.A. recycling	*No*	*No*	*Yes*	*No*	*Together with Pu*	*In target subass.*	*In ADS*	*In ADS*
Front-end facilities (needs per year)								
Nat Uranium (tons / year)	8360	7320	7600	-	-	-	7580	5400
Fabrication capacities (t/y)	820	820	820	340	340	340 FR 2 target mod.sub.	730 UOX 8,6 ADS	540 UOX , 90 PWR MOX 60 FR 1 ADS
Reactors – Fuel type								
% PWR UOX	100			-	-	-	89	66
% MOX	-	100	100	100 (FR)	100 (FR)	100 (FR)	-	30
% ADS	-	-	-	-	-	-	21	5,2
Back end facilities (needs per year): Processing and re-fabrication facilities								
Processing capacities (t/y)	0	820	820	340	340	340	740	690
Storage (annual masses) – 60 GWe/year – 400 TWHe								
TRU stored (t/y)	11,8	3,3	0,03	2	0,06	0,2	0,07	0,05
Gain (related to Once through)	1	4	400	6	200	60	170	240

Scenarios at steady state

- P&T strategies :
  - Efficient mass reduction in the geological repository
  - Some strong impacts on the main fuel cycle parameters, mainly at the fabrication step :

Fuel type	**PWR**		**FR**			**ADS**
Fuel cycle management	***MOX as reference***	***Pu+MA recycling***	***Pu-only recycling***	***Pu+MA recycling***	***MA target subass.***	***MA recyling***
At fabrication step						
Decay heat (W g^{-1} H.M.)	1	x 3	0,5	x 2,5	x 80	x 90
Neutron sources (n s^{-1} g^{-1} H.M.)	1	x 8 000	1	x 150	x 5 000 / x 10 000	x 20 000

iHM: initial Heavy Metal

In the frame of the french industrial situation, the choice of

F.N.R. as long term sustainable systems

allowing a reduction of TRU masses in the waste was proposed

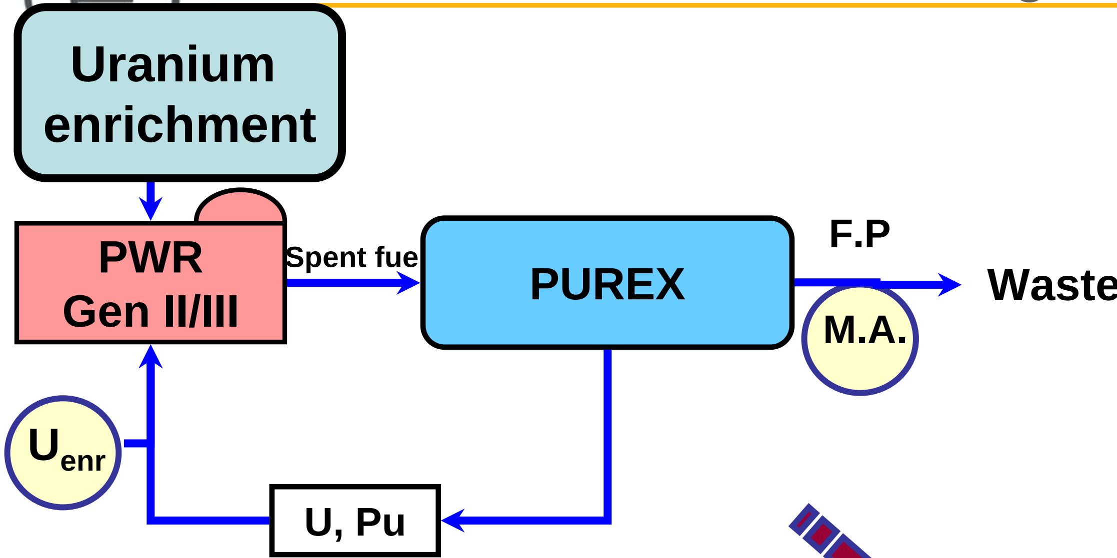
Americium and Pu recycling in GEN III system (EPR)

- Only Americium in PWR reactors :
  - Cm recycling produces Cf, very considerable source of neutrons and induces large difficulties on fuel cycle

  (The neutron source is multiplied by a factor 7900 in relation to the MOX case and creates both criticality and radioprotection difficulties.)

