DE LA RECHERCHE À L'INDUSTRIE

NUCLEAR FUEL CYCLES

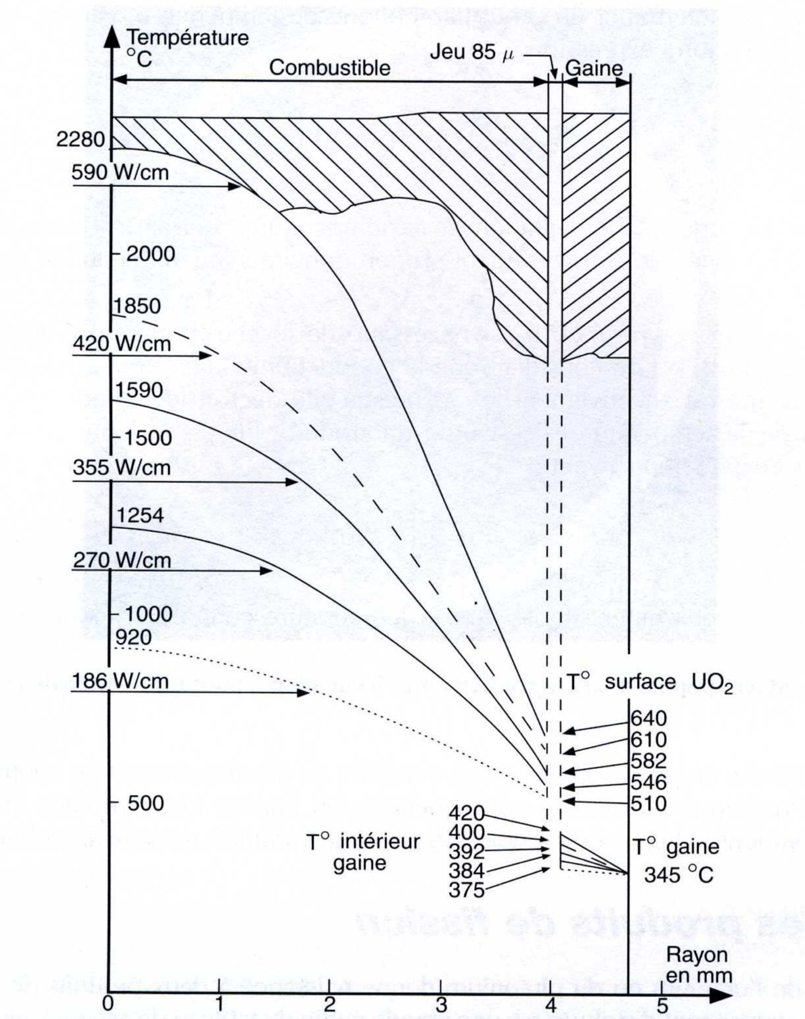
3 – USED NUCLEAR FUEL

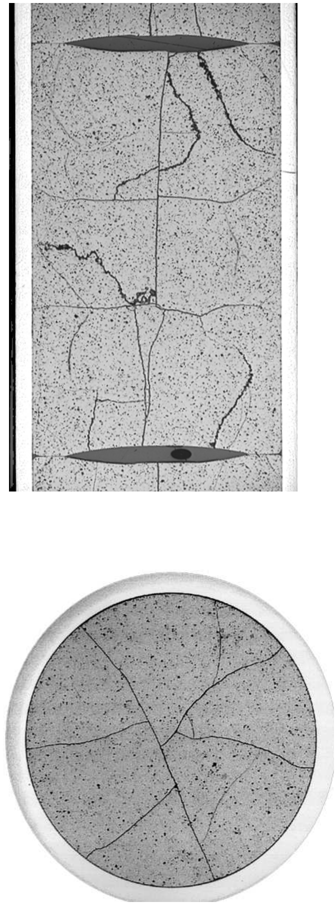
Bernard BOULLIS
bernard.boullis@cea.fr

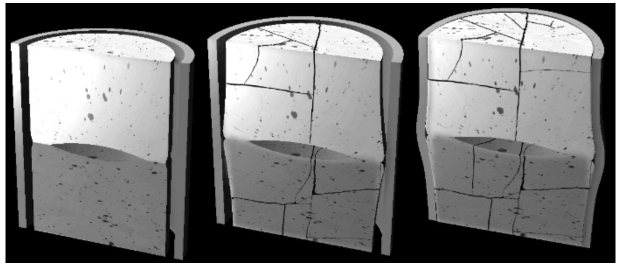
www.cea.fr

Joliot Curie School, St Pierre d'Oleron, 22-27 September, 2019

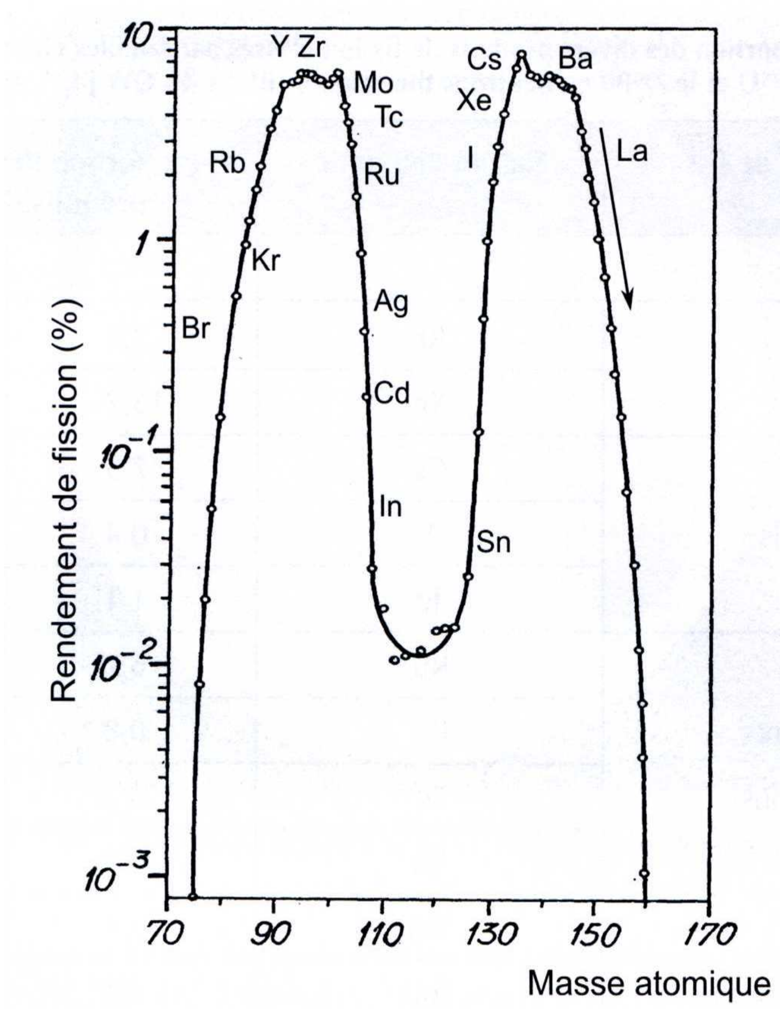
NUCLEAR FUEL CHANGES DURING IRRADIATION:
THERMO-MECHANIC EFFECTS







NUCLEAR FUEL CHANGES DURING IRRADIATION:
FISSION and CAPTURE NUCLEAR REACTIONS



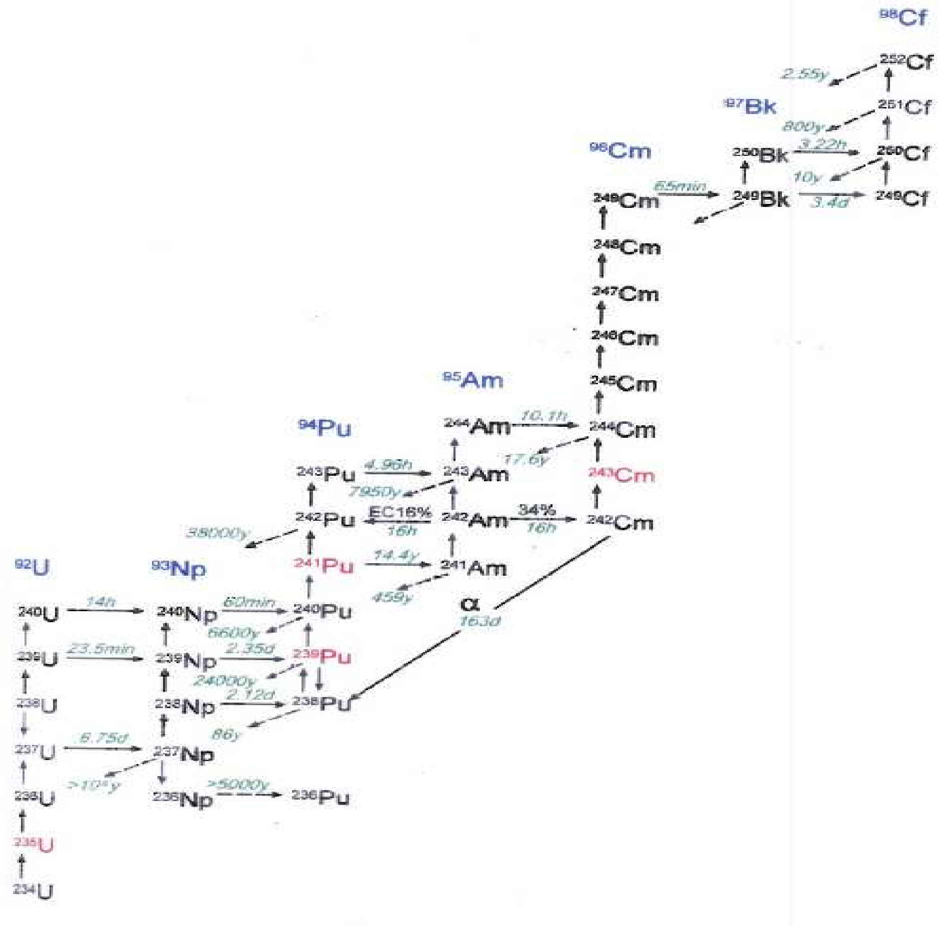


Fig. 5.22. — Transuranic element formation.

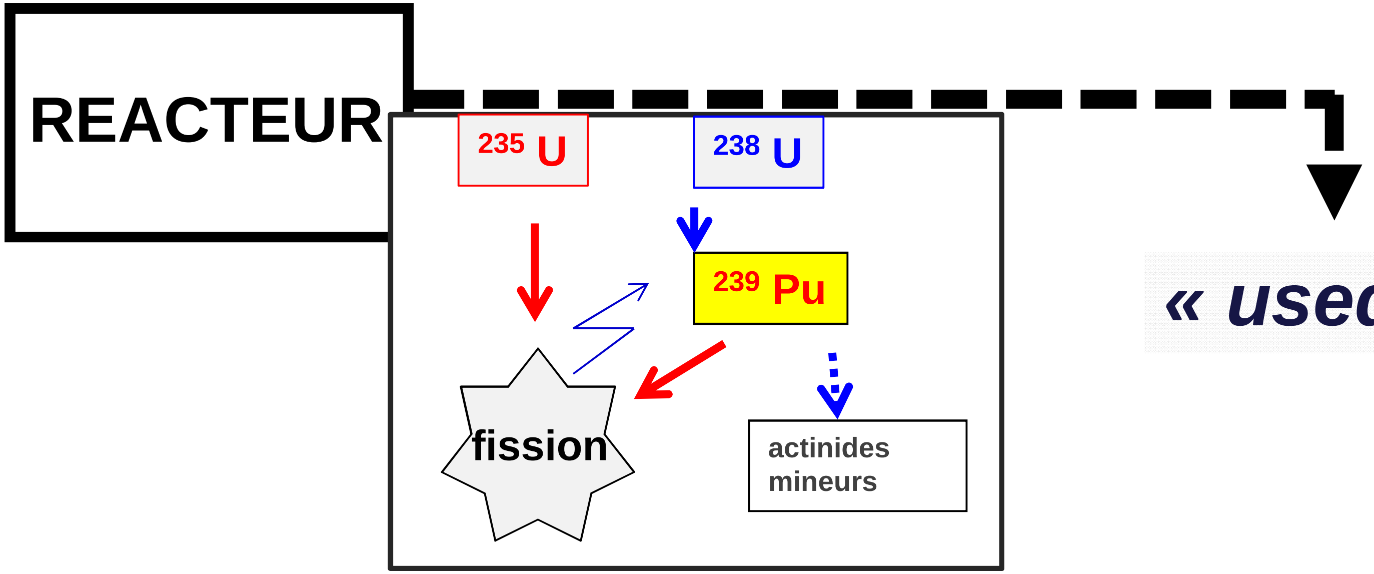
Horizontally: number of protons; vertically: number of nucleons.

↑ neutron capture (n, γ) ↓ (n, 2n) reaction

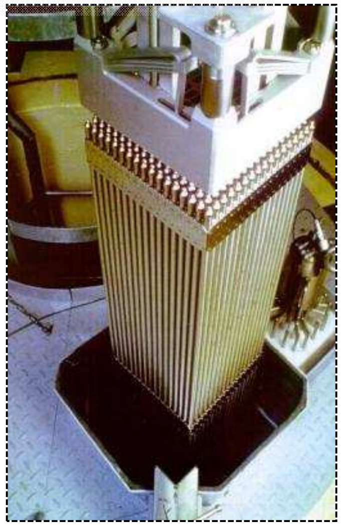
→ β⁻ decay ← β⁺ decay or electron capture (EC)

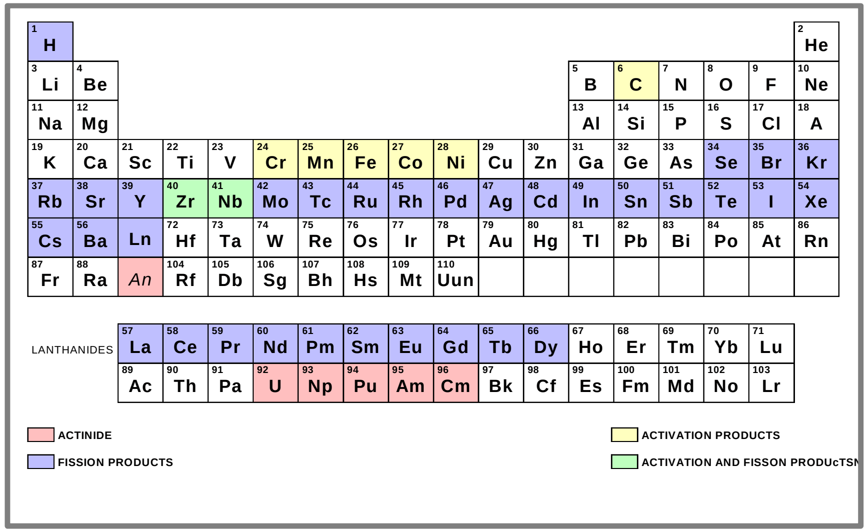
↙ α decay

USED FUEL CONTENT



« used fuel »





- **Uranium (95%)**
- **Produits de fission (4%)**
- **Plutonium (1%)**
- **Actinides mineurs (0.1%)**

(depending on “burn-up”)

USED FUEL « *BURN-UP* »

- ***FISSION YIELD* (Tf)** = **fissionned atoms / fissionnable atoms**

  [*fissionnable = fissile+ fertile (=«heavy atoms »)]* *(atom%)*

- ***BURN-UP* (BU) = extracted energy**
  per ton of irradiated initial heavy metal
  (M(G)W.day per ton)

- ***Standart UOX LWR spent fuel:***
  - **Tf** ~ 3-6 %
  - **BU** ~30-60 GWd/t

  1 fission: ~ 200 Mev

  BU (GWd/t)= 9.6 Tf(%)

M (annual discharge) .BU = annual energy provided

FRONT END ANNUAL NEEDS

- WORLD FLEET
  - fissions per year
  - *^{235}U consumption 330t*
  - *fission products amount*
  - *enriched uranium fuel loaded (~8000 tons)*
  - *natural uranium needs (60 000 tons)*
  - *depleted uranium amount (>50 000 tons)*

448 units, 392GWe, 2488TWhe

- FRENCH FLEET
  - fissile consumption :50t
  - BU: 40GWdays/t
  - Fuel loaded per year (tons)
    = total annual power/BU
    = 380/0.33 TWh /BU
    = **1200 tons**
  - Natural U annual needs:
    1200 (4-0.2) / (0.7-0.2)
    = **9100 tons** *(if Nw =0.2%; Np=4%))*
  - Depleted uranium amount *:*
    = **7900 tons**

58 units, 63GWe, 380TWhe

FRONT END ANNUAL NEEDS

- WORLD FLEET
  - *SWU needs ~ 50 MUTS*

448 units, 392GWe, 2488TWhe

- FRENCH FLEET

SWU annual needs:

SWU=P f(NP)+Wf(NW)-Ff(NF)

f(0.04)~3

f(0.071)~4.8

F(0.002)~6

SWU= (1200 . 3)+(7900 . 6)-(9100 . 4.8)

