



*The Abdus Salam
International Centre for Theoretical Physics*



2055-19

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

9 - 20 November 2009

Reprocessing and recycling of fast reactor fuel

Dominique Warin
*CEA/Nuclear Energy Division
Radiochemistry and Processes Department Marcoule
France*



IAEA/ICTP School on Physics and Technology of Fast Reactor Systems

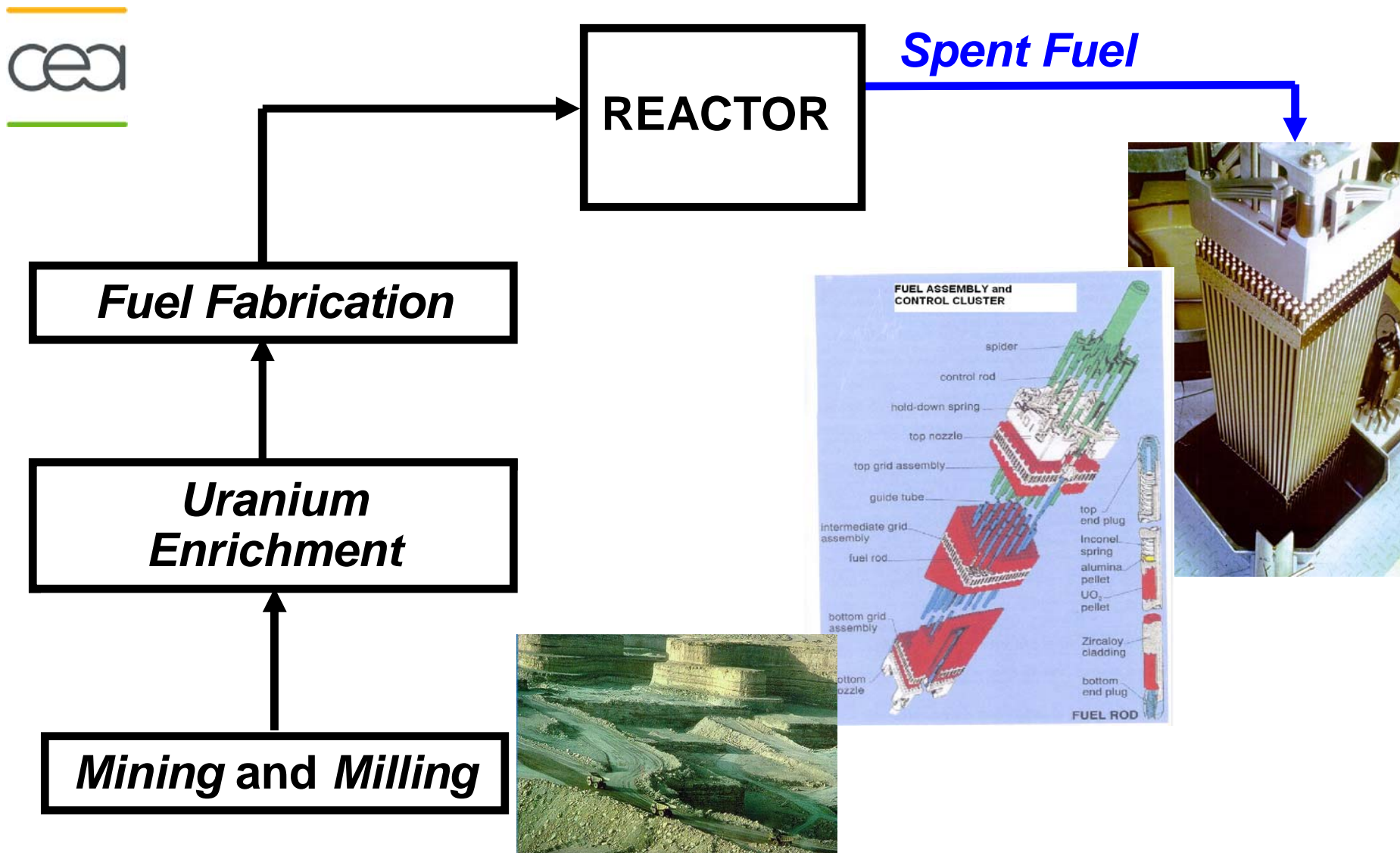
Reprocessing and recycling of fast reactor fuel

Dominique Warin

CEA / Nuclear Energy Division
Radiochemistry and Processes Department

Marcoule, France
dominique.warin@cea.fr

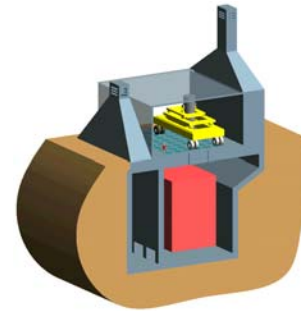
The nuclear fuel cycle



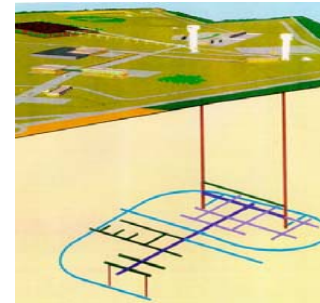
Spent fuel management : what options ?