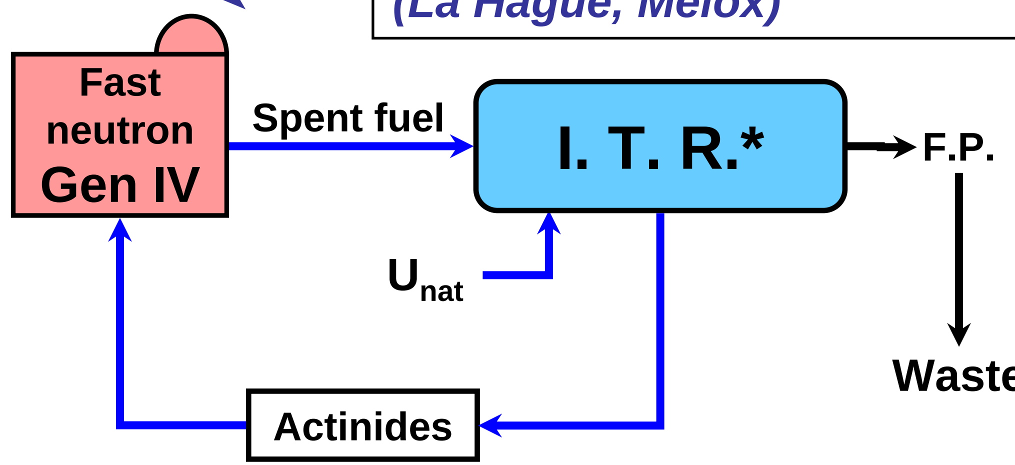
  - Np was not considered, because its recycling doesn't give important benefit on radiotoxicity or heat decay, but it could be add up.
- Concept used :
  - MOX UE (~8 % Pu, ~1% Am, ~5% U5) with higher moderation ratio
- Interim storage is necessary for Cm and Np

Transition scenarios between generations : fuel cycle facility



- Natural uranium resources
- Uranium enrichment
- Mono-recycling of Plutonium
  20 PWR - 900 30 % MOx
- Progress in ultimate waste form *(mass of Actinides, radiotoxicity, decay heat…)*
- Maximum utilization of existing fuel cycle plants *(La Hague, Melox)*

- Management of the (U,Pu) stockpile to deploy 4th generation fast neutron systems (> 2035)
- Recycling of MA (U, Pu, MA)
- Integral recycling of Actinides in fast neutron 4th systems
- Non proliferation