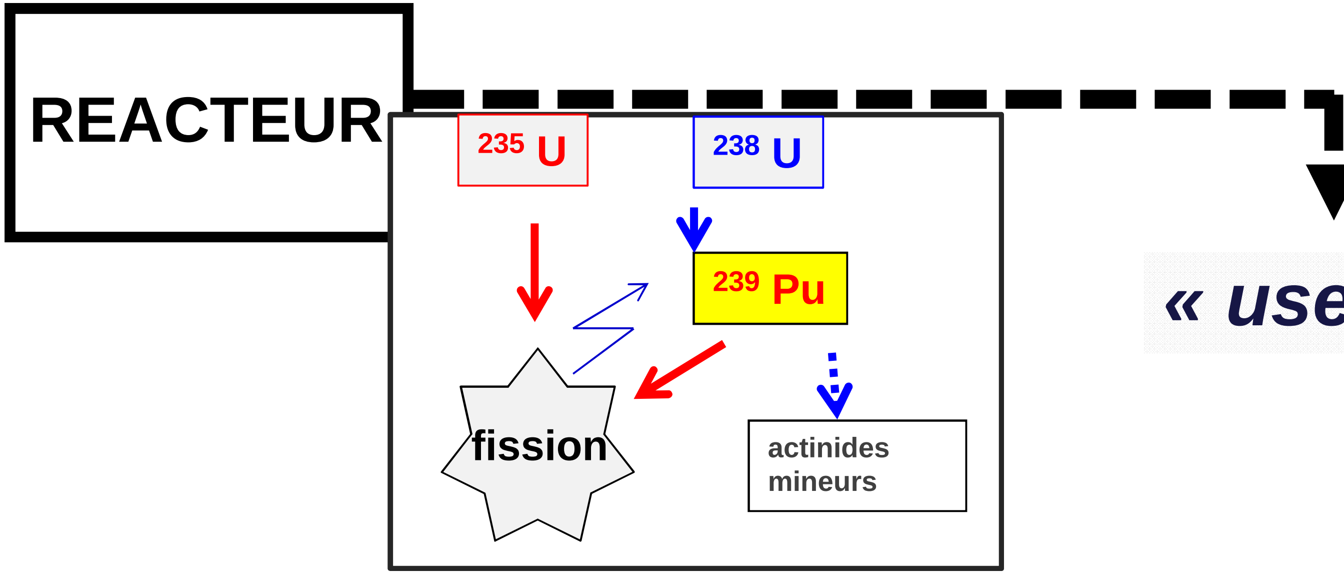
~ 7000 KUTS

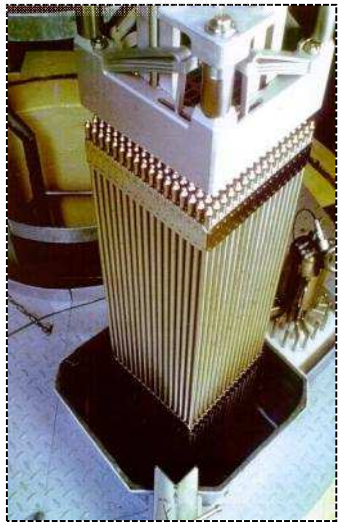
~ 7 MUTS

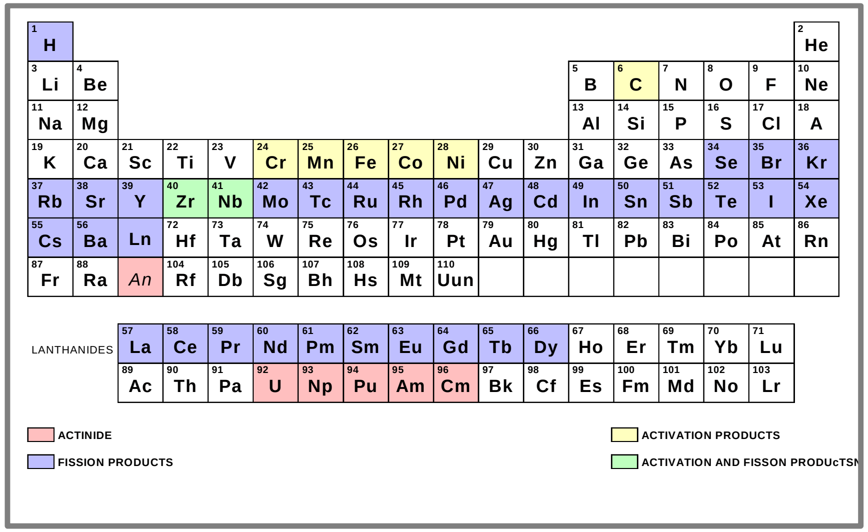
(all without used fuel recycling)

58 units, 63GWe, 380TWhe

USED FUEL CONTENT







- **Uranium (95%)**
- **Produits de fission (4%)**
- **Plutonium (1%)**
- **Actinides mineurs (0.1%)**

(depending on “burn-up”)

from uranium to transuranic elements (TRU)

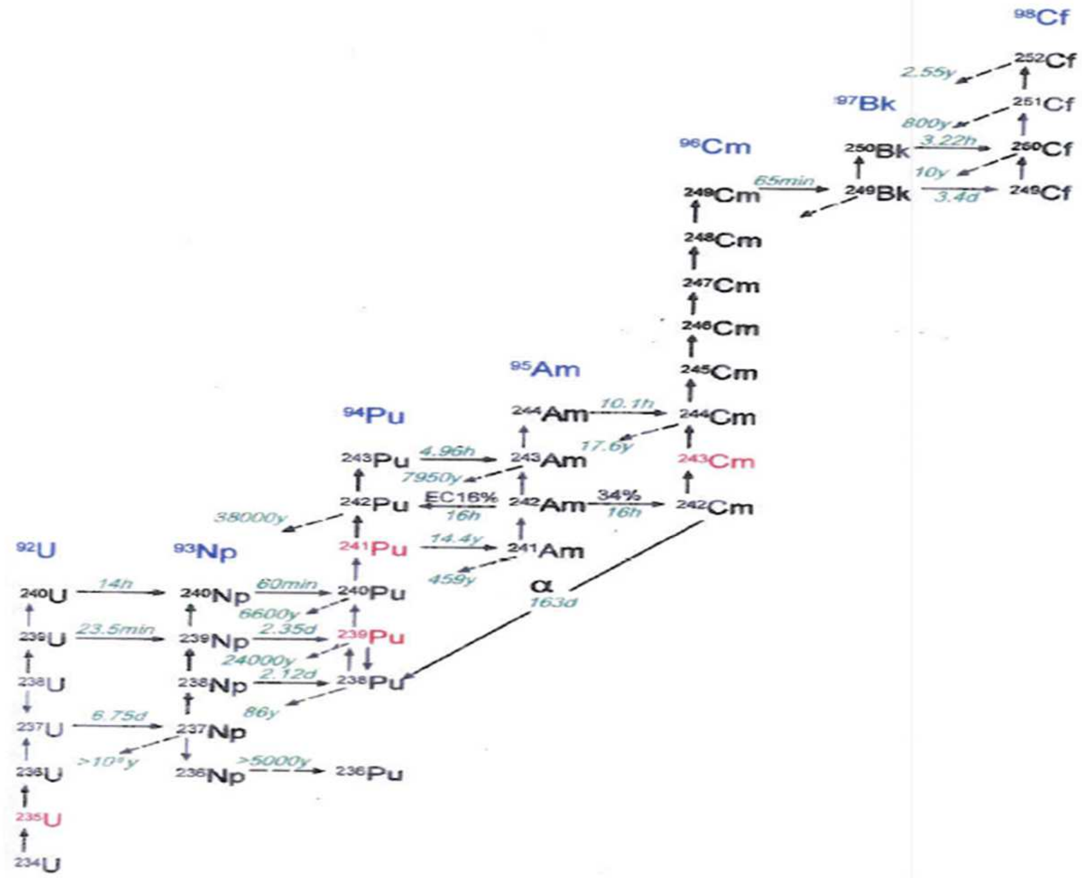


FIG. 5.22. — Transuranic element formation.
Horizontally: number of protons; vertically: number of nucleons.
↑ neutron capture (n, γ) ↓ (n, 2n) reaction
→ β⁻ decay ← β⁺ decay or electron capture (EC)
α decay

RADIONUCLIDES	PERIOD (years)
^{246}Cm	
^{245}Cm	8,50 10^{3}
^{244}Cm	18,1
^{243}Cm	
^{242}Cm	
^{243}Am	7,37 10^{3}
^{242}Am	
^{241}Am	433

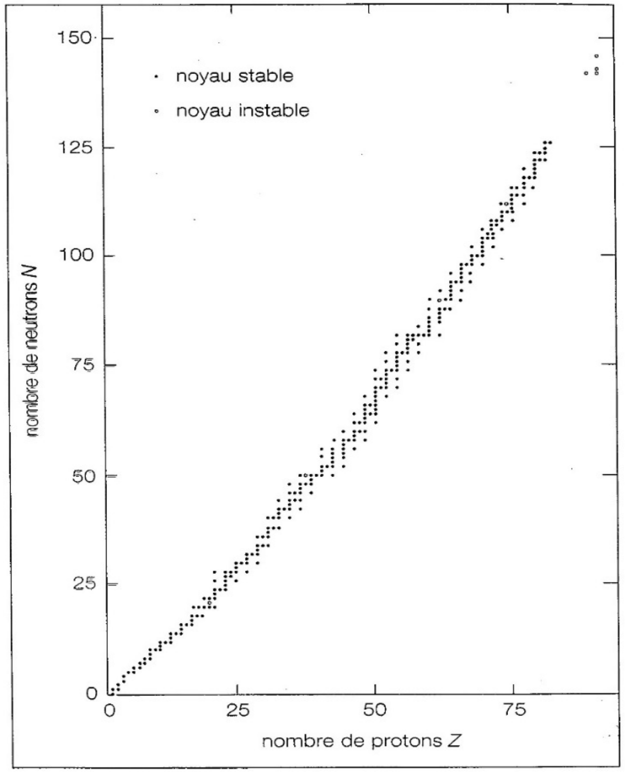
RADIONUCLEDES	**PERIOD (years)**
^{242}Pu	3,74 10^{5}
^{241}Pu	14,4
^{240}Pu	6,56 10^{3}
^{239}Pu	2,41 10^{4}
^{238}Pu	87,7
^{237}Np	2,14 10^{6}
^{238}U	**4,47 10^{9}**
^{236}U	**2,34 10^{7}**
^{235}U	**7,04 10^{8}**
^{234}U	**2,46 10^{5}**
^{233}U	**1,59 10^{5}**
^{232}U	**70**

ABOUT FISSION PRODUCTS

<u>SOLIDS</u> (oxides, metallic compounds) and <u>GAS</u> (>25% atom gas per fission)
(both solid and gaseous swelling)

Rare Gas :	5 kg	(Kr, …)
Alcali :	4 kg	(Cs, …)
Alcalino-terreux :	2 kg	(Sr, …)
Lanthanides :	10 kg	(La, Ce, …)
Chalcogènes :	0,5 kg	(Se, …)
Molybdènium :	3 kg	(Mo)
Halides :	0,2 kg	(I, …)
Technétium :	1 kg	(Tc)
Platinoïds :	4 kg	(Ru, Pd, …)
Others:	(0,3 kg)	

UOX 33GWd/t average content



Sub-assembly, 6 months cooling:~150 000 Bq, 20 kW

Mainly radioactive β$^-$ emitters
TRU:α emitters, PA : β$^-$ emitters

from RADIOACTIVITY (Bq) to its EFFECTS (dose, Gy/Sv)

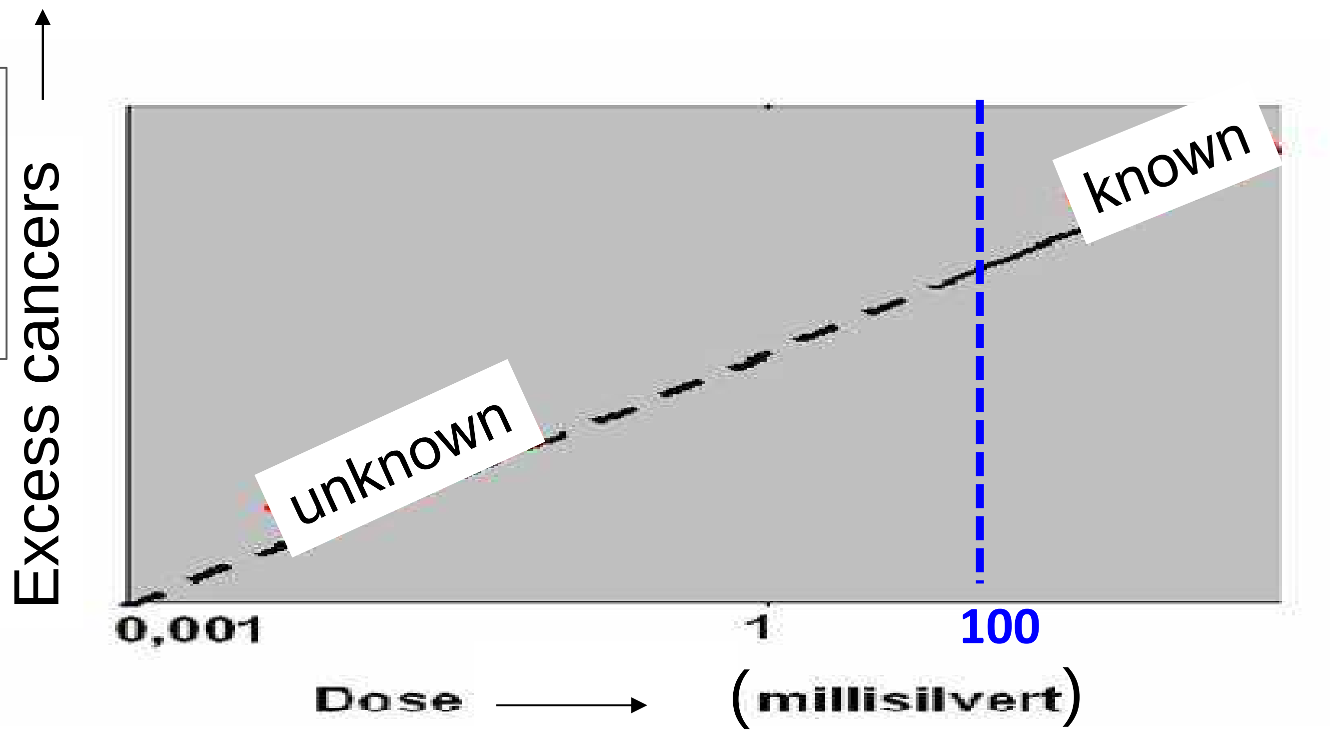
10 Sv	Letal dose (whole body)
100 mSv	*Lowest dose with cancerogen effect detected*
20 mSv	Worker's maxi permitted dose (/year)
2 mSv	Natural irradiation average level (France, /year)
1 mSv	Public maxi permitted dose (/year)

1 Gy = 1J/kg

For dose calculations

1 Sv = 1 Gy x p

p ponderation factor



RADIOTOXICITY: engaged dose after intake *(by ingestion, by inhalation)*

for each nuclide:

$$\textbf{Dose}_{(Sv)} = \textbf{Activity}_{(Bq)} \times \textbf{\textit{DPUI}}$$

Dose coeffficients (DPUI) take into account :

- fraction of the element transfered to blood from intake ;
- element *metabolism* ;
- *nature and energy* of the radiations…
- … and the same for decay products !

for each radionuclide : Dose Coefficients DPUI, ingestion and inhalation.

(ICRP published data, up-dated :*ICRP 67*, *ICRP 72*)

DPUI for some radionuclides *(from: ICRP 67)*

RADIONUCLIDE	DPUI (*) INGESTION (Sv/Bq)
^{14}C	$5,8.10^{-10}$
^{99}Tc	$6,4.10^{-10}$
^{129}I	$1,1.10^{-7}$
Ln	$\sim 10^{-10}$
^{239}Pu	$2,5.10^{-7}$
^{241}Am	2.10^{-7}

(*) DPUI = Dose Per Unit of Intake or « Dose Coefficient »
in french : « facteur de dose », FD)

RADIOTOXICITY OF A SUBSTANCE

Definition: engaged dose after intake (by ingestion) of all the radionuclides present in the substance at the moment.