cea

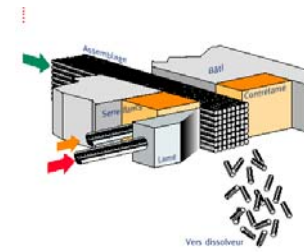
- Interim storage



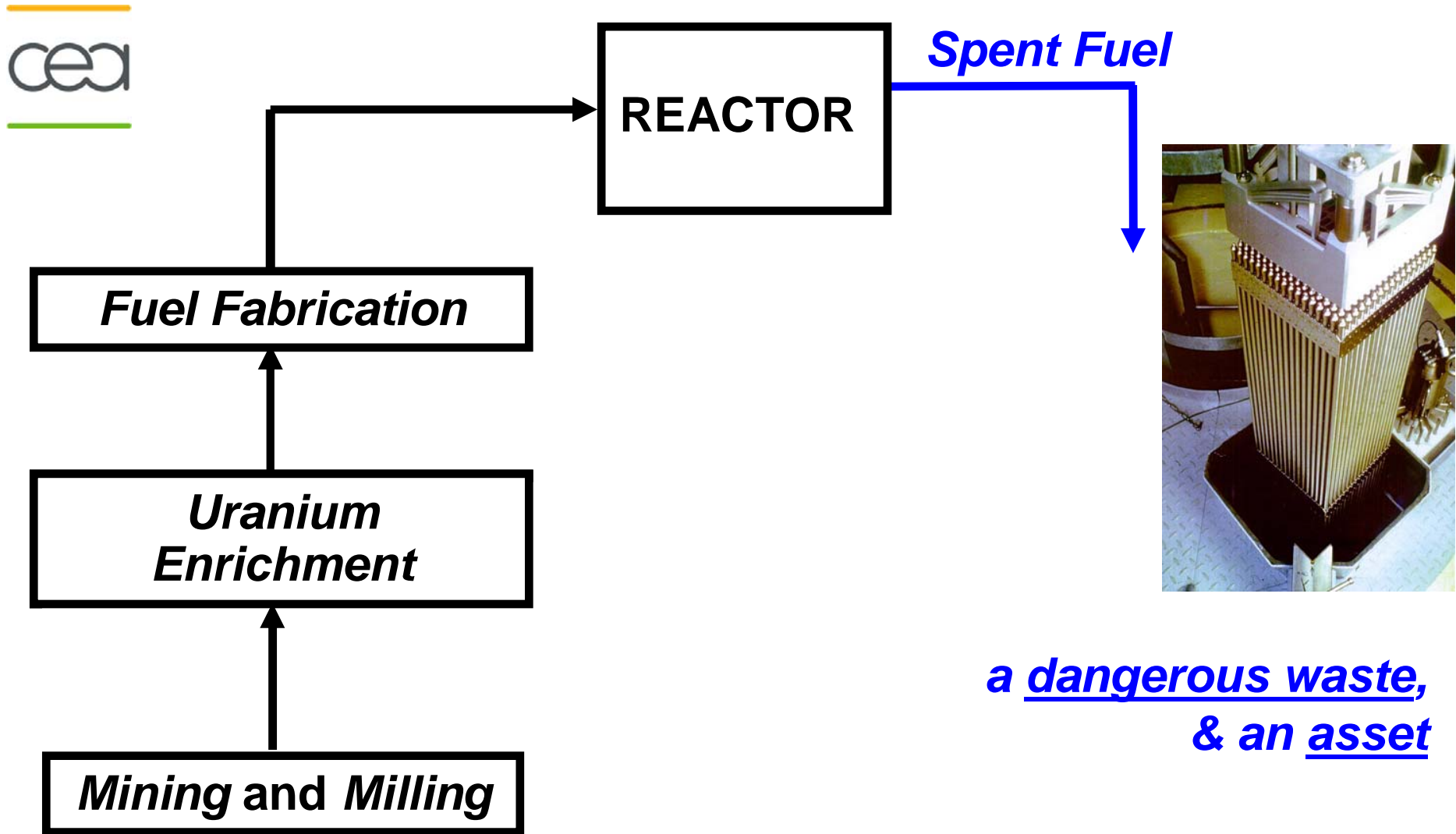
- Direct disposal



- Process and recycle

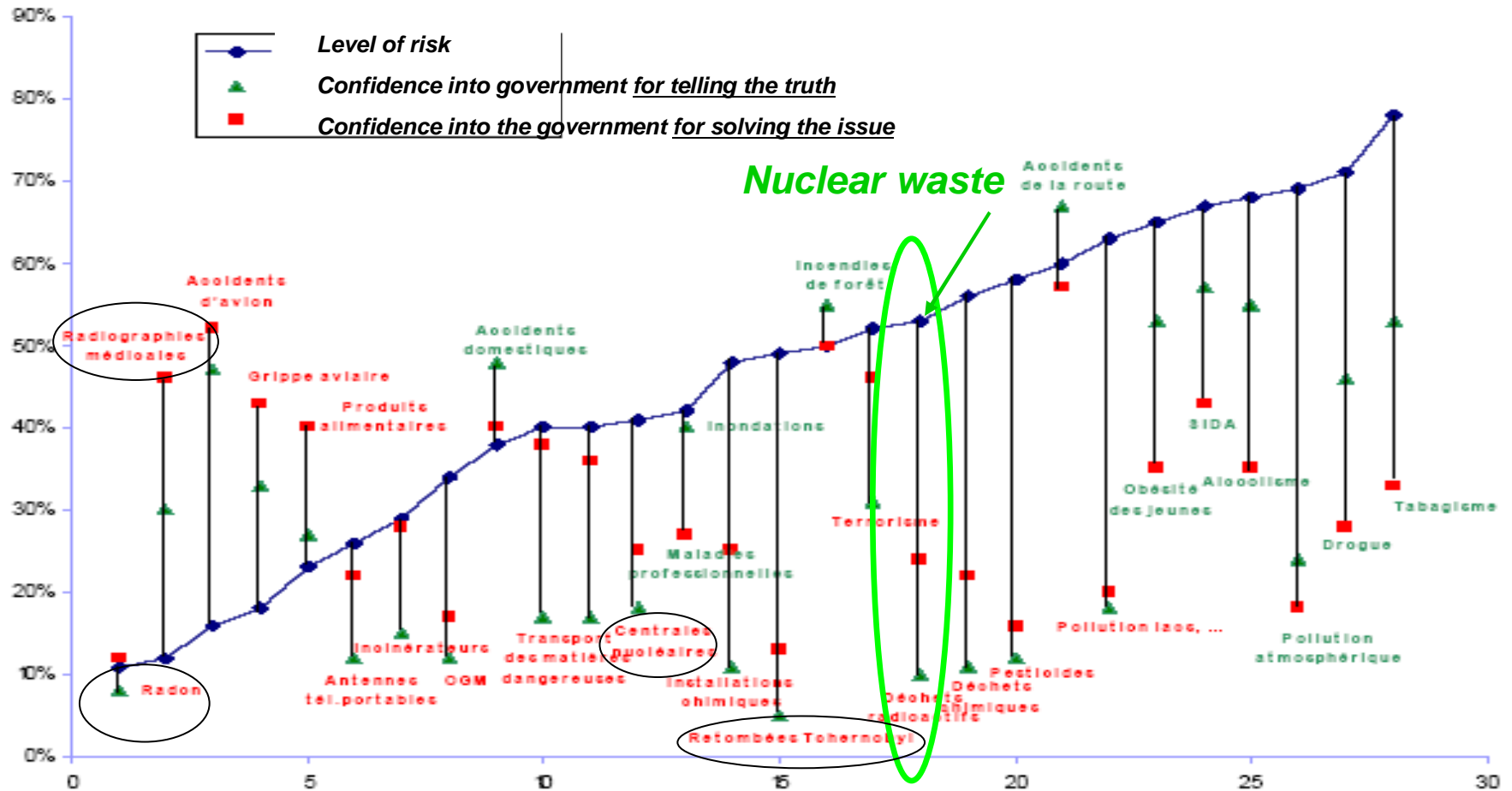


The nuclear fuel cycle

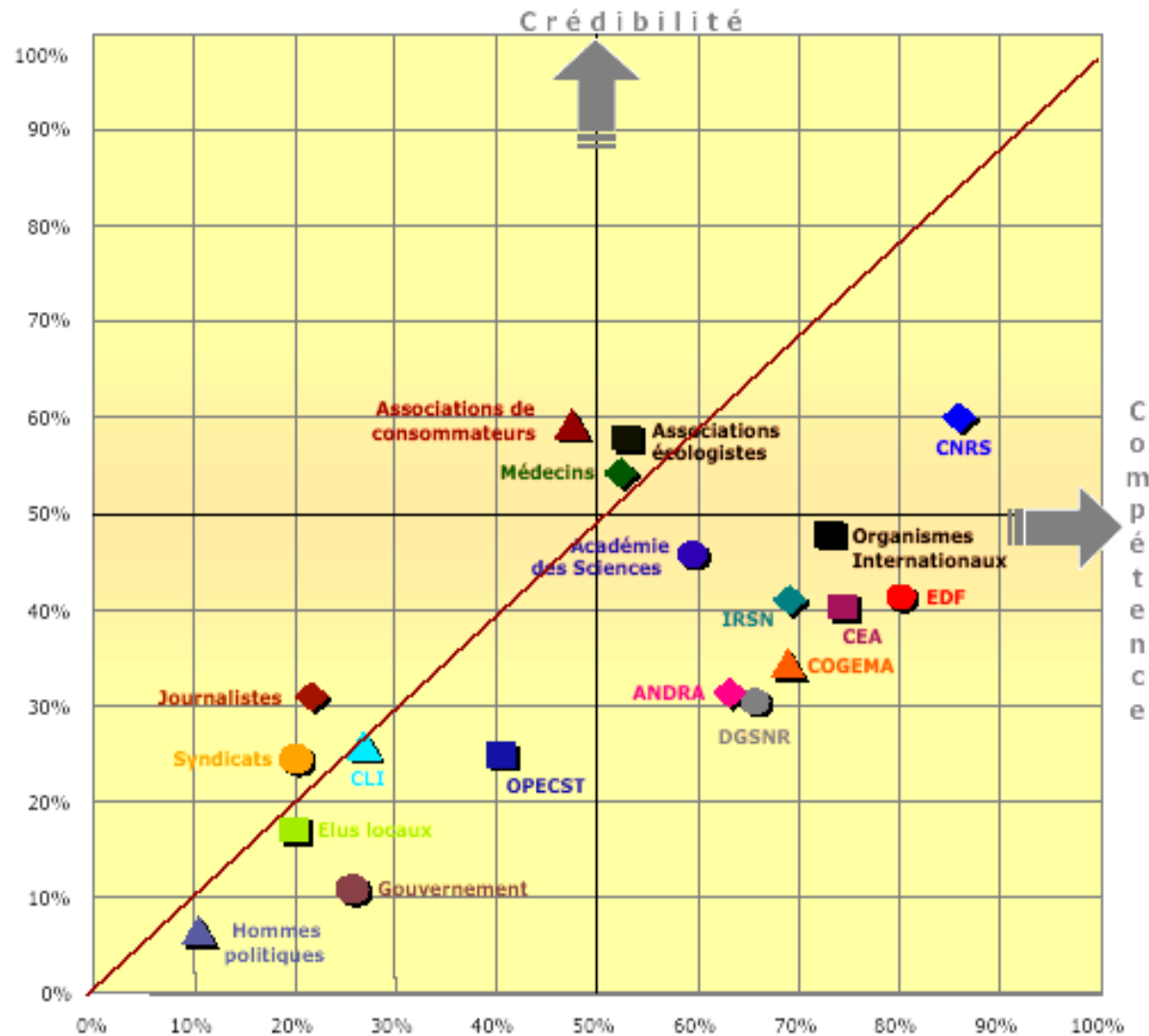


About risks...

Source: "IRSN barometer" 2006



Nuclear acceptance : credibility and competence



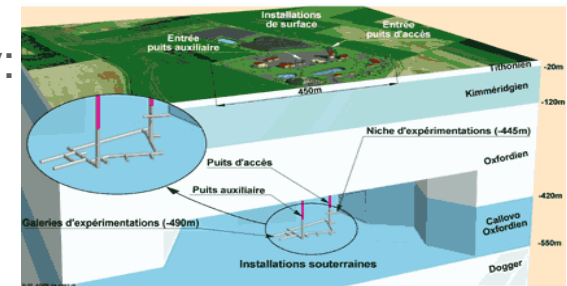
The French Act of December 1991, for 15 years

Waste management in France: which goals?