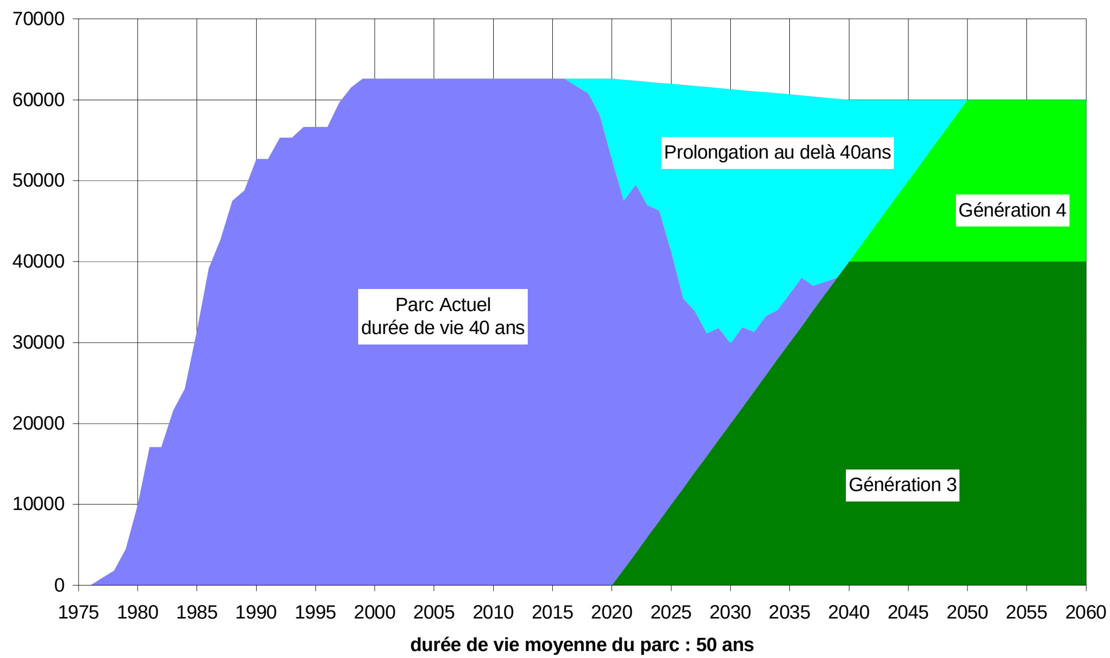


***I.T.R. : Integrated treatment & re-fabrication**

Scenario for the renewal of power reactors in France *(EDF)*

- **Major role of LWRs over the 21st century**
  - Operating PWRs (Gen II): lifetime extension (> 40 years)
  - Gen III/III+ PWRs: relacement of current PWRs around 2015 – Operation over most of the 21st century
- **~2040 – Transition from PWRs to Gen IV Fast neutron systems**

Source: EDF ENC 2002

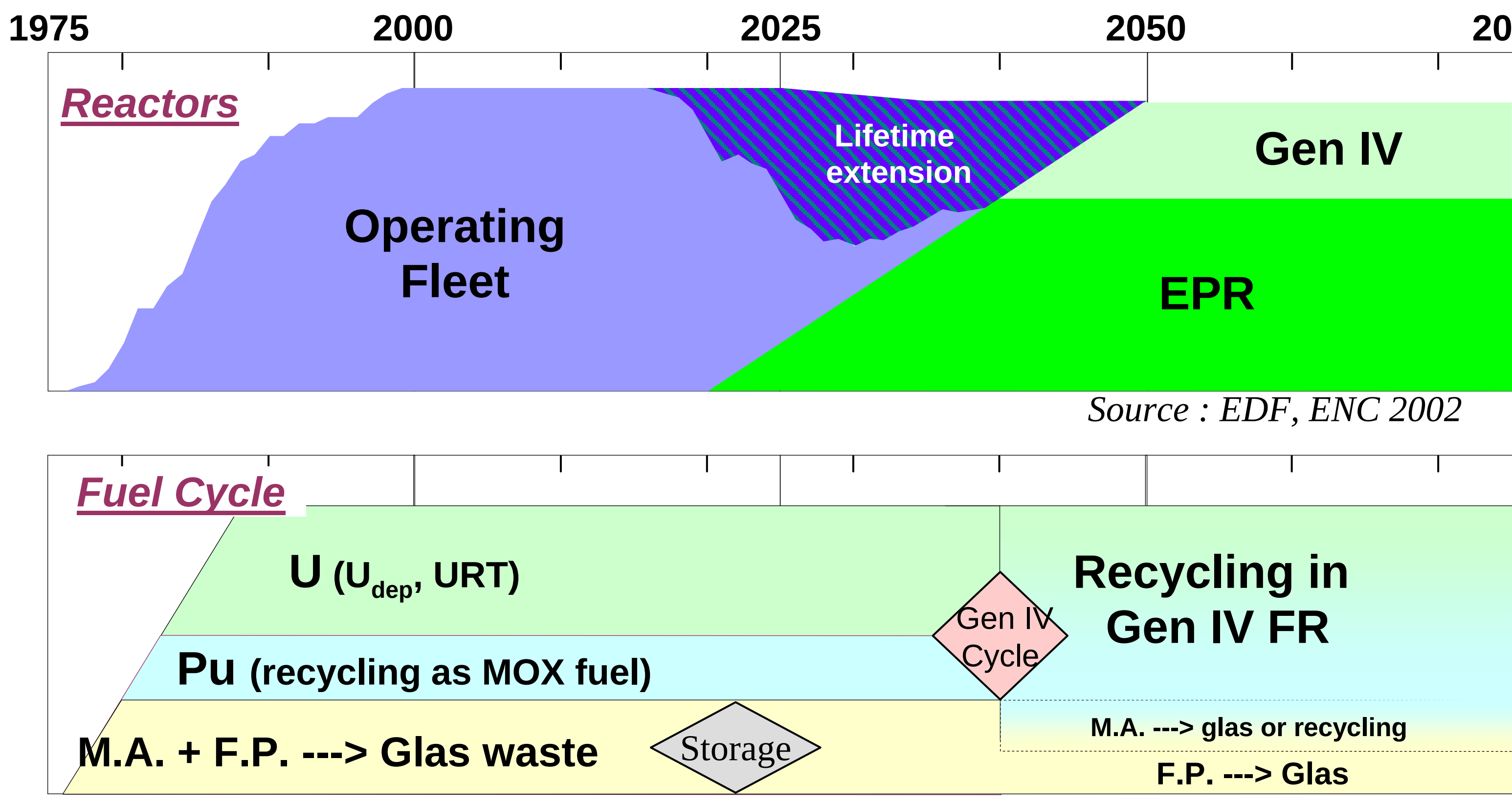


Introduction of Fast Reactors and New Fuel Cycle Plant around 2040

2040: - Deployment of Fast neutron systems *(SFR or GFR)*

- New spent fuel treatment plant (Ganex) – 2 options:

- **Recycling of U-Pu and separate management of MA** *(to waste or interim storage)*
- **Integral recycling of U-Pu-MA**



Delayed deployment of Fast Reactors (> 2050)

2040: New spent fuel treatment plant at La Hague (Ganex) :

- U-Pu (Co-management) for mono-recycling of Pu in PWRs with MA to waste

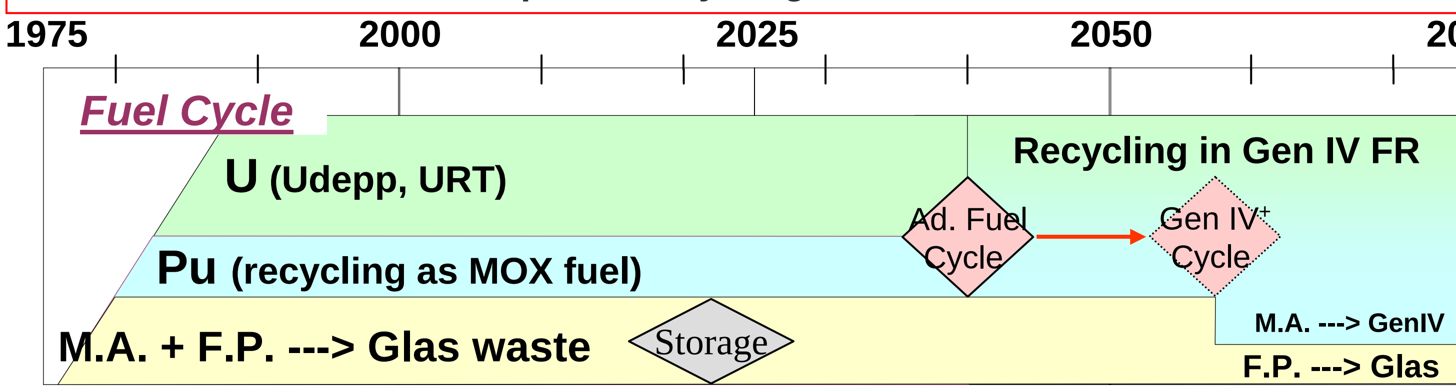
> 2050: Deployment of Fast neutron Systems (2080 in our study)

- U-Pu (co-management) for multi-recycling of Pu in Fast Reactors with MA to waste

- U- Pu-AM (co-management) for integral multi-recycling in Fast Reactors when deployed with, for the interim period :

- Single Pu recycling in PWR

- Multiple Pu recycling in PWR



** +: Addition of specific needs of Gen IV systems (front end, re-fabrication workshops)*

Main results (LWRs up to 2070)

Inventories (t)	One recycling Pu (MOX) **Alternative 1**			Multiple recycling Pu (MOX-UE) – **Alternative 2**			Once-through cycle (UOX)			
	2035	2050	2070	2035	2050	2070	2035	2050	2070	2100
Natural U (annual values – aggregates)	**7400 / 410 10^3**	**7500 / 520 10^3**	**7500 / 670 10^3**	**7160 / 410 10^3**	**7100 / 520 10^3**	**7000 / 660 10^3**	**8360 / 420 10^3**	**8360 / 550 10^3**	**8360 / 720 10^3**	**8360 / 970 10^3**
UTS (annual, M SWU/yr)	**5,8**	**5,8**	**5,8**	**5,3**	**5,1**	**5,1**	6.4	6.4	6.4	6.4
Pu (Total)	**396**	**479**	**596**	**373**	**398**	**400**	**474**	**612**	**793**	**1062**
MA (Total)	**76**	**120**	**178**	**76**	**125**	**191**	**99**	**138**	**191**	**271**
% fuel with TRU in fleet	12%	10%	10%	23%	26%	33%	0%	0%	0%	0%
TRU in storage	**389**	**529**	**703**	**128**	**175**	**240**	**573**	**750**	**984**	**1333**