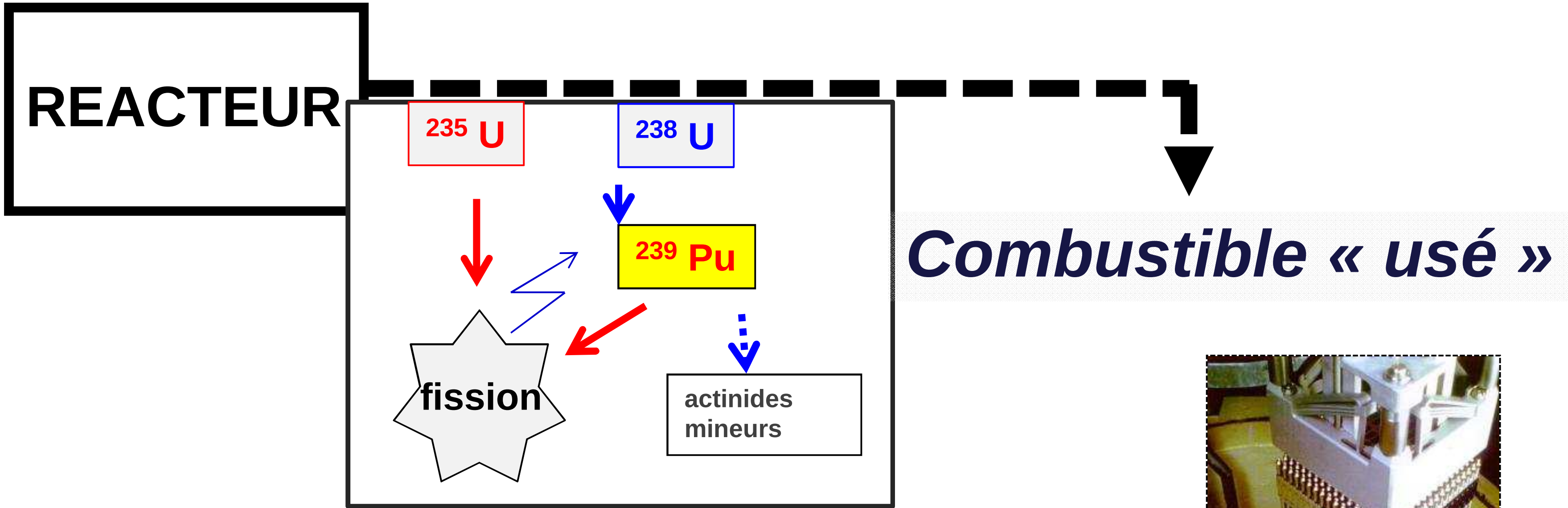
« RADIOTOXIC INVENTORY » at time t

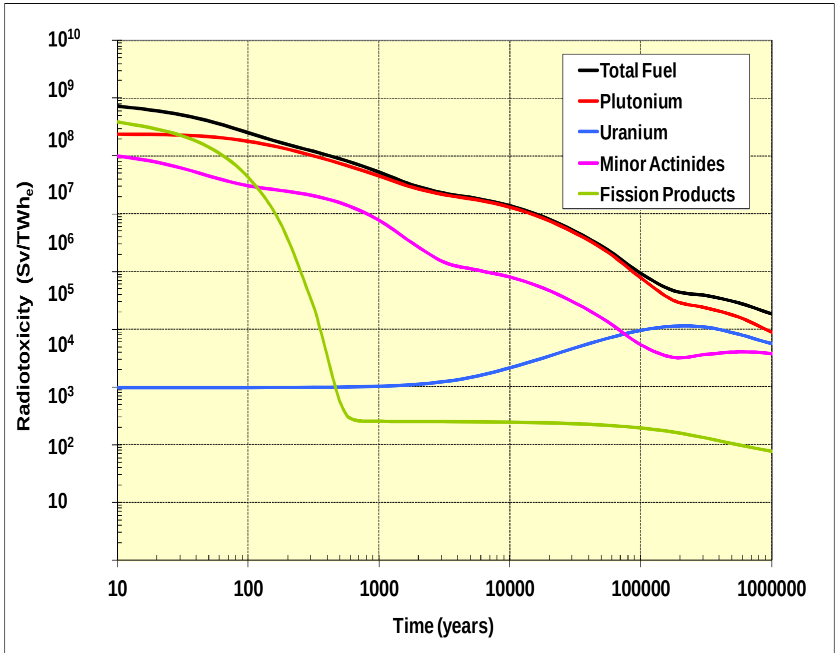
- Radiotoxicity **(t) =** $\sum_{RN} \mathbf{a_{RN}(t) x FD_{RN}}$ *(Sv)*

a_{RN} (t) : radionuclide activity RN at time t

FD_{RN} : DPUI of radionuclide RN.

USED FUEL





- Uranium (95%)
- Produits de fission (4%)
- Plutonium (1%)
- Actinides mineurs (0.1%)

USED NUCLEAR FUEL CONTENT

^{235}U~ 0.7-1%

Uranium (95%)

Fission Products (4%)

Plutonium (1%)

Minor Actinides (0.1%)

Used FuelAverage Content

ISOTOP	NATURAL U	USED FUEL
232		ε
234		0.02 %
235	0.7 %	1.09 %
236		0.45 %
238	99.3 %	98.5 %

ISOTOP	USED UOX	USED MOX
238	1.8 %	3.9 %
239	58.3 %	37 %
240	22.7 %	30 %
241	12.2 %	14.8%
242	5 %	14.4%

Fissile isotops ~ 75%

DE LA RECHERCHE À L'INDUSTRIE

NUCLEAR FUEL CYCLES

4 – USED FUEL MANAGEMENT

Bernard BOULLIS

bernard.boullis@cea.fr

www.cea.fr

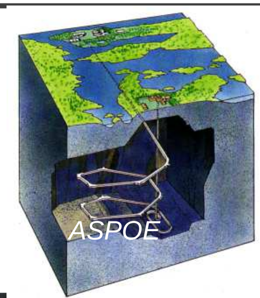
Joliot Curie School, St Pierre d'Oleron, 22-27 September, 2019

USED FUEL MANAGEMENT OPTIONS

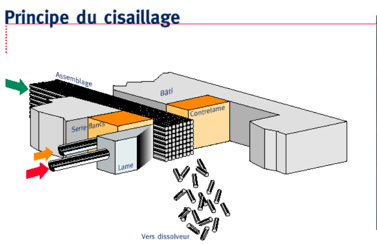
world cumulative amount ~ 300 000 tons
(~ + 10 000 tons /year)

- INTERIM STORAGE

- DIRECT DISPOSAL



- RECYCLING



USED FUEL INTERIM STORAGE

<u>INTERIM STORAGE in FRANCE</u>:

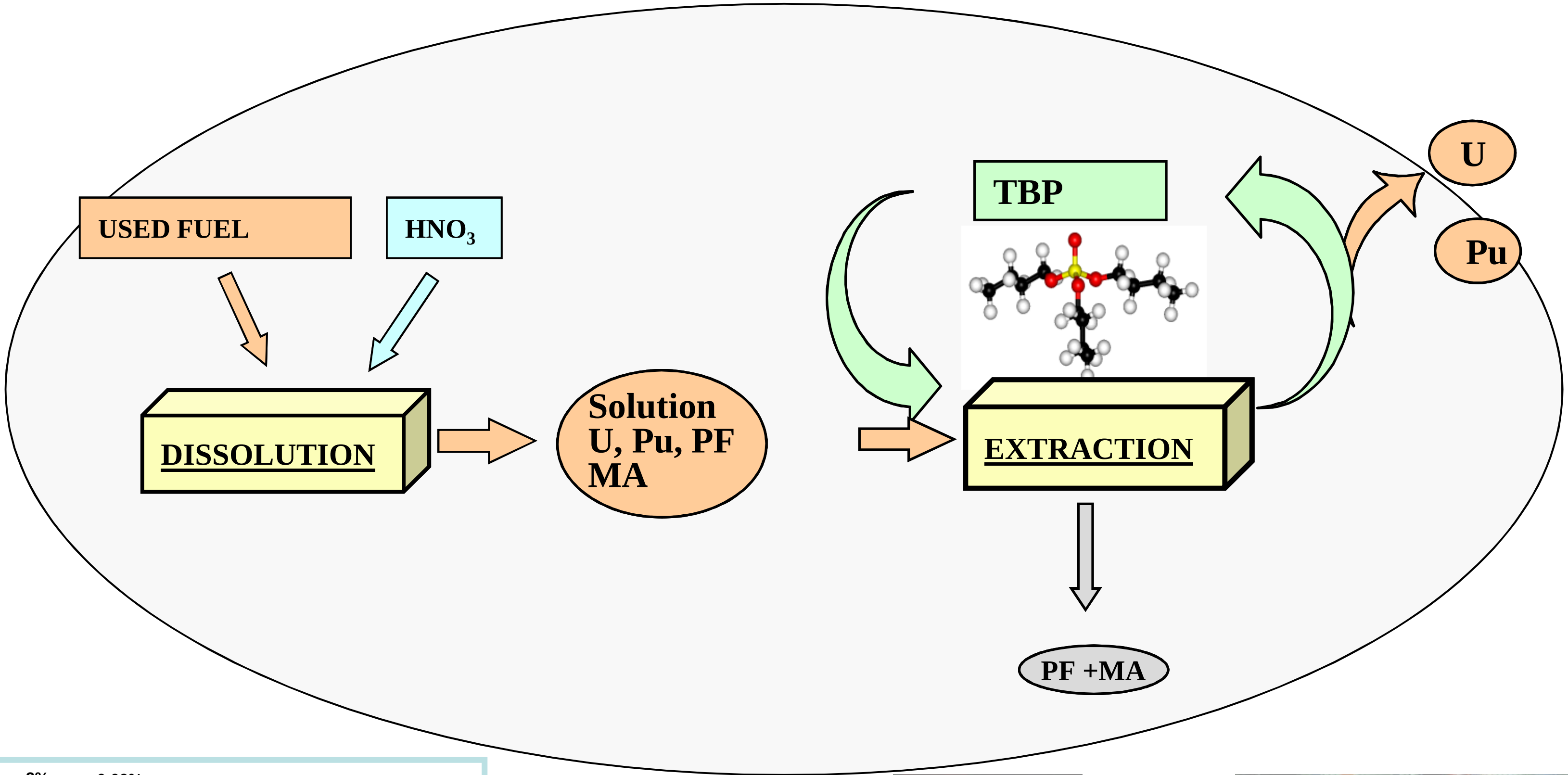
Pool (reactor site) : 1 to 3 years

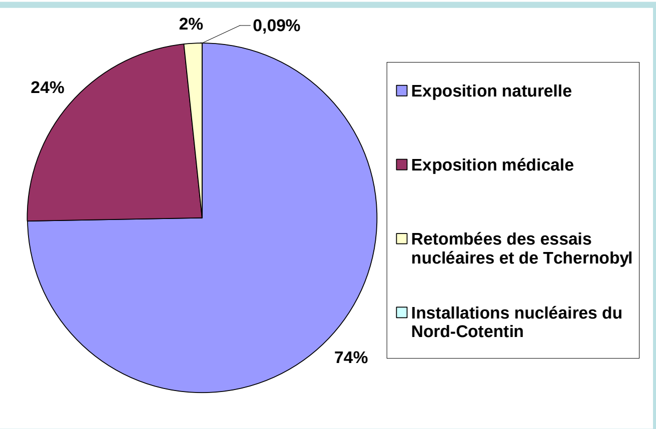
Pool (centralized site): 5… 10 years…

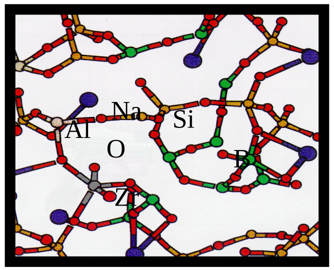
Casks (dry) only for transportation

REPROCESSING (RECYCLING) TODAY

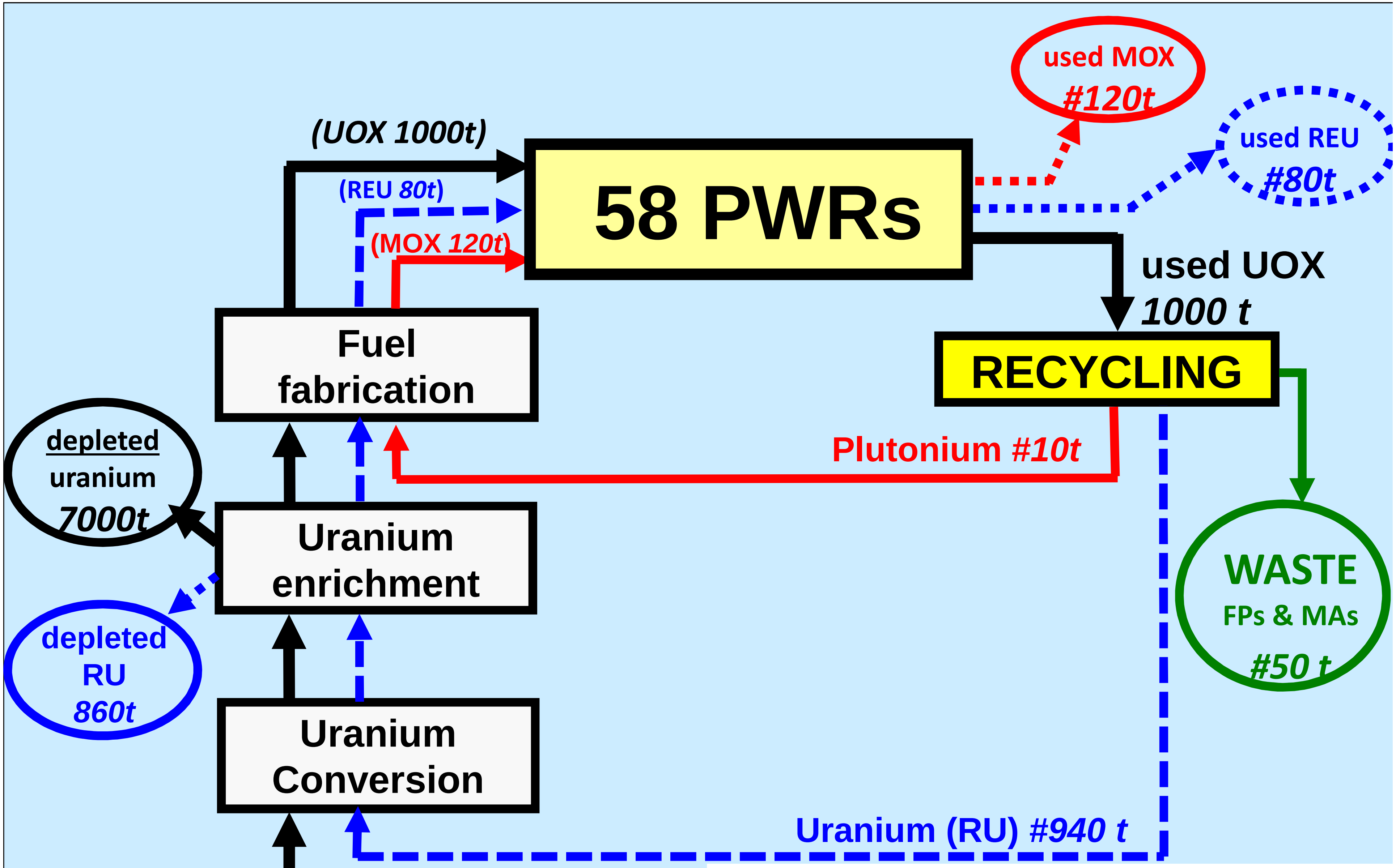
(La Hague ORANO plant)





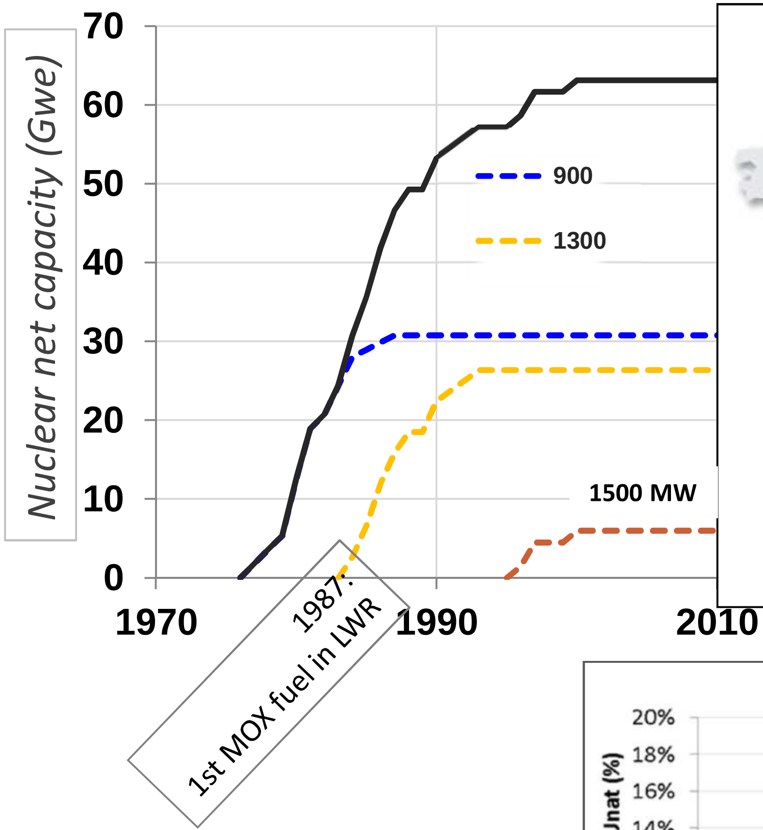


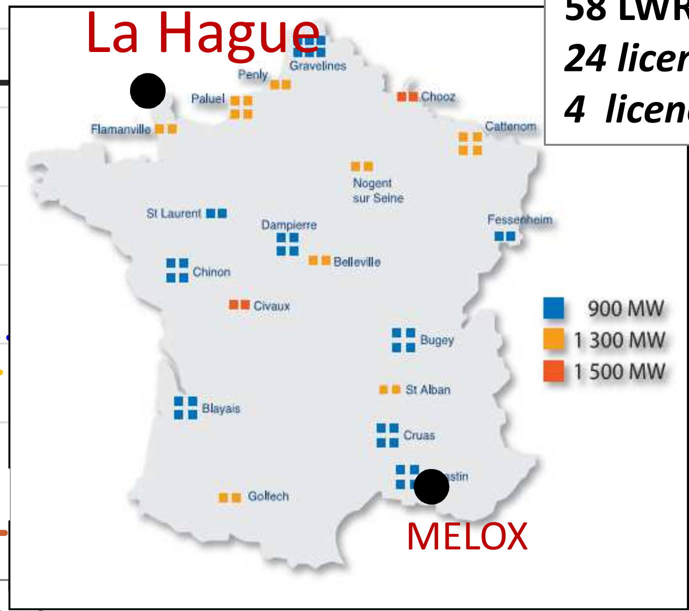
 # 10-15 glass canisters / réactor /year



RECYCLING : (PRINCIPLE OF) THE FRENCH FUEL CYCLE *(approximative amounts, per year)*