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

Goal 2 research : to study the feasibility of reversible or irreversible repository in deep geological formations

Positive results of Underground Laboratory:
Dossier Argile 2005 : clay in Meuse/Haute Marne site



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

Sub surface long term storage demonstration gallery constructed at CEA-Marcoule ; long term storage demonstration containers (scale 1)



⇒ **Deadline set in law : 2006**

The main results of P and T research programs

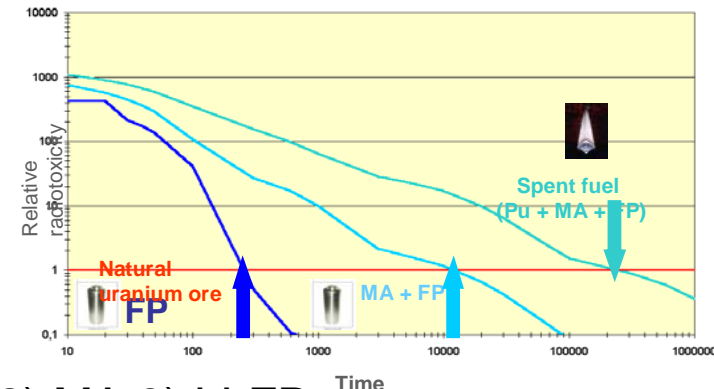


- **New possibilities, continuous progress :**
 - Extraction molecules have been developed at lab scale, and pre industrial demonstrations have proved successful : **industrial partitioning is feasible**
 - Transmutation of Fission Products (I, Cs, Tc) is either not feasible or unrealistic
 - MAs transmutation is unrealistic in PWR
 - Transmutation 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**
 - Feasibility of transmutation in ADS is not obtained today, and research must go on
 - **Realistic scenario : Transmutation can be carried out only with the new generation of fast neutron reactors, as a contribution to sustainable nuclear energy by homogeneous or heterogeneous mode**

Recycling strategy



- Aim of recycling : to minimize the **quantity and radiotoxicity** of long lived nuclear waste
- Potential gain : to reduce at hundreds years the life time of future waste (compared to the level of initial U_{nat}) and to ease the conditions of a deep geological repository



- Main radiotoxicity contributors : 1) Pu 2) MA 3) LLFPs (years)
- Fundamental hypothesis : closed cycle, GEN IV fast neutron reactor implemented at mid or long term (whatever the cooling : Na, gaz,...)
- Reference strategy :
 - PWRs: Pu management by multi recycling
 - GEN IV SFR or GFR: full actinides recycling, beginning 2040
 - Option of the « double strata » PWRs – Accelerator Driven Systems
- Demonstrations of scientific and technical feasibilities,

before pre industrial development of recycling

Recycling strategy : systems evolution...



(Fuel cycle)

- The new requirements :

- Save resource
- Minimize environment impact (*waste nocivity*)
- Increase resistance vs proliferation risks

- New purposes, other than electricity (*heat, H2, dessalinization*)...
- *And obviously : safety, costs...*

(Reactors)

Save resource : Uranium ore

cea

Ore: 0.5 -10 kg U/ ton



$^{235}\text{U} : 0.7\%$
 $^{238}\text{U} : 99.3\%$

Average earth crust : few grams U/ ton

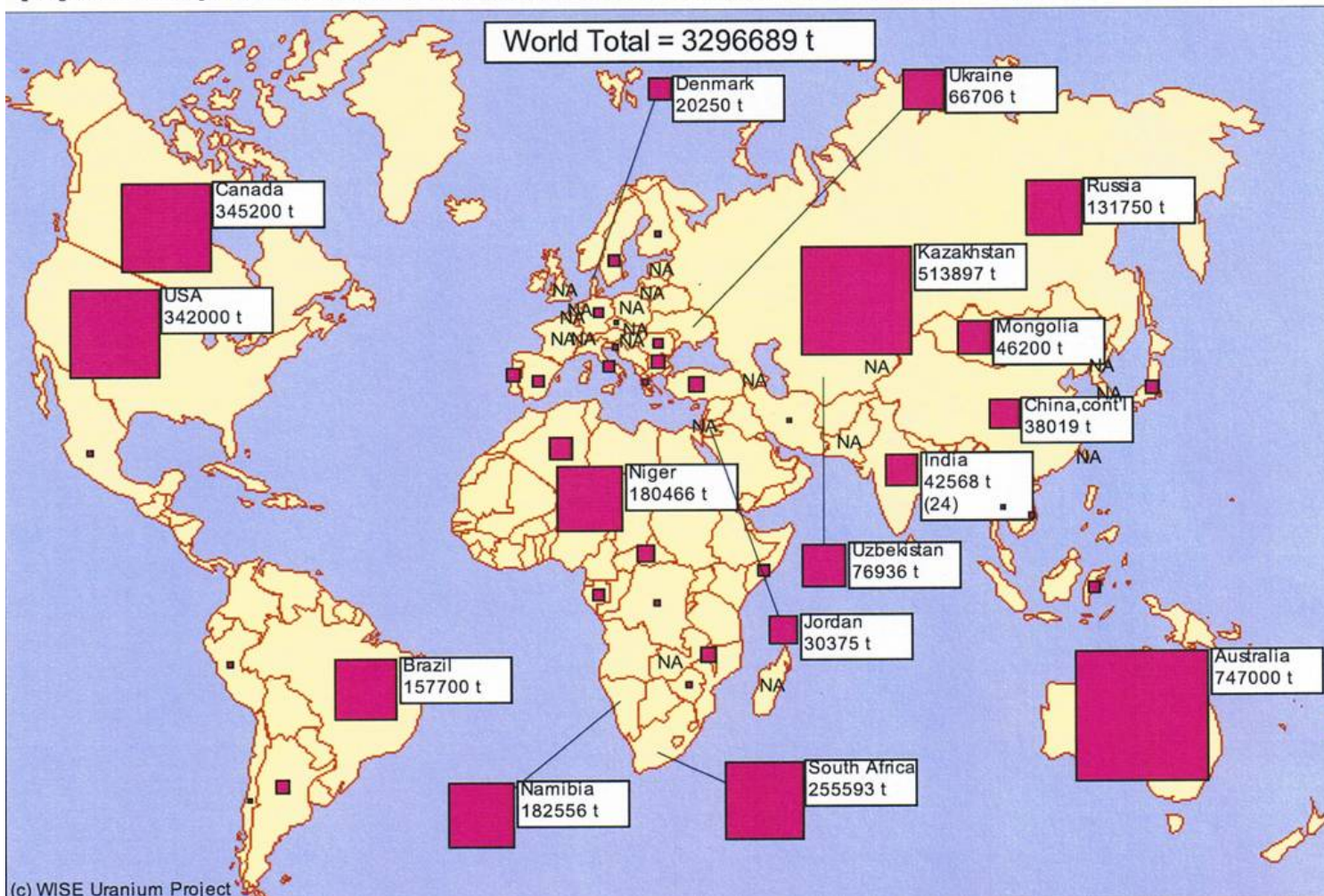
Seawater: few mg U / ton water

Save resource : Uranium resources



Uranium Resources (RAR - \$130/kg U)