Pu recycling in PWR :

-10% of saving natural ressources compared to once-through cycle

- Equilibrium of the Pu inventories
- Increase in MA inventories

Main results (FRs, introduced at 2035)

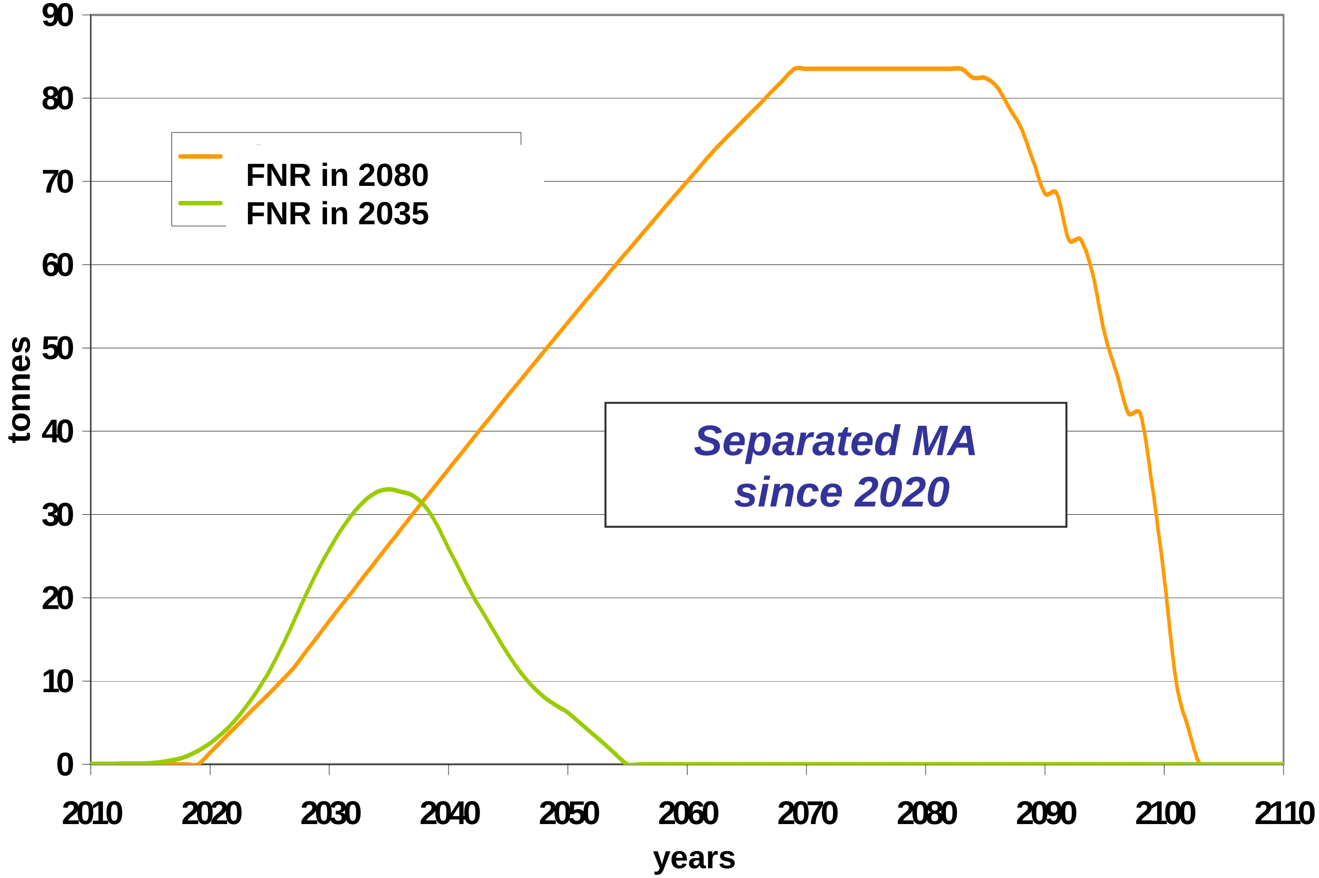
	One recycling of MOX in PWR and Pu recycling in 4th generation FR system (SFR), M.A. disposed in storage				One recycling of MOX in PWR and global multiple recycling (Pu, Np, Am, Cm, ...) in 4th generation FR system (SFR)				One recycling of MOX in PWR and global multiple recycling (Pu, Np, Am, Cm, ...) in 4th generation FN (GFR) system			
Inventories	2035	2050	2070	2100	2035	2050	2070	2100	2035	2050	2070	2100
Natural U (annual values – aggregates)	7850 / 430 10^3	4200 / 510 10^3	4200 / 600 10^3	0 / 660 10^3	7850 / 430 10^3	4200 / 515 10^3	4200 / 600 10^3	0 / 660 10^3	7850 / 430 10^3	4200 / 515 10^3	4200 / 600 10^3	0 / 660 10^3
UTS (annual, M SWU/yr)	5,9	3,3	3,2	0	6	3,2	3,2	0	6	3,2	3,2	0
Pu (Total)	**450**	**566**	**672**	**802**	**455**	**576**	**685**	**848**	**455**	**577**	**698**	**815**
MA (Total)	**70**	**106**	**149**	**205**	**76**	**96**	**105**	**86**	**76**	**89**	**76**	**64**
TRU total	**519**	**671**	**821**	**1007**	**531**	**672**	**790**	**934**	**531**	**666**	**774**	**879**
% fuel with TRU in fleet	0%	50%	50%	100%	0%	50%	50%	100%	0%	50%	50%	100%
TRU in storage	**65**	**103**	**149**	**208**	**27**	**28**	**29**	**30**	**27**	**28**	**29**	**30**