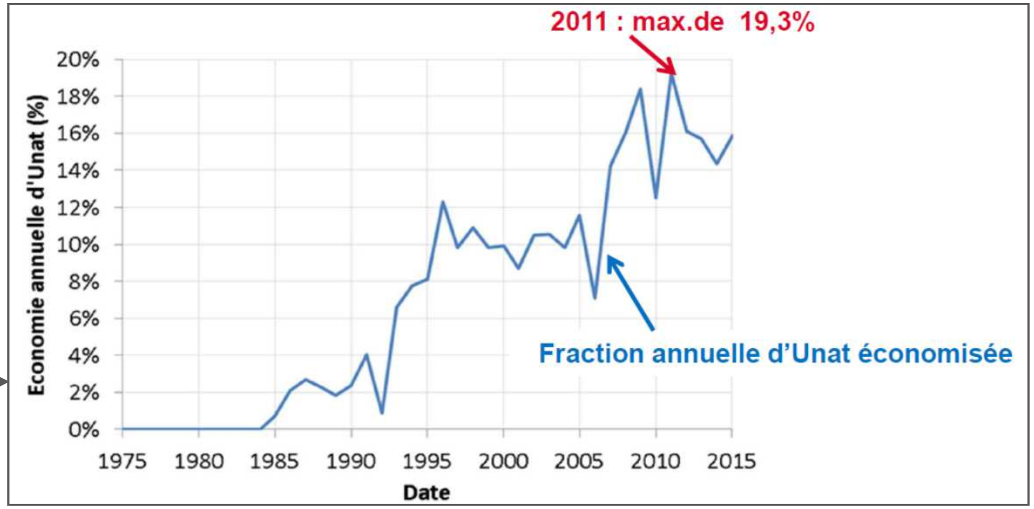
THE FRENCH NUCLEAR FLEET





58 LWR
24 licenced for MOX
4 licenced for REU

Estimated uranium annual savings
(cumul. ~25 000tons)



CURRENT RECYCLING FRENCH STRATEGY :
"THE NATIONAL INVENTORY", 2015 (ANDRA)

- ***used UOX*** **:**
  2013 : 12000 t (# stabilized)

- ***used MOX*** **:**
  2013: 1500 tons *(increasing 120 tons/year)*
  2035 : 4000t (about # 250t Pu)

- ***used REU*** **:**
  2013 :420 t
  2035 :1800 t

- ***depleted U:***
  2013 : 290 000 t
  2035: 450 000 t

THE CURRENT FRENCH NUCLEAR FLEET

- **Power: 63.2 GWe *(MSI: 1977 -1999)***

- **34 units 900 MWe *(31 GWe)***

- FESSENHEIM (1 2)	[1977]	UOX	***6 reactors100% NEU***
- BUGEY (1 2 3 4)	[1979-1980]	UOX	
- CRUAS (1 2 3 4)	[1984-1984]	URE	***4 reactors 100% REU***
- TRICASTIN (1 2 3 4)	[1980-1981]	MOX : >1996 et>	***24 réactors 70% NEU/30% MOX***
- SAINT LAURENT (1 2)	[1981]	MOX : >1987	
- BLAYAIS (1 2 3 4)	[1981-1983]	MOX : >1994 (2013)	
- DAMPIERRE (1 2 3 4)	[1980-1981]	MOX : >1992 et>	
- CHINON (1 2 3 4)	[1982-1987]	MOX : >1998 et >	
- GRAVELINES (1 2 3 4 5 6)	[1980-1985]	MOX :> 1989 et>	

- **20 units 1300 Mwe *(26 GWe)*** *[>1986]* *all 100% NEU*

- **4 units 1450 Mwe *(6 GWe)*** *[> 1996]* *all 100% NEU*

- ***(1 réactor 1650 MWe** (EPR Flamanville)**)*** *[>2020(?)]*

NUCLEAR ENERGY : *THE LONGER TERM*

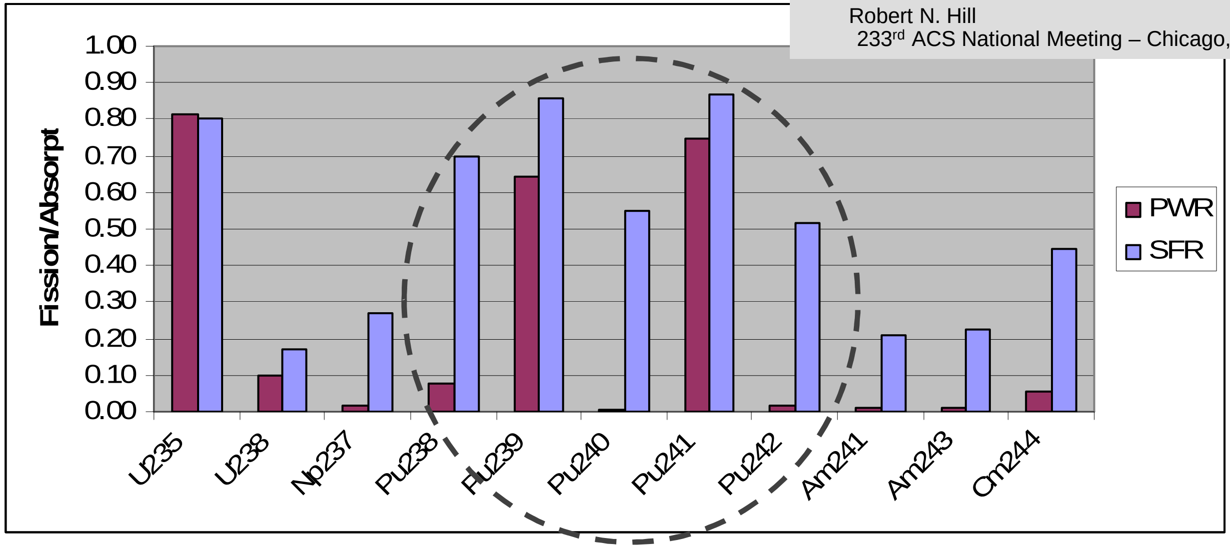
- **A SUSTAINABLE MANAGEMENT OF NUCLEAR MATERIALS & WASTE :**

  - *extension of the use of natural resource ;*
    (only less than 1% of extracted uranium is currently used)
  - *minimizing waste amount and nocivity*
    (and/or used fuel interim storage growth)

- **POSSIBLE ROUTES**
  - *multi-recycle U and Pu in suitable reactors*
    [fast neutron reactors favors Pu fission vs. capture ,
    allowing efficient Pu multi-recycle (more difficult in LWRs)]
  - *other options: …TWR…Th-based cycle…*

WHY FAST NEUTRON REACTORS?

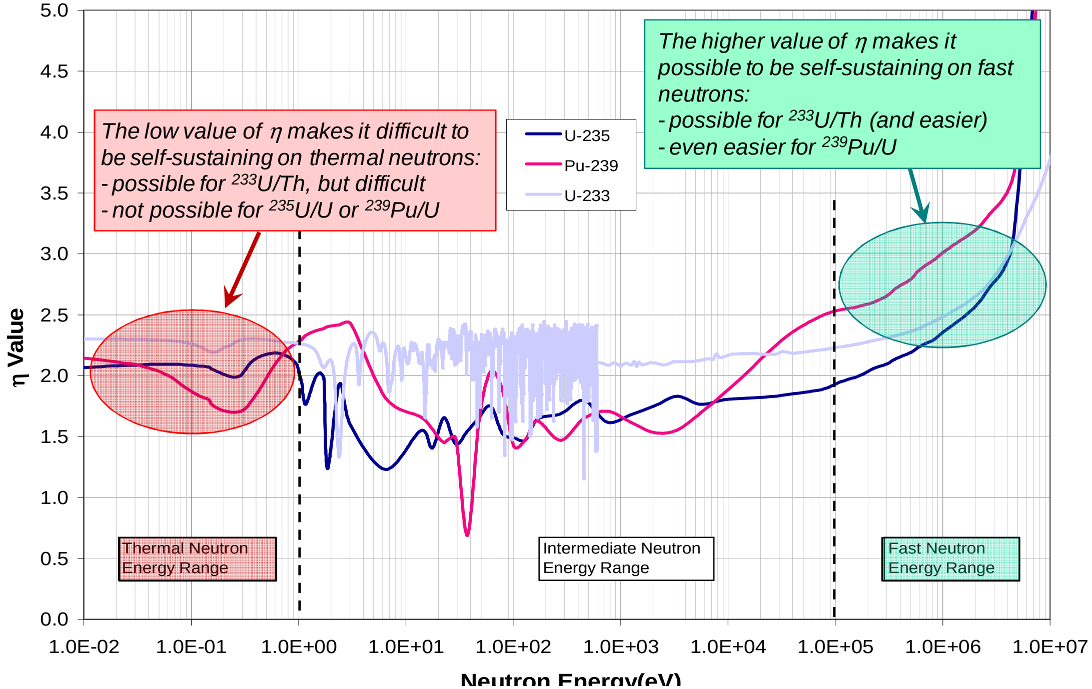
Robert N. Hill
233rd ACS National Meeting – Chicago,, 2007



Fission to absorption ratio

neutrons after fission

from INL Idaho National Laboratory



PU MULTIRECYCLE IN LWRS

[UOX et MOX 46 GWj/t

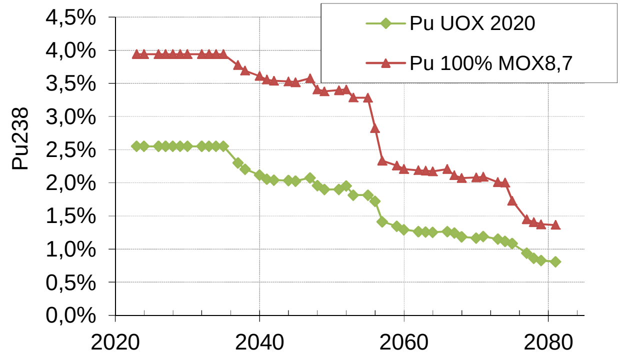
Pu isotop	Pu in REP -UOX (%)	Pu in REP -MOX (%)
238	2.48	3.8
239	53.3	39.9
240	24.8	31.1
241	12.1	13.4
242	7.3	11.8

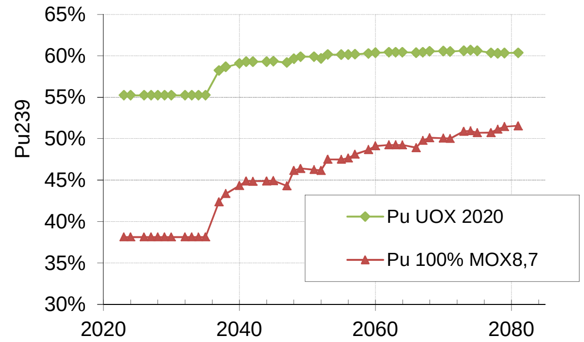
(^{239}Pu eq) # 45% → (^{239}Pu eq) # 25%

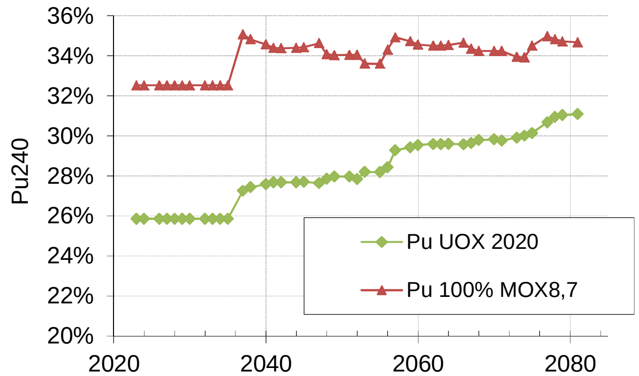
Pu multirecycle in LWRs

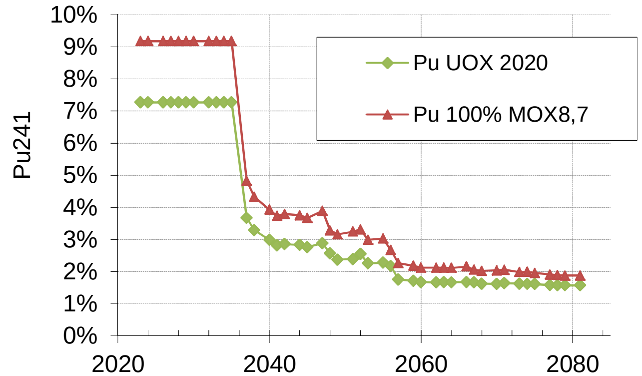
- *towards higher isotops (and minor actinides)*
- *higher and higher[Pu] needed in MOX fuel (reactivity)*
- *Multirecycle very difficult to impossible ([Pu] capped, safety)*

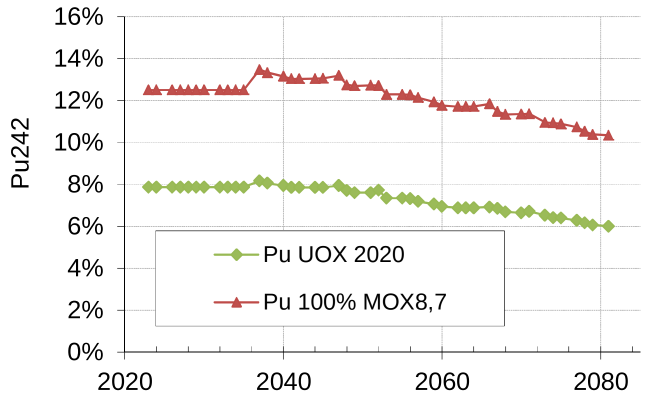
Pu MULTIRECYCLE IN FRs : *an exemple (ASTRID CEA project)*

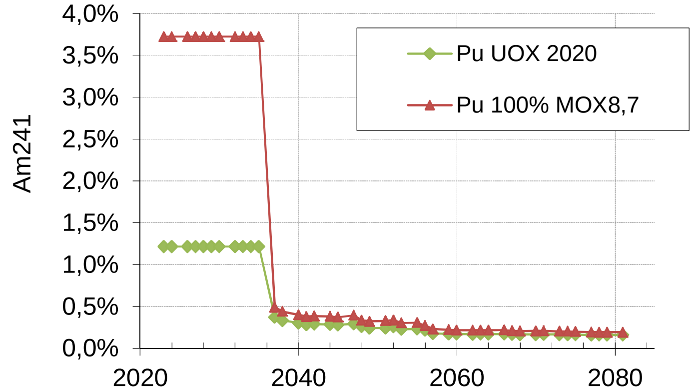


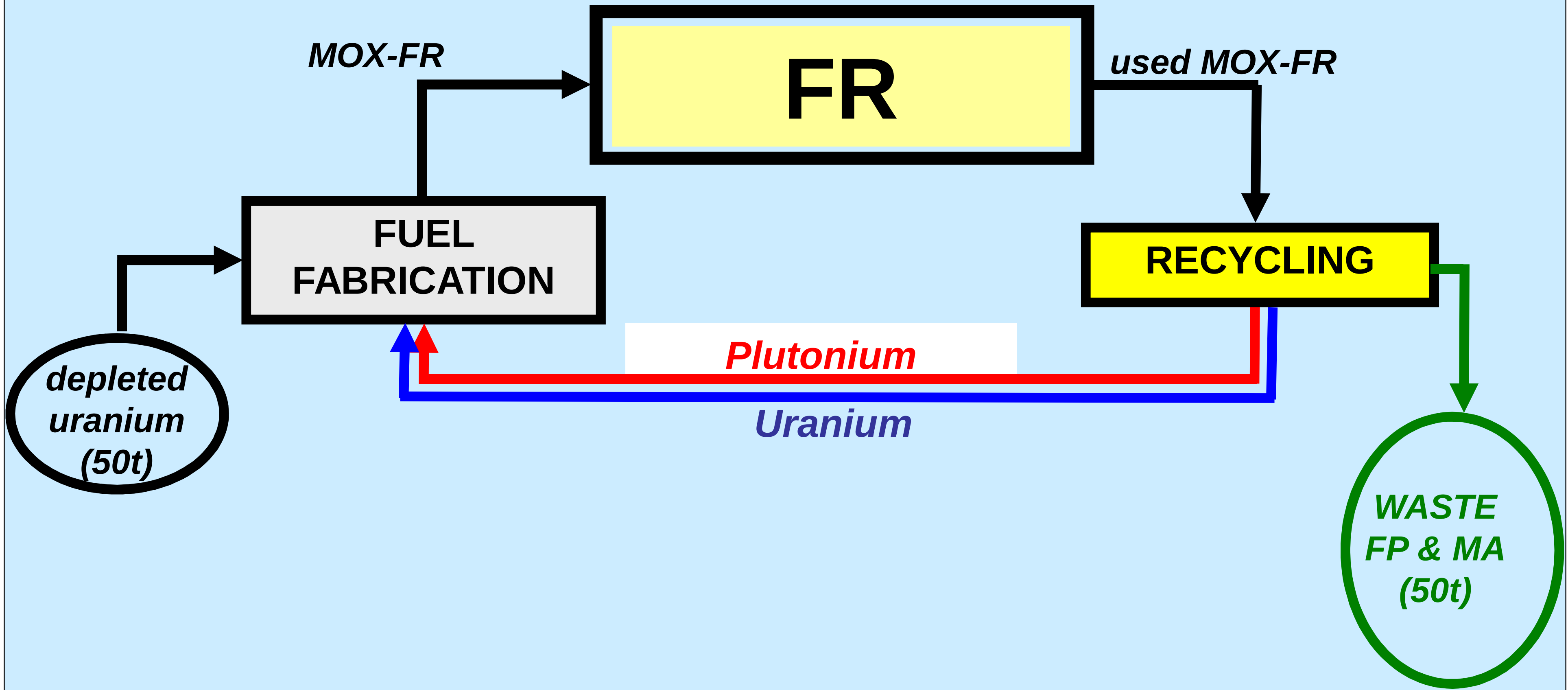








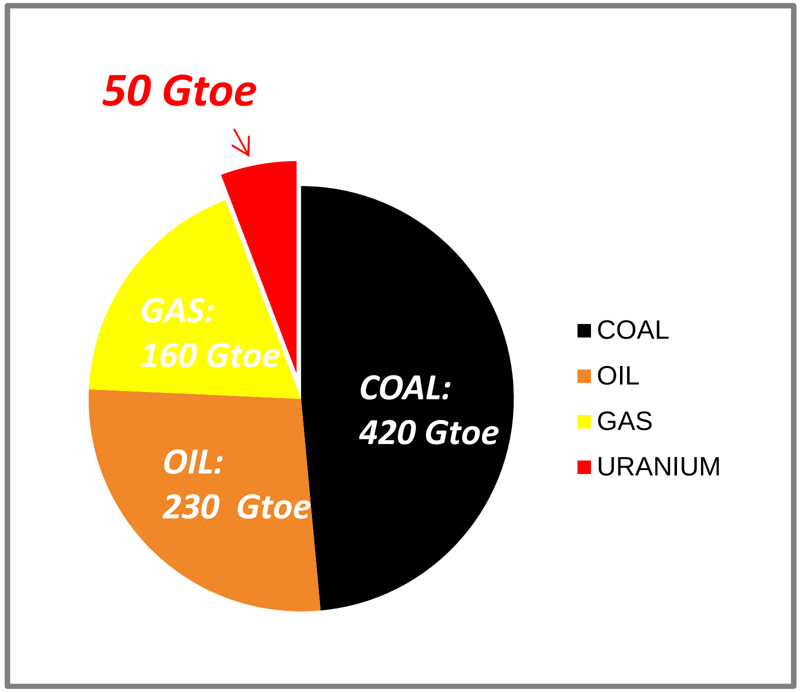




(HYPOTHETIC) FR FUEL CYCLE

(approximative amounts for 400TWh)