[t U] Reasonably Assured Resources, recoverable res. as of 1/1/2005, Cost range < US\$130/kg U (OECD 2006)



Uranium world production and consumption

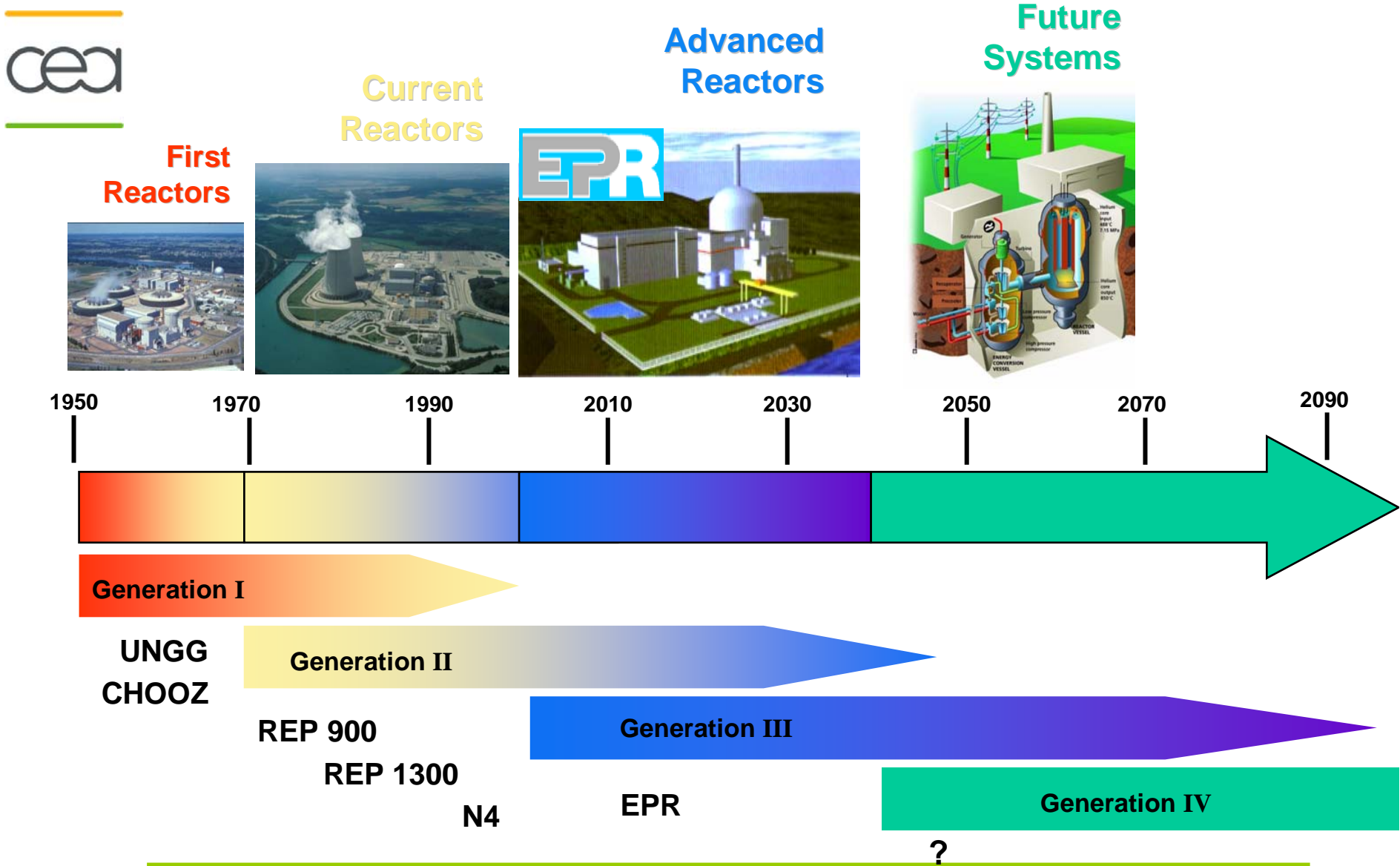


Production et consommation d'uranium naturel dans le monde



2005 records : 41 600 tons produced
67 500 tons needed

The Evolution of Nuclear Power to fast systems



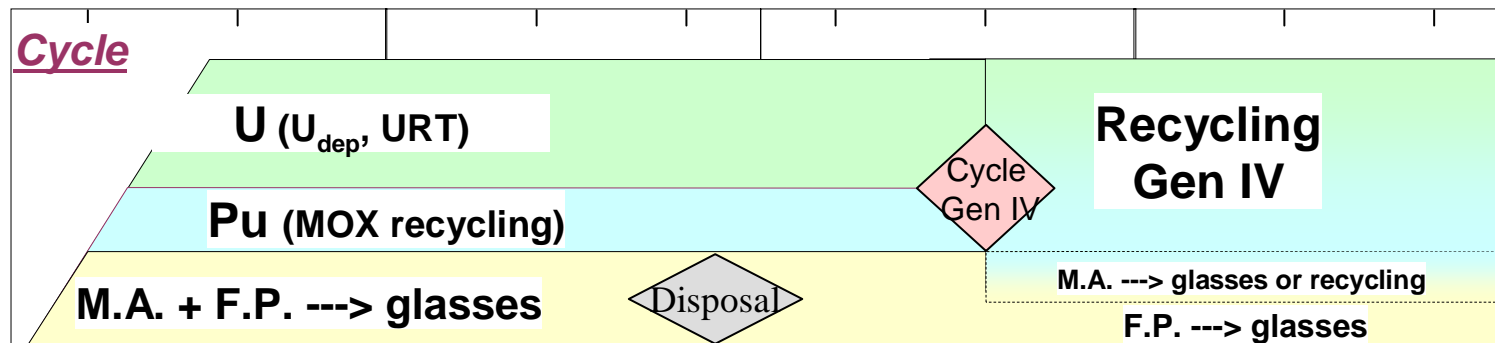
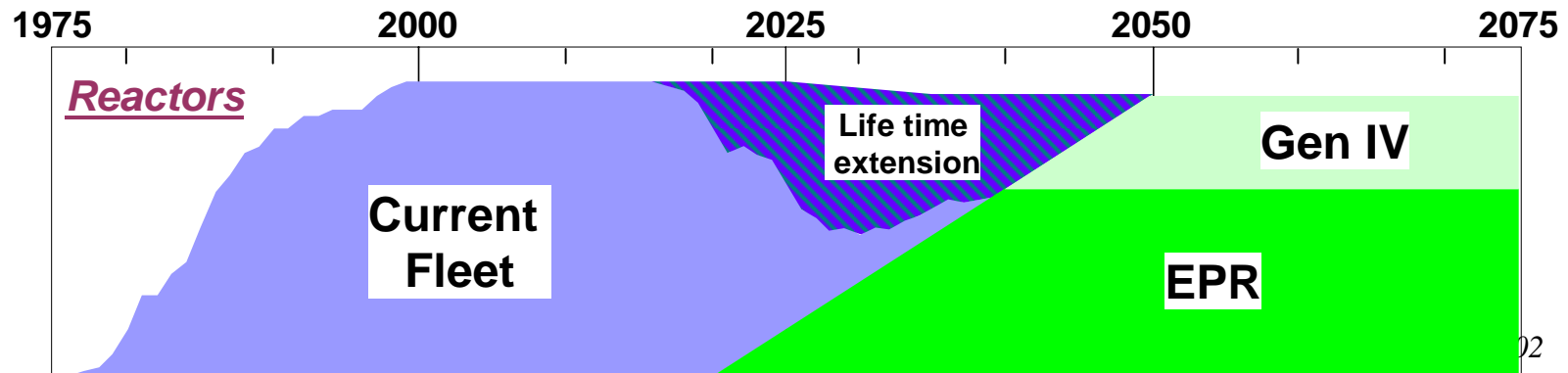
Transition scenarios between generations in France



➤ Major role of LWRs in the 21st century

- ❖ Current PWRs (Gen II): life time extension (> 40 years)
- ❖ Gen III/III+ PWRs : current PWRs replacement around 2015 (EPR) – Operation during 21st century

➤ A transition scenario between LWRs and Fast Neutrons Systems



Nuclear acceptance and a responsible management of spent fuel and waste



A responsible management of nuclear spent fuel :

- Recycles **96%** of spent fuel materials
- Saves **30%** of natural resources
- Costs less than **6%** of the kWh total cost
- Reduces by **5%** the amount of wastes
- Reduces by **10%** the waste radio toxicity
- Adapted technologies allow a safe conditioning of wastes to guarantee their long term confinement and stability, during dozens of thousands of years