- **Pu+MA recycling :** strong reduction in masses disposed in the wastes and stabilization of the TRU inventories
- **Pu only recycling :** increase of MA in the wastes
- FNR permits to **save natural ressources** – No needs in uranium enrichement facilties

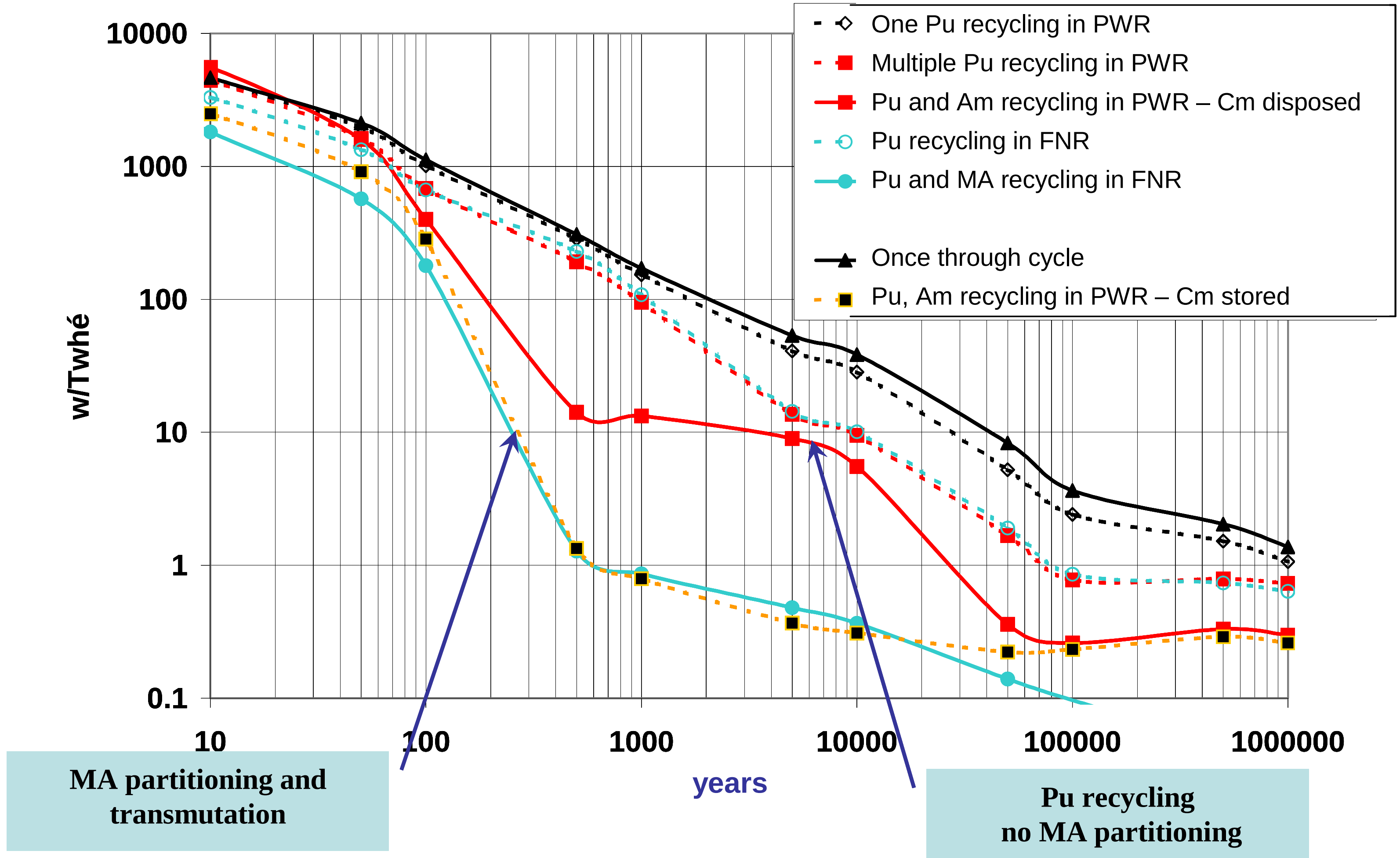
Recycling of M.A. from interim storage in Fast Reactors (P§T since 2020)