URANIUM among OTHER FOSSIL RESOURCE…

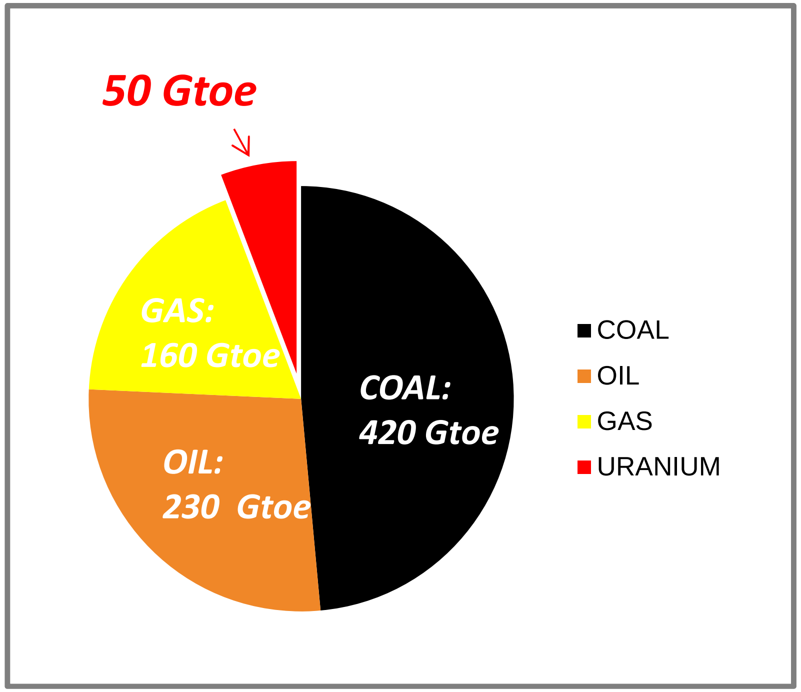


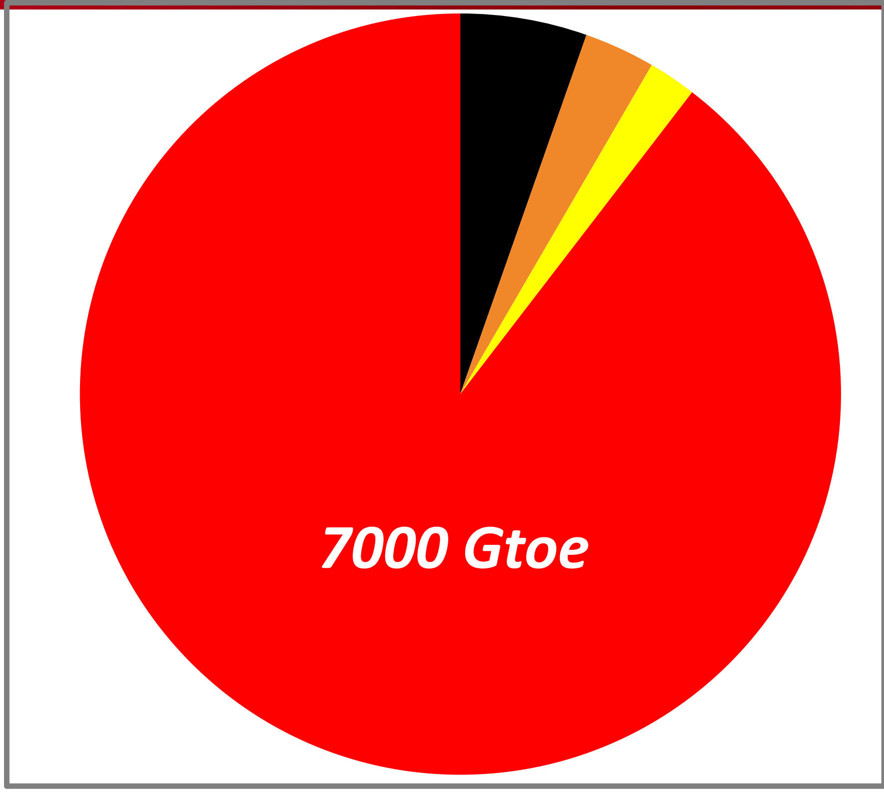
Uranium use
in <u>thermal neutrons</u> (*current*) reactors

<u>Identified</u> <u>conventional</u> resource

(BP statistical review, 2013 and OECD/NEA, 2012)
(OIL 235 Gt, COAL 860Gt, GAS 187 Tm³,URANIUM 4Mt)

URANIUM among OTHER FOSSIL RESOURCE…





Uranium use
in thermal neutrons (*current*) reactors

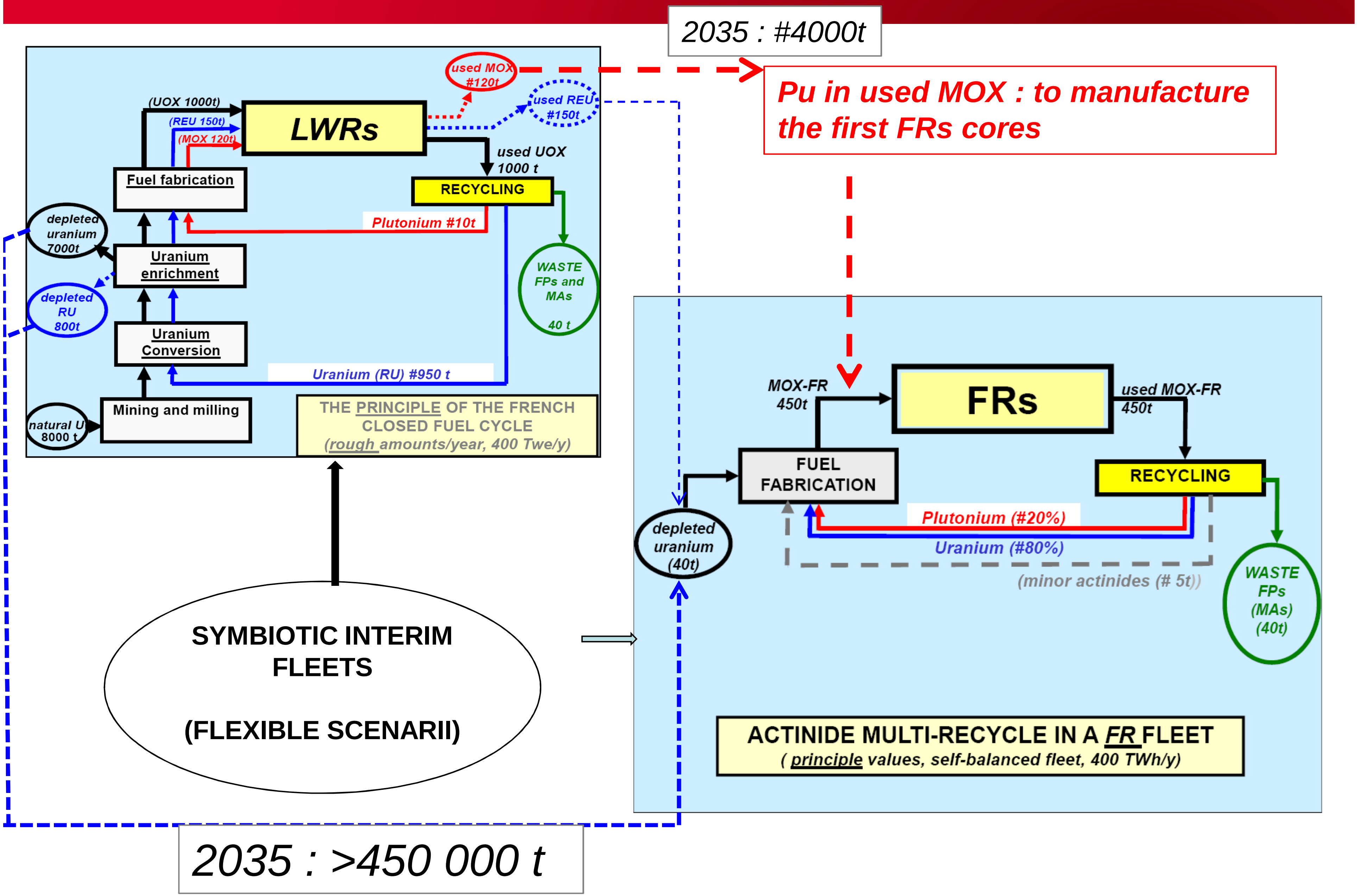
Uranium & Plutonium multirecycled
(in fast neutron reactors)

Identified conventional resource

(BP statistical review, 2013 and OECD/NEA, 2012)

(OIL 235 Gt, COAL 860Gt, GAS 187 Tm³,URANIUM 4Mt)

from current LWRs to FRs…. the storyline…



URANIUM ANNUAL NEEDS:
DIRECT DISPOSAL vs. RECYCLING OPTIONS

- WORLD FLEET
  - fissions per year
  - ^{235}U consumption
  - enriched fuel needs
  - natural uranium needs
  - SWU needs
  - HLW amount

- MONORECYCLE
  - uranium savings (U recycle)
  - uranium savings (Pu recycle)
  - SWU savings

- MULTIRECYCLE
  - uranium savings

DE LA RECHERCHE À L'INDUSTRIE

cea

NUCLEAR FUEL CYCLES

5 – PARTITIONING & TRANSMUTATION
OF LONG-LIVED RADIONUCLIDES

Bernard BOULLIS

bernard.boullis@cea.fr

www.cea.fr

Joliot Curie School, St Pierre d'Oleron, 22-27 September, 2019

FINAL HL WASTE MANAGEMENT: *OPTIONS…*

(1) <u>SOURCE TERM MINIMIZATION</u>:

(limit waste production?)

(« transmute »?)

(2) <u>WAIT</u> …

(interim storage for decay)

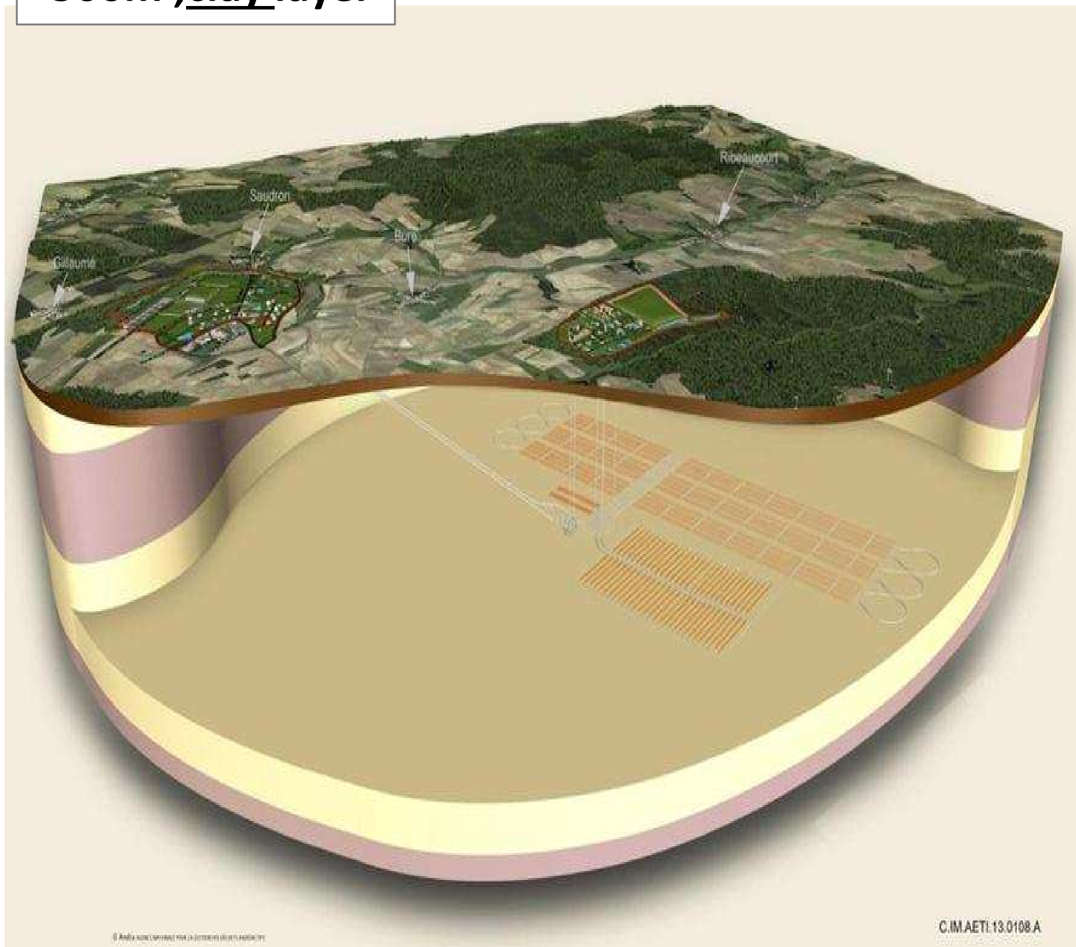
(3) <u>CONFINE…</u>

(prevent (?),delay ,« filter » final impact)

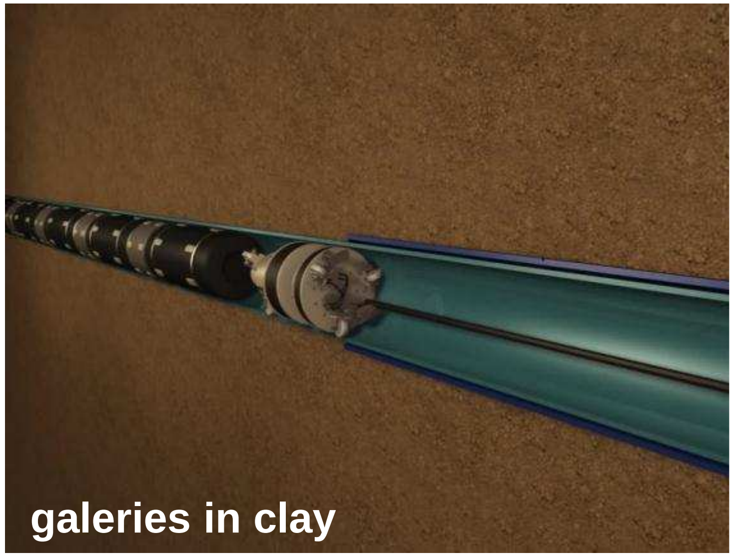
- *high durability matrixes & canisters*
- *disposed into high stability geological places ?*