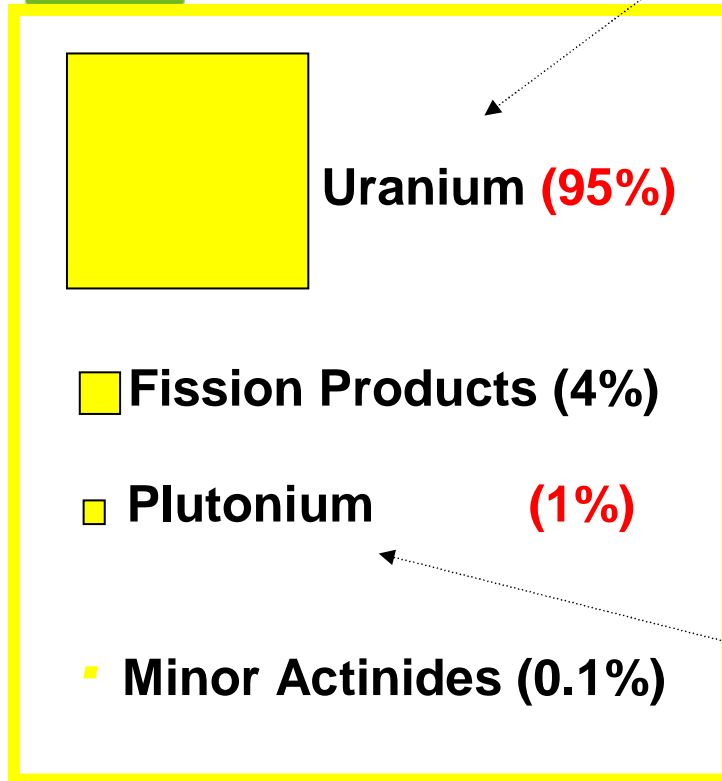


Recycling, such as implemented today in France, GB or Japan, gives time and opens a large range of options for the satisfactory management of nuclear wastes.



more sustainable policy
better public acceptance

The spent nuclear fuel



Average Content

$^{235}\text{U} = 0.9\%$



Fissile isotopes = 75%

UOX Nuclear spent fuel

(UOX : 3,5 % ^{235}U ; 33 GWj.t⁻¹)



U : 955 kg.t⁻¹
 Pu : 9,6 kg.t⁻¹
 AM : 0,8 kg.t⁻¹
 PF : 34 kg.t⁻¹

1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	<i>Ln</i>	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	<i>An</i>	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Uun								

LANTHANIDES	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
ACTINIDES	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

NOYAUX LOURDS

PRODUITS D'ACTIVATION

PRODUITS DE FISSION

PRODUITS DE FISSION ET D'ACTIVATION

UOX and MOX spent fuels : typical content



	UOX (45 GW/t)	MOX (45 GWj/t)
URANIUM	~ 940 kg	~ 890 kg
PLUTONIUM	~ 11 kg	~ 56 kg
FISSION PRODUCTS	~ 48 kg	~ 48 kg
MINOR ACTINIDES	~ 1 kg	~ 6 kg

(for 1 tonne initial HM)