- **Impact of shifting the deployment of Fast neutron Reactors from 2035 to 2080:**

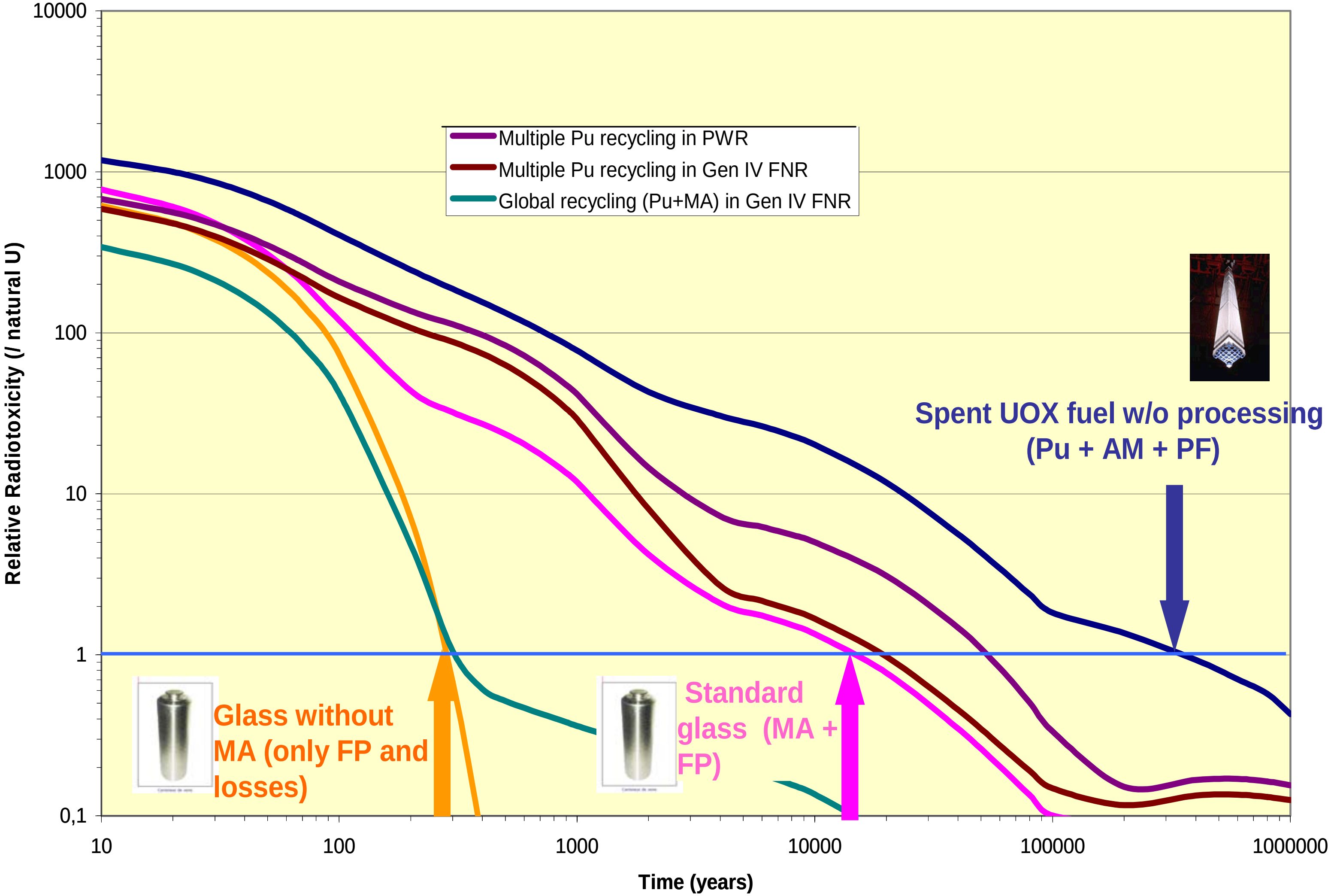
  The time needed to recycle Minor Actinides (MA) is comparable to the time needed to deploy Fast neutron Reactors