FINAL HLW MANAGEMENT:
THE FRENCH CASE

CIGEO PROJECT:
-500m ,clay layer

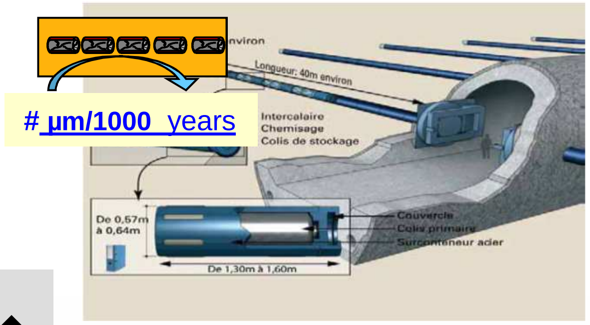


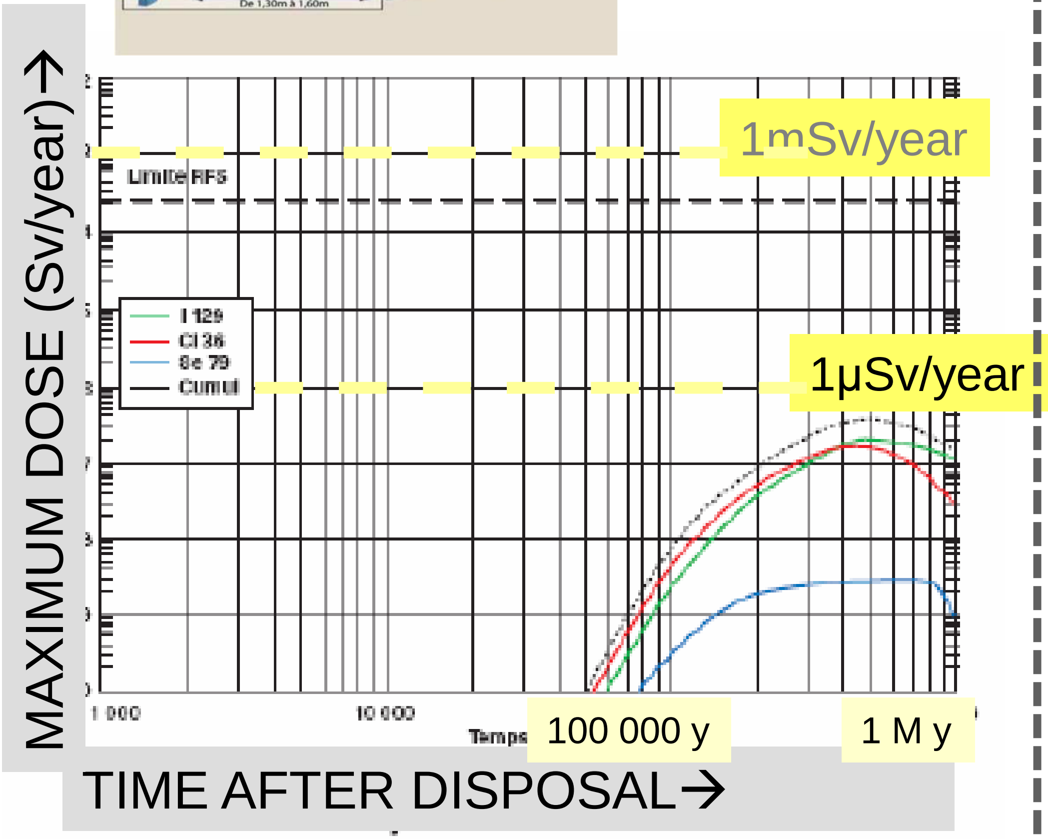
150 liters,
FPs #15%, #2kW,
>15 000 TBq



galeries in clay

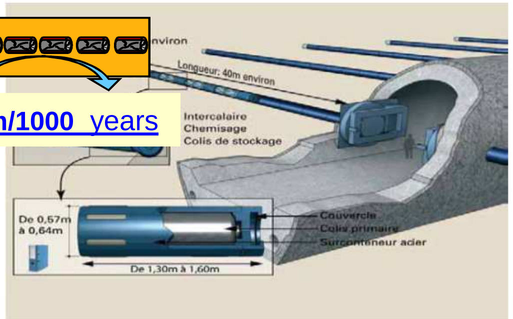
FINAL HLW MANAGEMENT:
THE FRENCH CASE

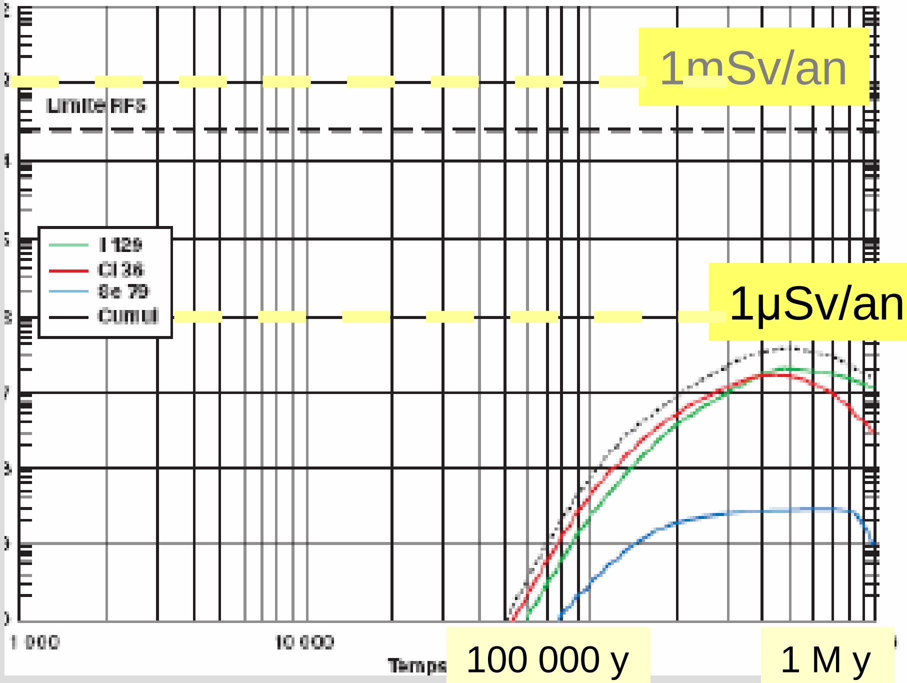




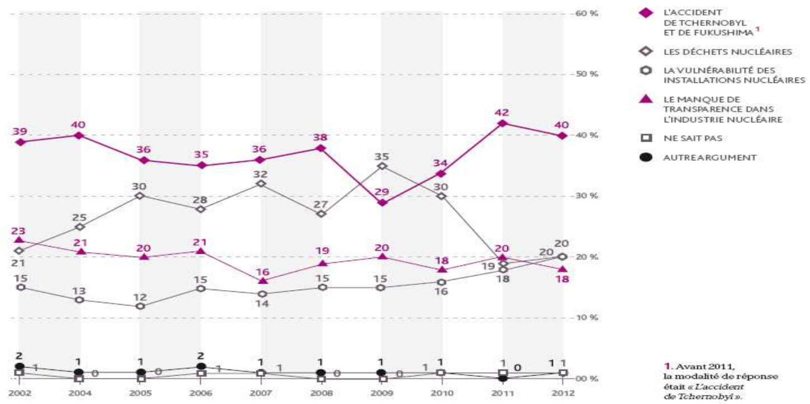
(ANDRA, « clay report », 2005)

DECHETS NUCLEAIRES:
OPTIONS DE GESTION – STOCKAGE VL





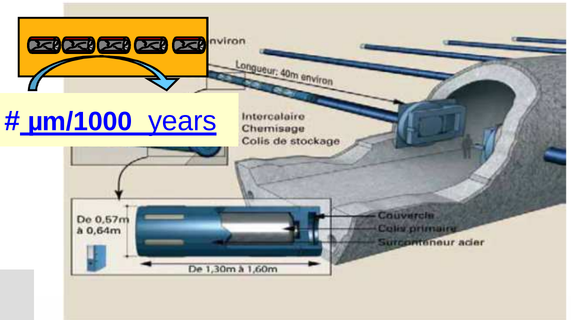
(ANDRA, « Rapport argile », 2005)

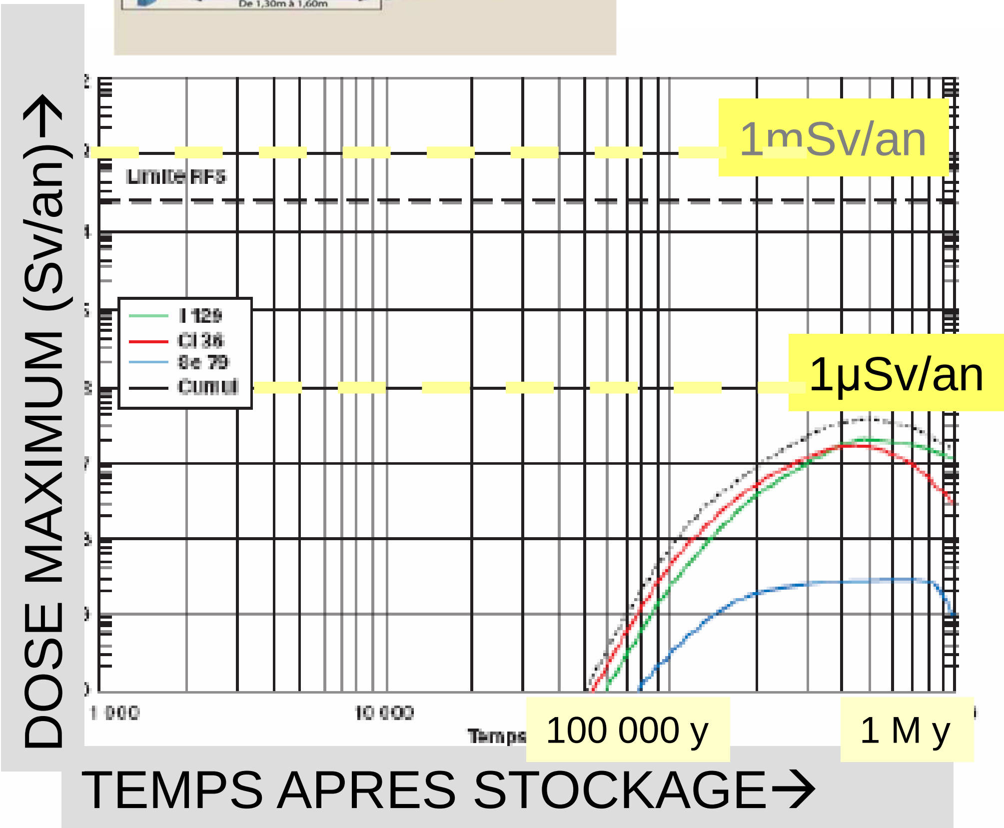


Baromètre IRSN,2013

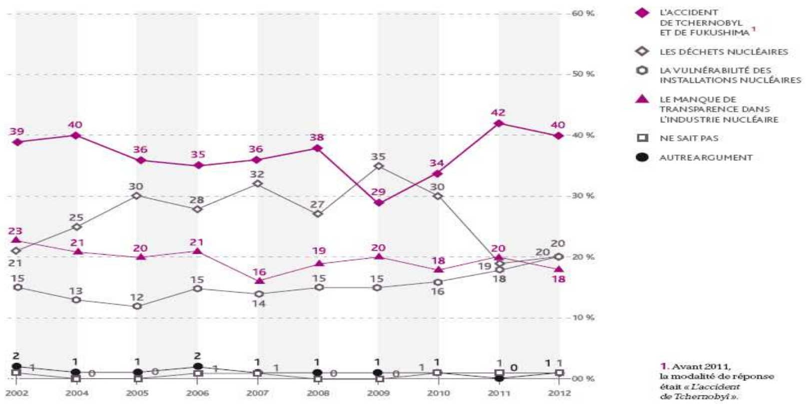
against nuclear power, why ?

DECHETS NUCLEAIRES:
OPTIONS DE GESTION – STOCKAGE VL



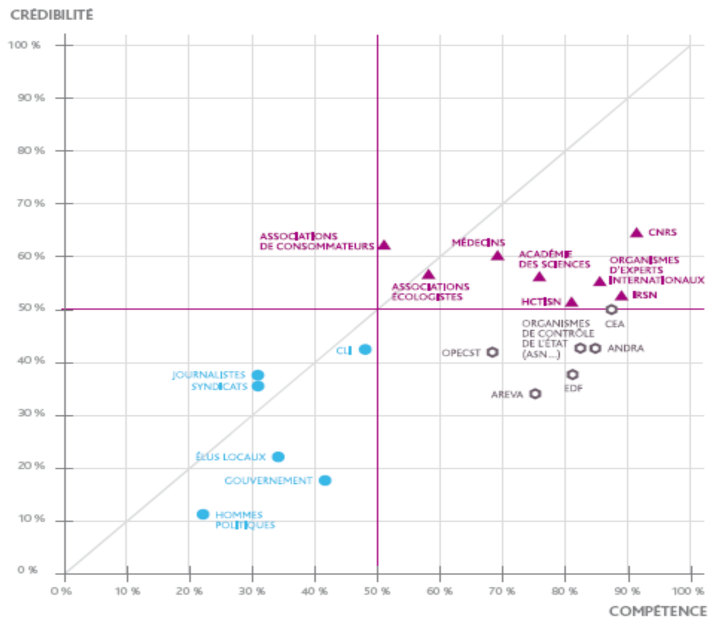


(ANDRA, « Rapport argile », 2005)

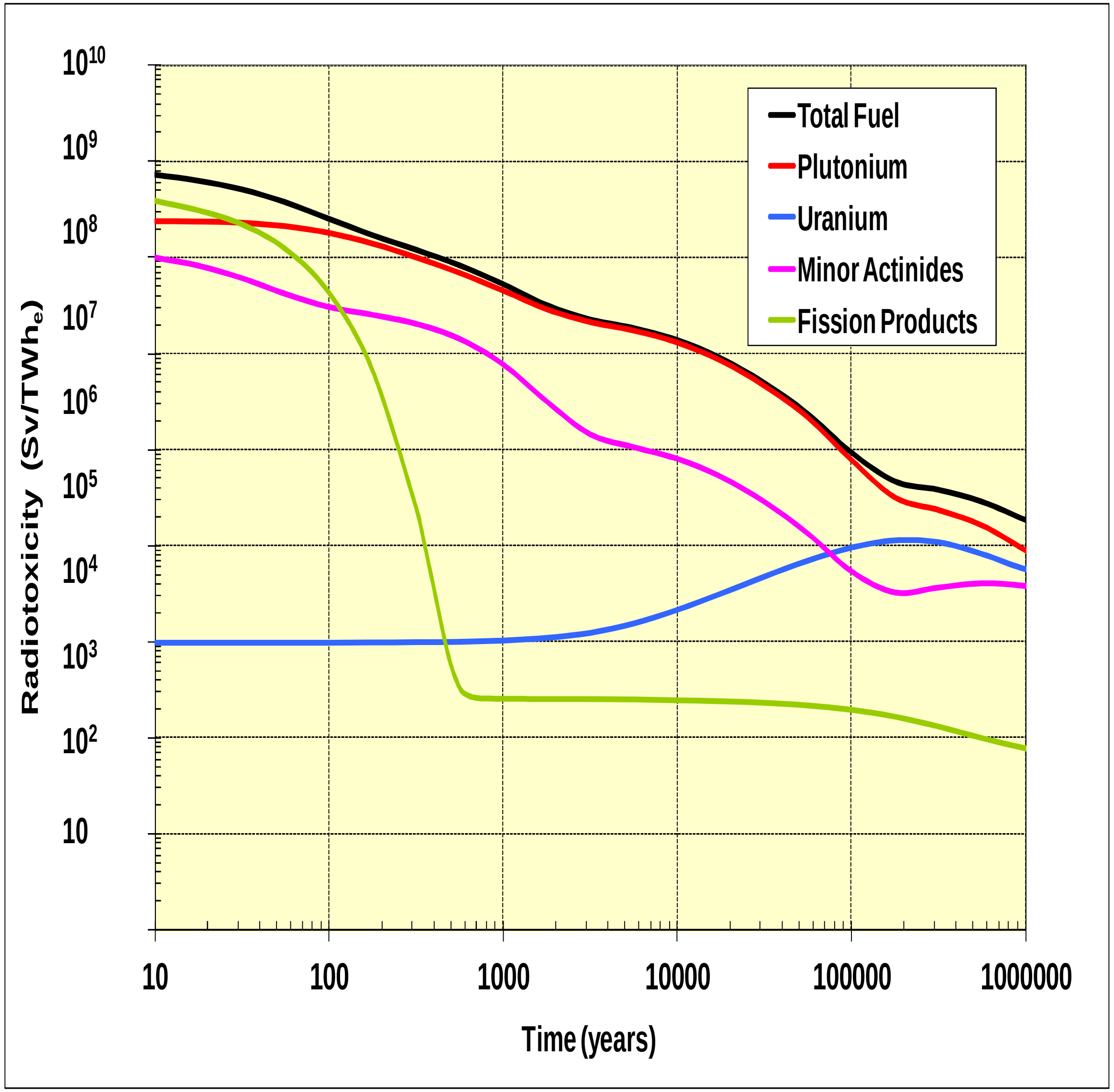


Baromètre IRSN,2013

against nuclear power, why ?