Spent fuel : radioactive content

AVERAGE BU ~ 33 000 MWd/t



6 months after unloading : assembly : 150 000 TBq (18 kW)

3 years after : 30 000 TBq (3 kW)

FISSION PRODUCTS

β^- mainly

very diverse radioactive periods

CAPTURE PRODUCTS

Actinides (Pu 1 %, Minor Actinides ~ 0.4 %)

α and long radioactive periods

ACTIVATION PRODUCTS

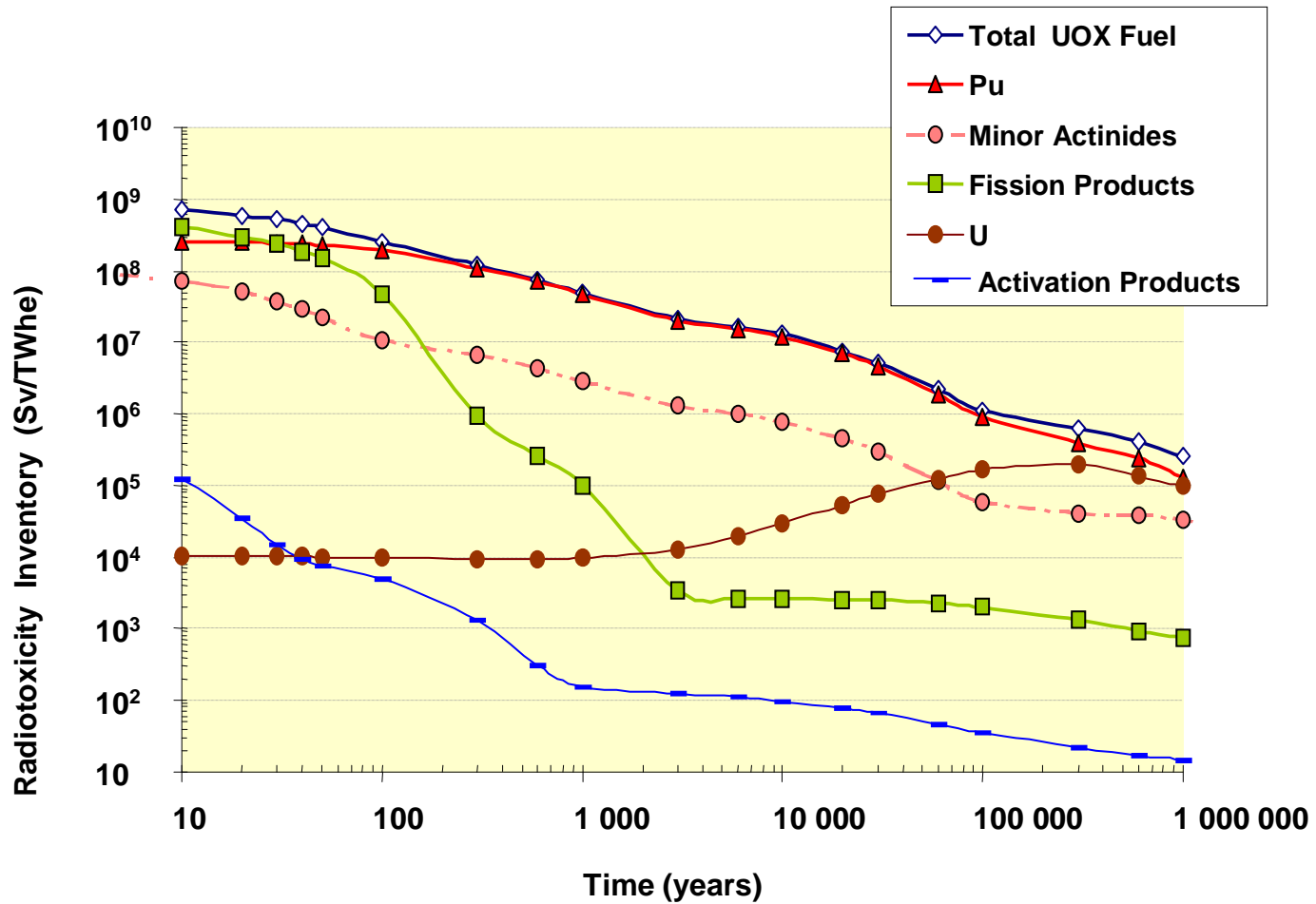
structure metallic materials (Zr isotops)

or fuel initial contaminants

(^{14}C from ^{13}N or ^{17}O traces in initial oxide)