Thermal output of the wastes



Impact of the actinides management strategy on the raditoxicity / Activity of ultimate Waste



Investment cost evaluation for the period 2000 – 2150 and uranium consumption

	FNR in 2040 - GAM	FNR in 2080 - GAM	Once through PWR	FNR in 2040 – no GAM
Investment and uranium expenses				
Total investment	1	*1.07*	*1.27*	0.98
U nat cost (max)	1	*2,2*	*8,2*	1
U nat cost (min)	1	*1,7*	*8*	1
Fuel cycle invest.	1	*1.1 to 1.5*	*1.7 to 3.8*	0.98
Thermal output of waste (TRU § FP) or spent fuels (at 2050)				
After 60 years of cooling	1	1.10	2.5	2.0
After 200 years of cooling	1	1	21	15
Mass disposed in the geological site				
At 2100	1	1	44	7

In relatve unit compared to first column

Reactor cost dominates

P&T cost is a balance between

- reduction of investment in front-end and geological storage
- and increase in fuel fabrication and processing

FNR allows to reduce strongly uranium expenses

Actinides Management allows to reduce mass and thermal output of wastes

Synthesis

The partitioning / transmutation scenario implemented in Gen IV FR in (2025 – 2040) allows also to **minimise** the:

- **mass disposed** in the final waste at the end of the century,
  - **by a factor 40-50 or more compared to the once through cycle**
  - **by a factor close to 10 compared to a plutonium recycling (in PWR or FR) without minor actinides recycling**
- **thermal output** of the final wastes, allowing a strong and rapid decrease of power with time,
- **potential radiotoxicity** inventory (and radioactivity) in the final disposal,
- **natural uranium consumption**, reduced by 40% at 2100

Gen IV FR systems are **able to transmute all actinides together with no partitioning, with low penalties**

Keeping and **improving non proliferation** characteristics and **eliminating needs for enrichment** technologies.

This studies have to take into account in more detail the **industrial schedule** to renew fuel cycle facilities for different strategies.

Conclusion :
Future Nuclear Energy Systems R&D Strategy in France

- **Promising prospects for an industrial transmutation strategy based on renewed nuclear plants around 2040: Fast Reactors and renewed spent fuel treatment plant**
  - **Optimum use of existing nuclear plants** *(reactors and spent fuel treatment)*
  - **Flexible separation/recycling scenario with stepwise progress on spent fuel treated and waste form**
  - **Towards an integral actinide recycling if optimum for the fuel cycle back-end**
  - **Potential economic assets of Fast Reactors to limit global expenses over the period (2000 – 2150): Reactors + Fuel cycle plants + Natural uranium**

- **R&D orientations and main milestones**
  - ***Group Actinide separation process GANEX (2008-2015)***
  - ***Development of M.A. transmutation fuels*** *(Phénix, Joyo, Monju, Bor-60, BN-600…)*
  - ***International demonstration of Global Actinide Management in Monju (2015-2025) (+ TRU fabrication plant & La Hague Ganex Micro-Pilot plant)***

→ ***Demonstration of all steps of transmutation based on next generation industrial nuclear plants, and platform of international convergence on Global Actinide Management strategy***

Decision making steps

• release of reports :

- Parliament office's report in March,

- CEA's & Andra's preliminary reports in June 2005 to be supplemented with final reports in December,

- NEA/OECD peer-review reports (reviewing Andra's and CEA's studies) expected in January 2006,

- CNE's 2005 report # 11 in June & global report expected in January 2006.

• National public consultation on the HL LL waste long term management strategy options closing at year end,

• draft bill on HL LL waste long term management available January 2006,

• bill discussion and vote by Parliament before mid 2006.