FINAL WASTE RADIOTOXICITY



MINOR ACTINIDES IN USED FUEL

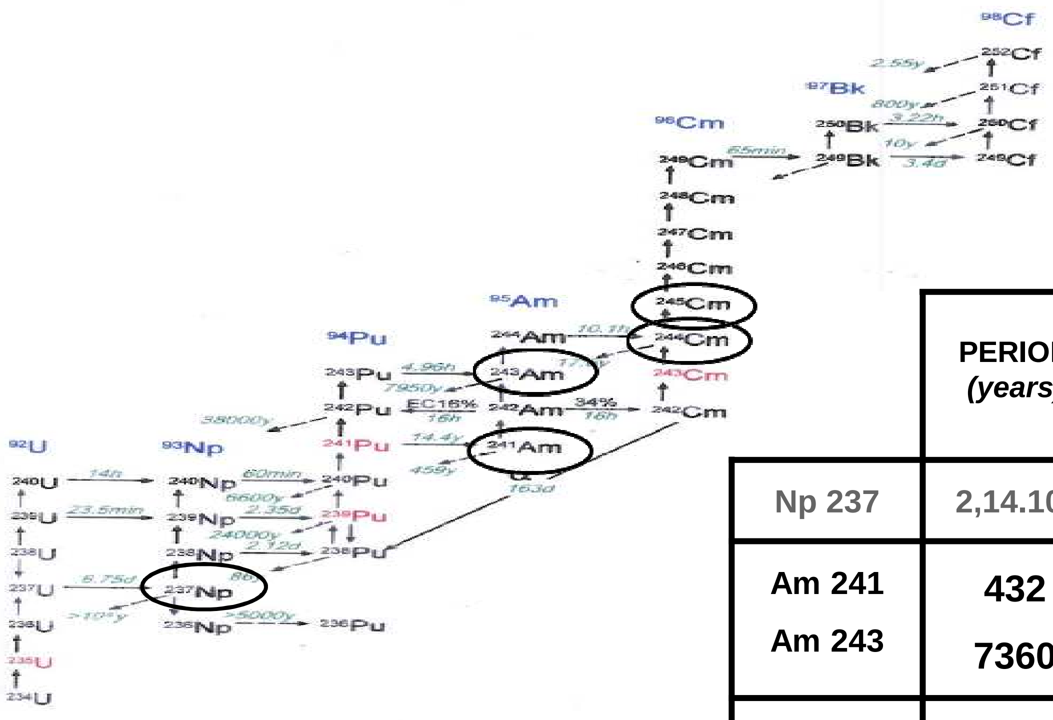


FIG. 5.22. — Transuranic element formati
Horizontally: number of protons; vertically: number
↑ neutron capture (n, γ) ↓ (n, 2n) reaction
→ β⁻ decay ← β⁺ decay or
↙ α decay electron capture (EC

	PERIOD (years)
Np 237	2,14.10⁶
Am 241	432
Am 243	7360
Cm 244	18
Cm 245	8500

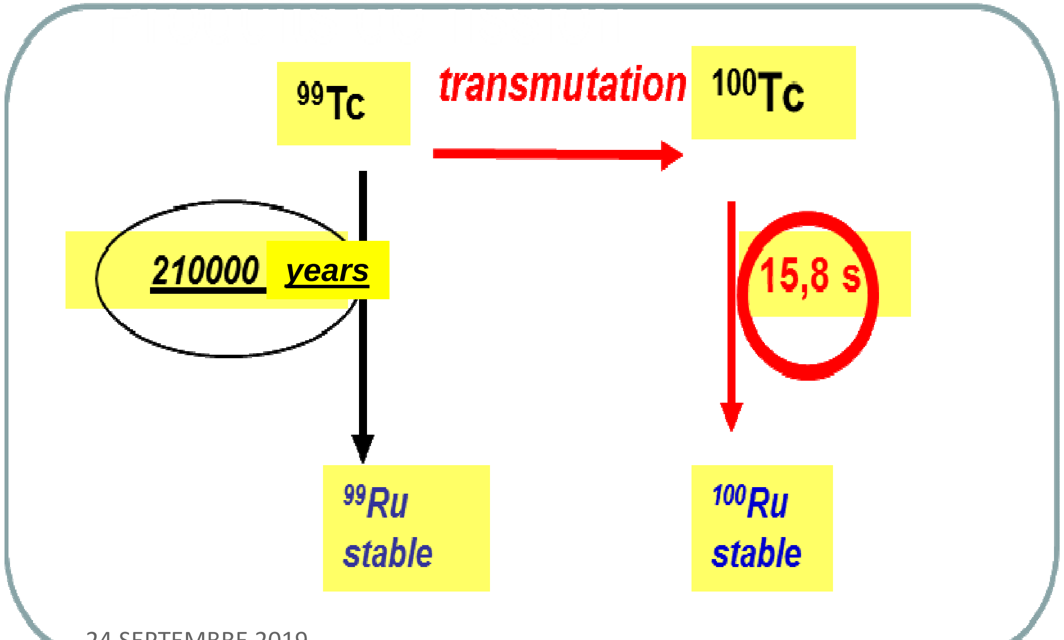
	UOX	MOX	FR (Astrid)
TC (GWj/t)	45	45	100
decay	5 years	5 years	5 years
Np (kg/TWh)	1.7	0.5	0.4
Am (kg/TWh)	1.9	15	3.7
Cm (Kg/TWh)	0.1	2.7	0.2

MOX used fuel decay	5 years	10 years	15 years	30 years
^{241}Am (kg/TWhe)	9.6	14.3	18	24.6

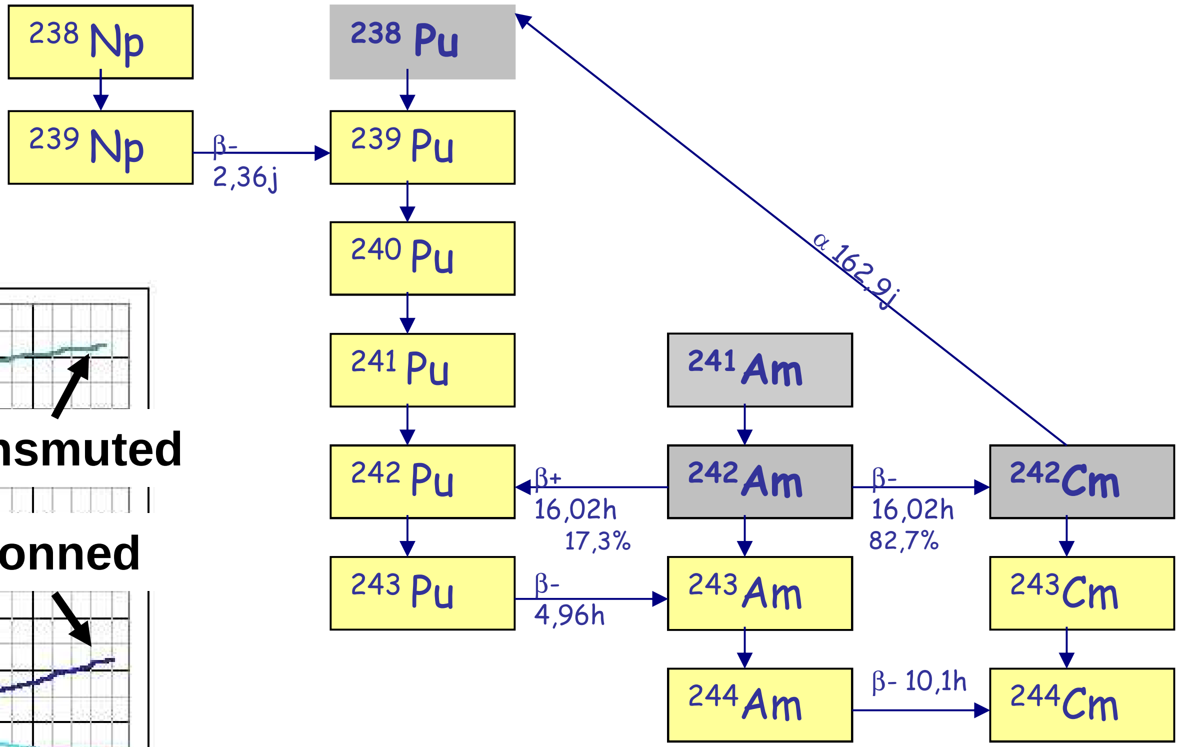
« TRANSMUTATION », WHAT IS THAT ?

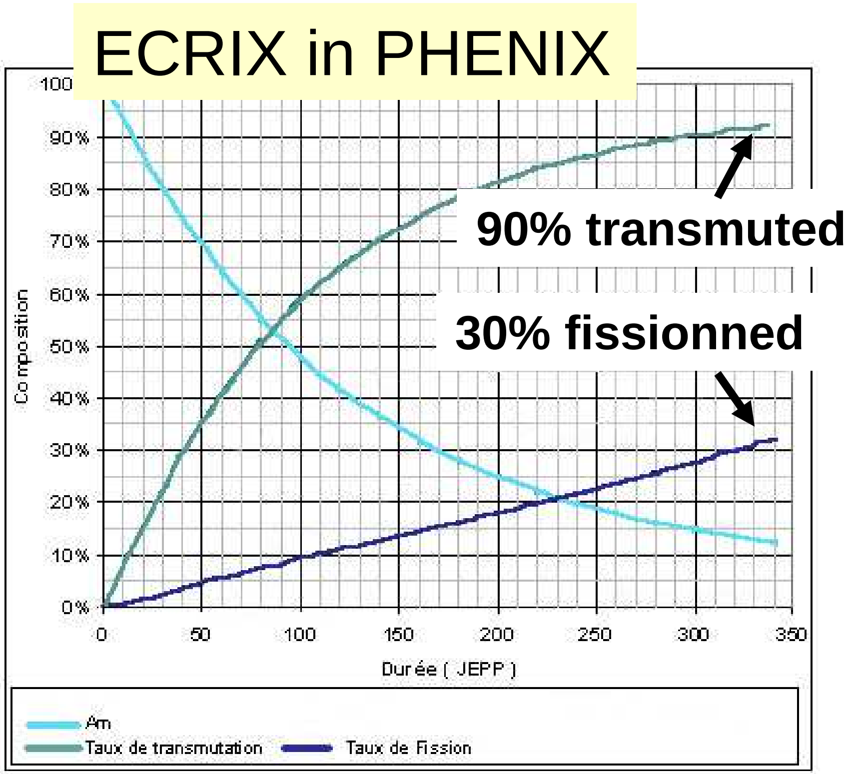
(1) LLRN (or precursors) <u>partitioning</u>

(2) LLRN <u>transformation into short-lived radionuclides</u>



^{241} Am TRANSMUTATION

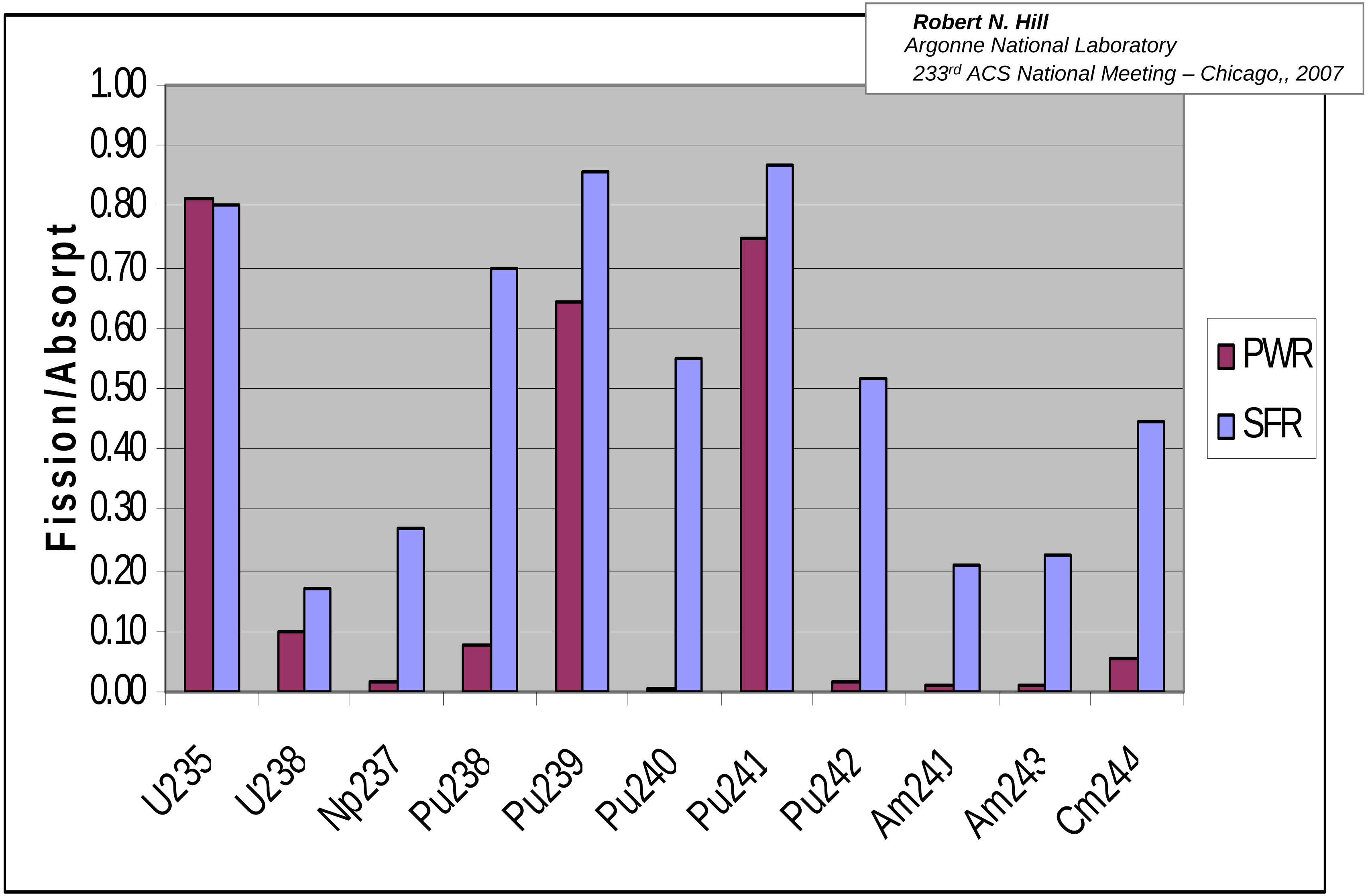




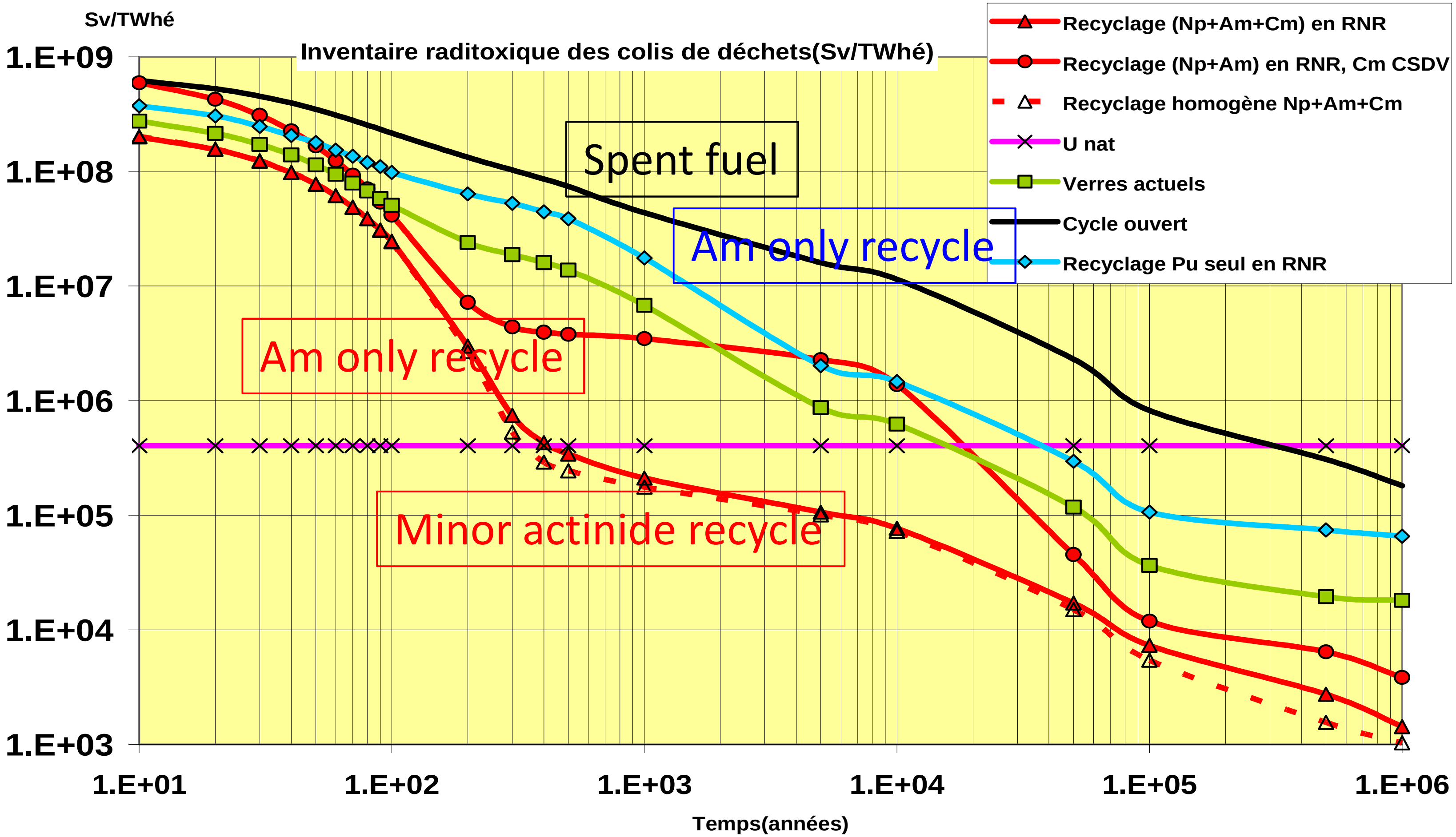
- only fission is useful

- if capture: formation of *other long-lived radiotoxic actinides (« déplace le problème »)*

MA TRANSMUTATION : WHY FAST NEUTRONS ?

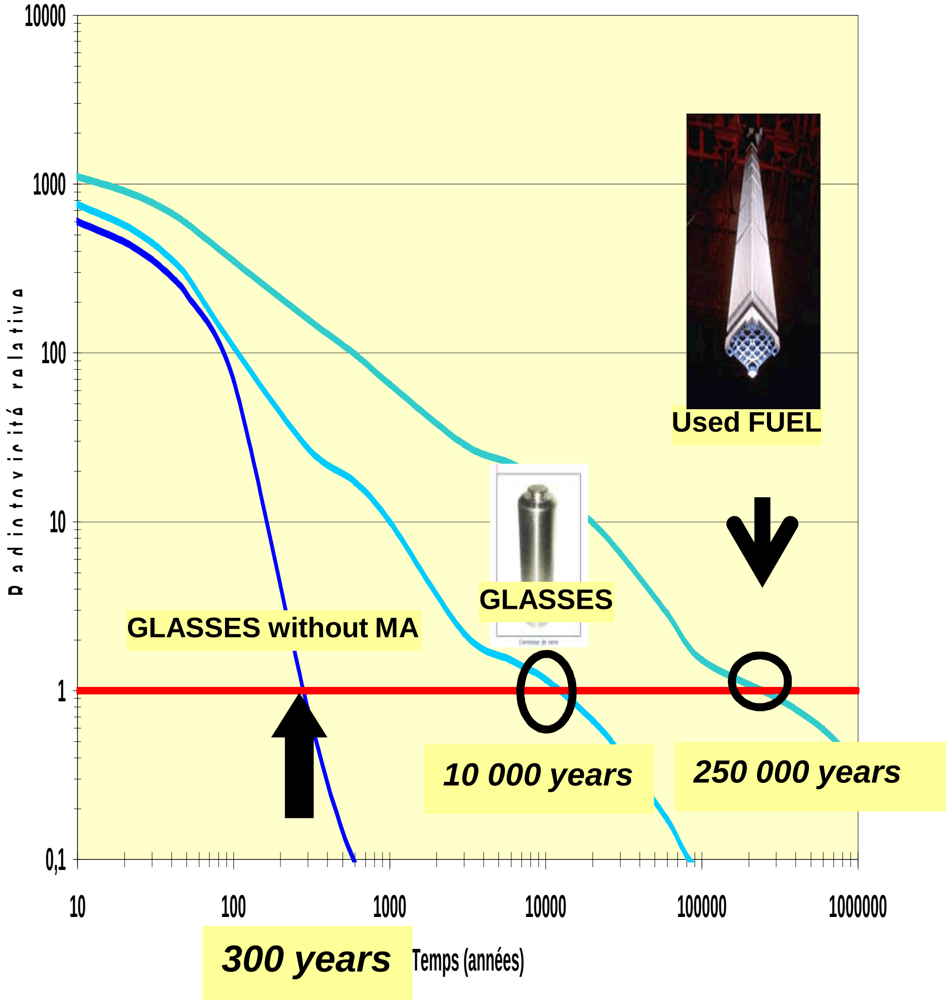


FINAL WASTE RADIOTOXICITY

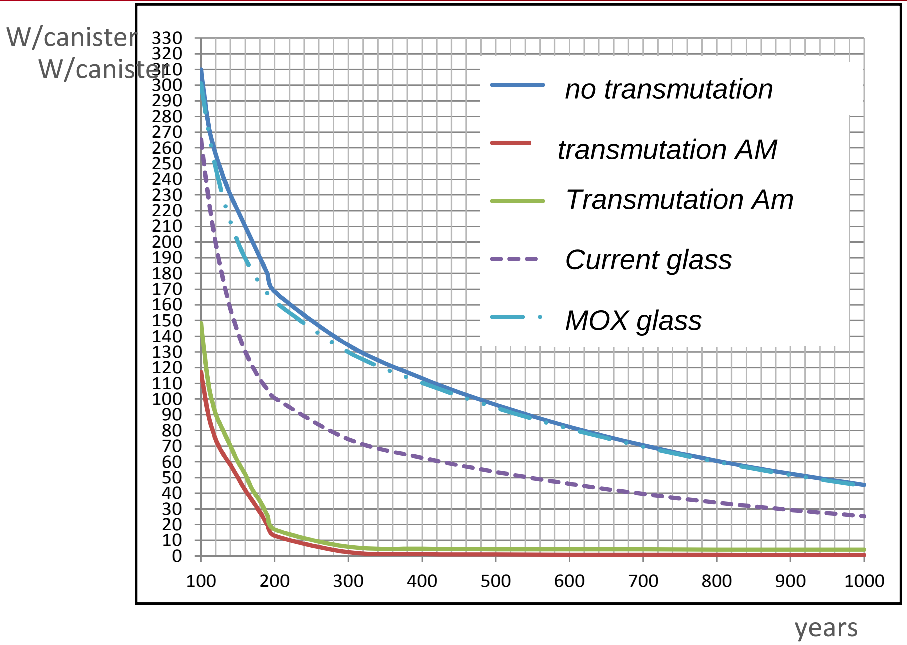


Minor actinide recycle : radiotoxicity decrease of about 200 times [*300years-10000years]*
Am only recycle no Cm recycle): smaller radiotoxicity reduction.

FINAL WASTE FEATURES FOR DIVERSE OPTIONS



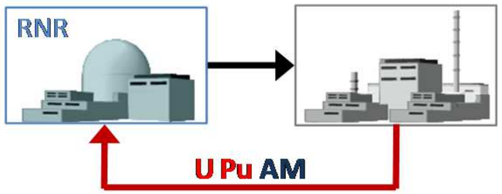
FINAL WASTE:
RESIDUAL RADIOTOXICITY



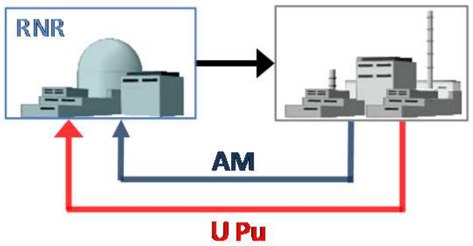
FINAL WASTE:
RESIDUAL HEAT

TRANSMUTATION: *HOW ?*

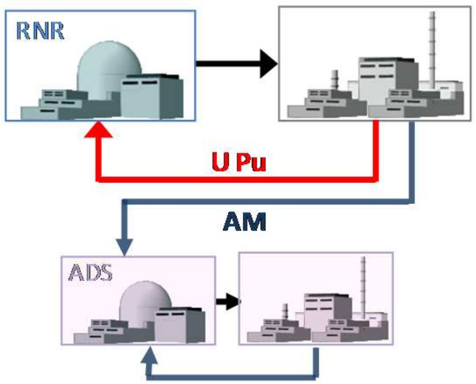
- **<u>HOMOGENEOUS MODE</u>**: ***MA diluted in fuel***



- **<u>HETEROGENEOUS MODE</u>** : ***MA in dedicated fuels***
  (ex : MA-bearing blankets)

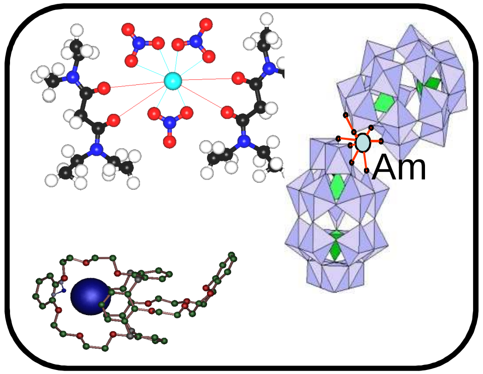


- **<u>DEDICATED STRATA</u>** : ***MA transmuted in specific devices***
  (ex: ADS)

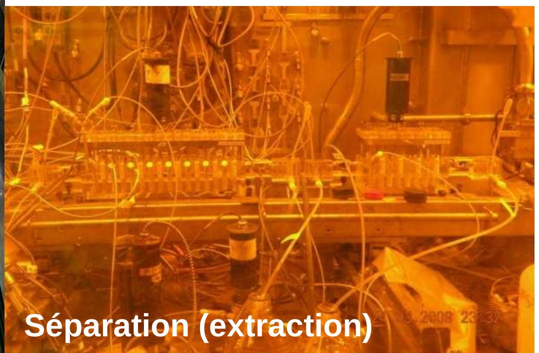


MINOR ACTINIDE RECOVERY

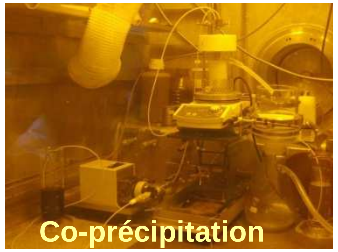
- additional steps to PUREX
- new extracting molecules,
  designed for MA selective extraction
- new separation processes
- laboratory experiments , genuine fuels, kg scale

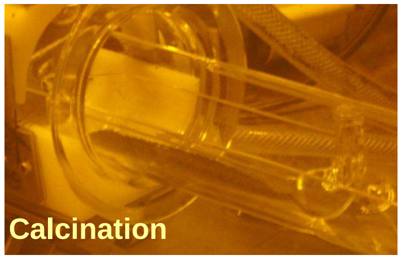


ATALANTE experiments, 2000-2012



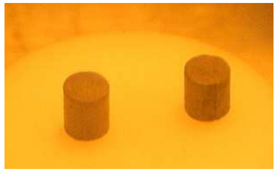
(plusieurs kg, 2005 - 2010)

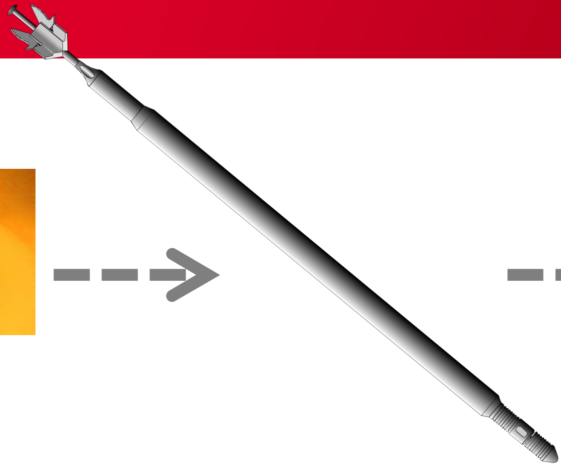


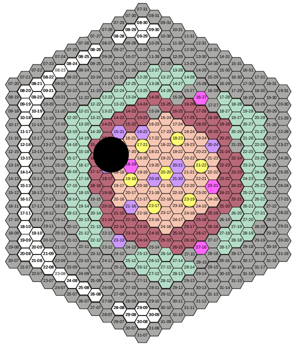


(dizaines de g, 2012)

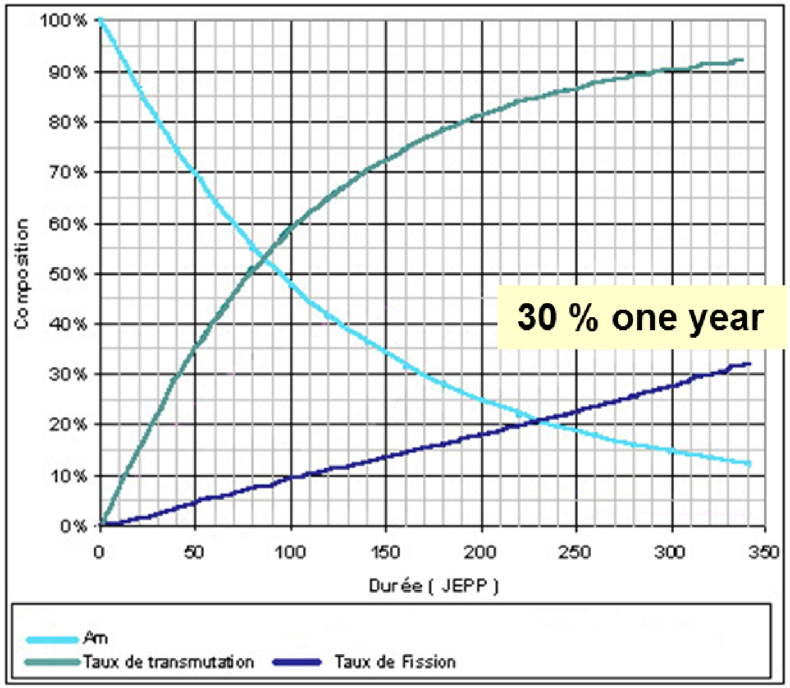
MINOR ACTINIDE TRANSMUTATION

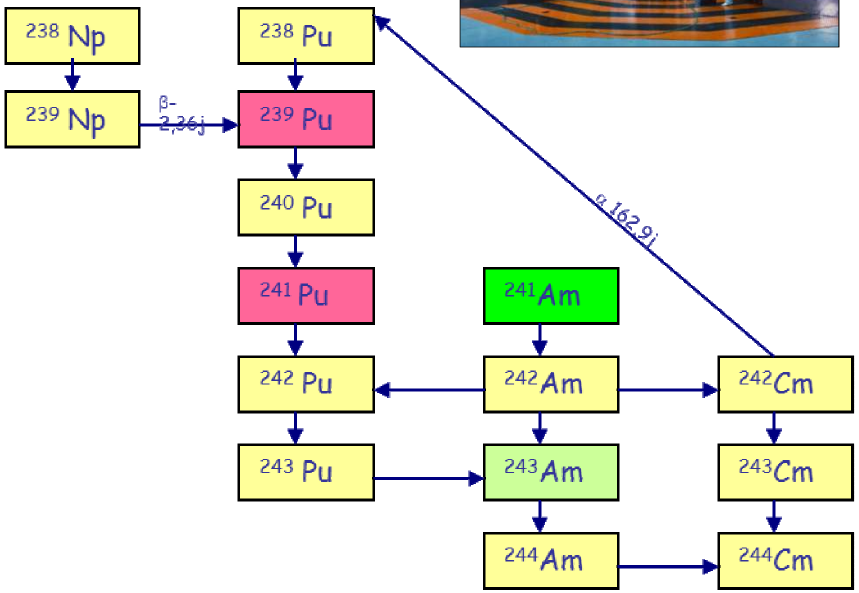






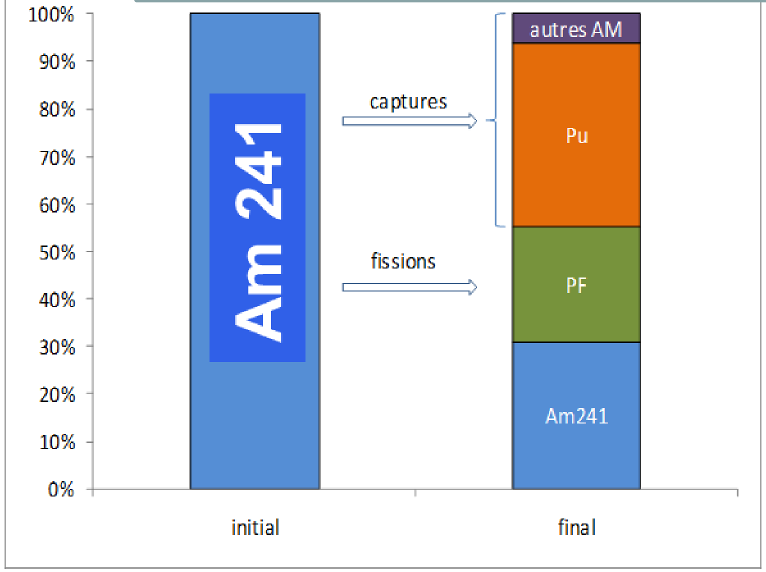
TRANSMUTATION EXP. IN PHENIX

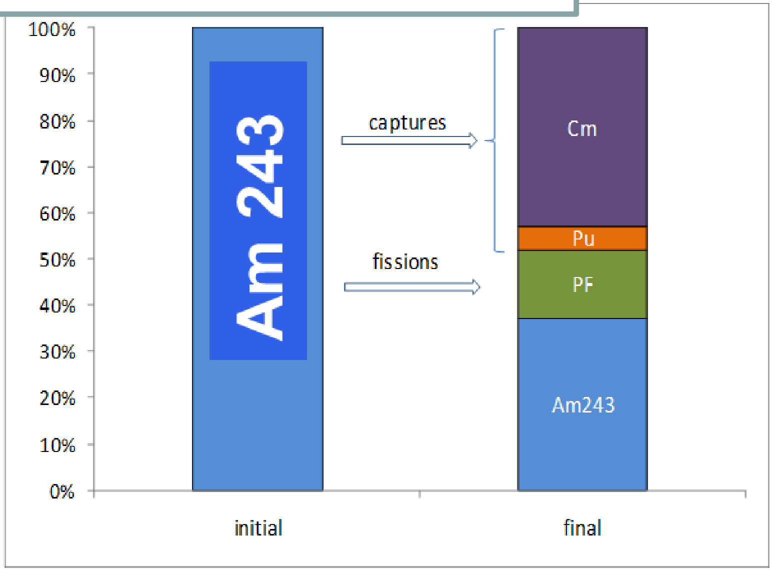




MINOR ACTINIDE TRANSMUTATION

CEA calculations, 5 years in FR core (ASTRID)

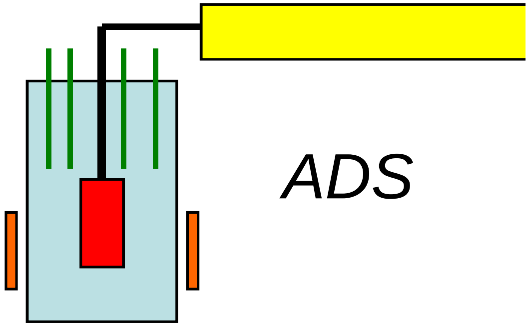




Dedicated transmutation devices:

-decoupling power generation/waste management

- *sub-critical core safer with high MA content*



ADS

(MYRRHA project, SCK-CEN)