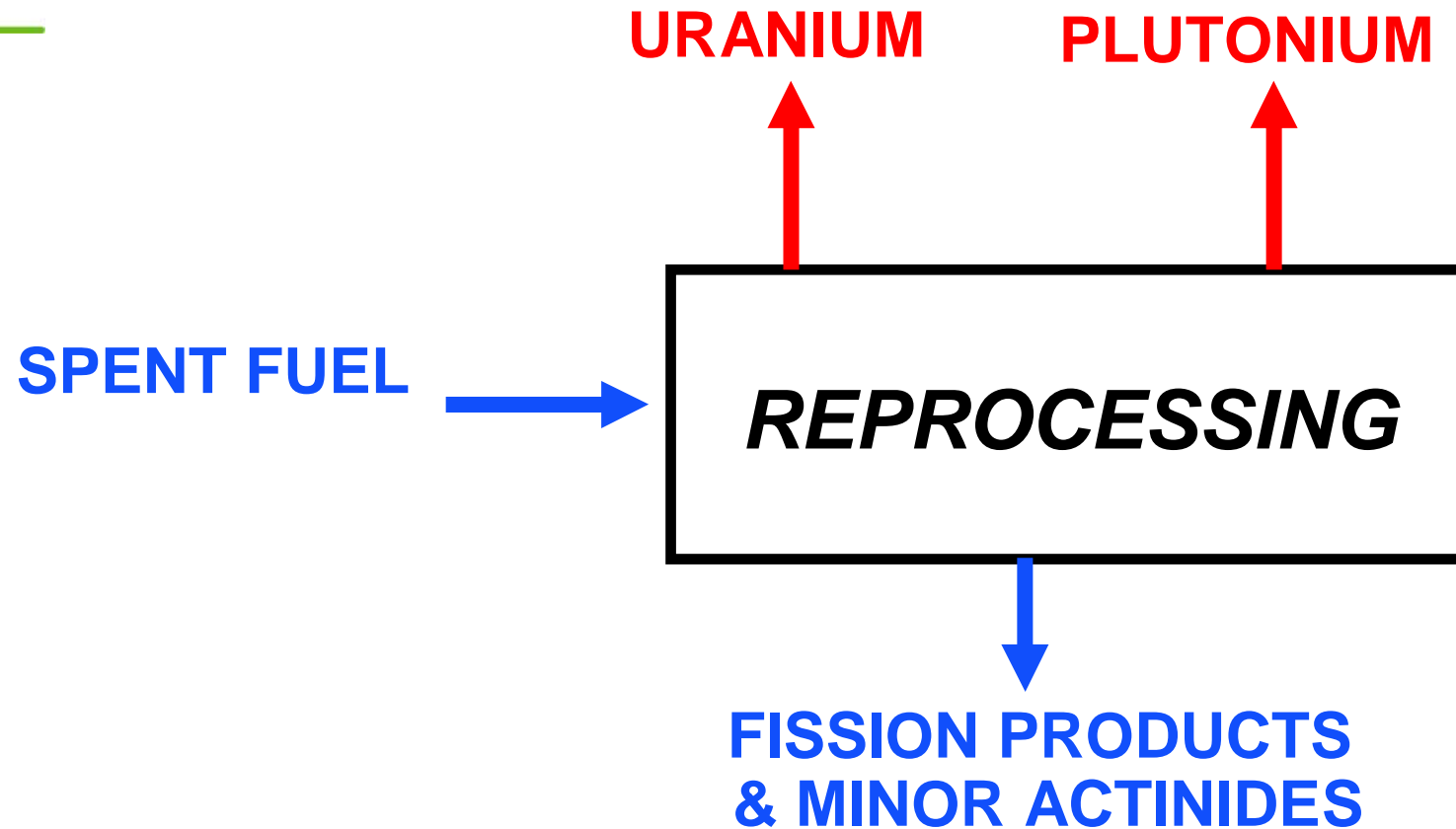
(^{36}Cl from ^{35}Cl traces in initial oxide)

Radiotoxicity of LWR-UOX Spent Fuel

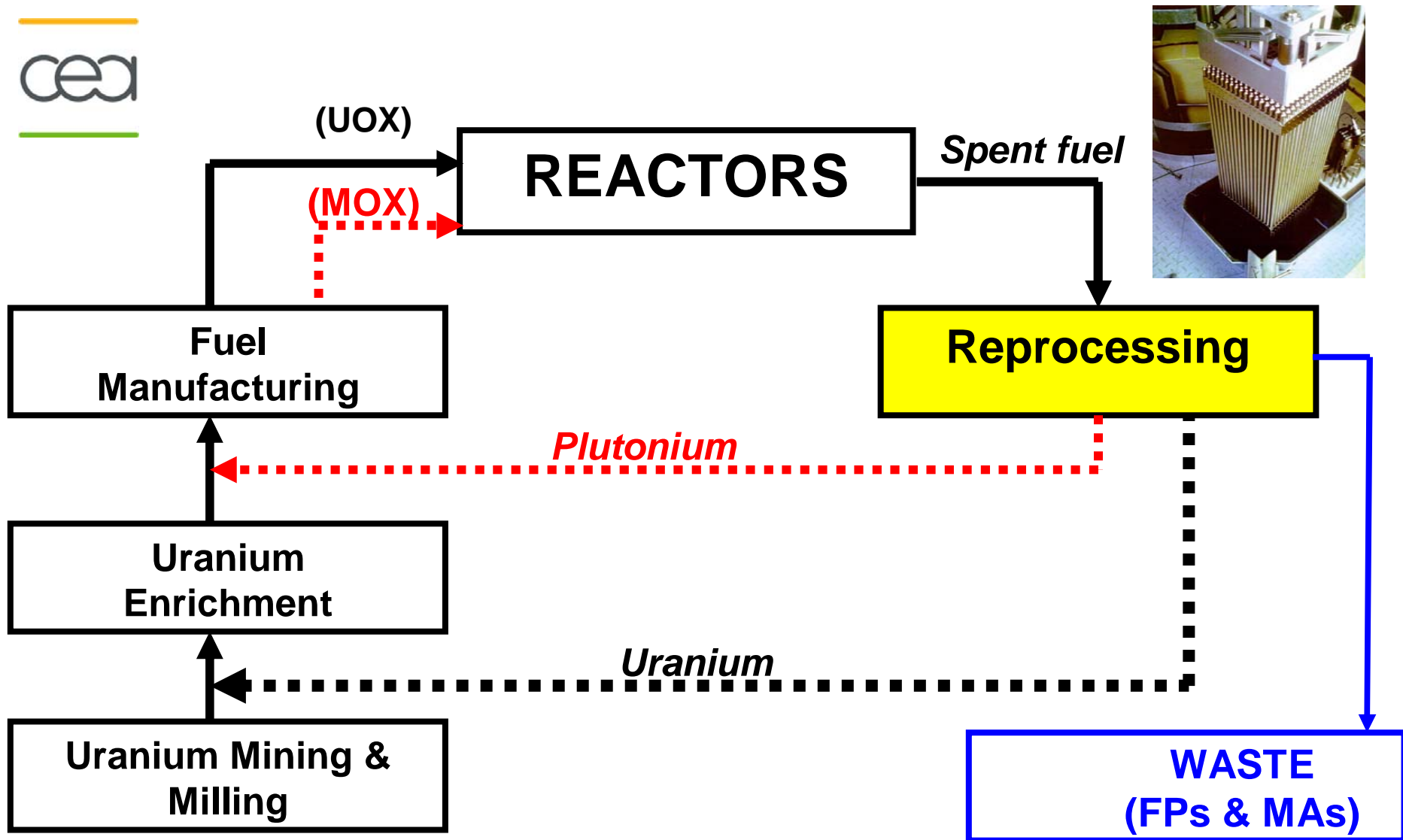


Ingestion Doses Coefficients from ICRP72

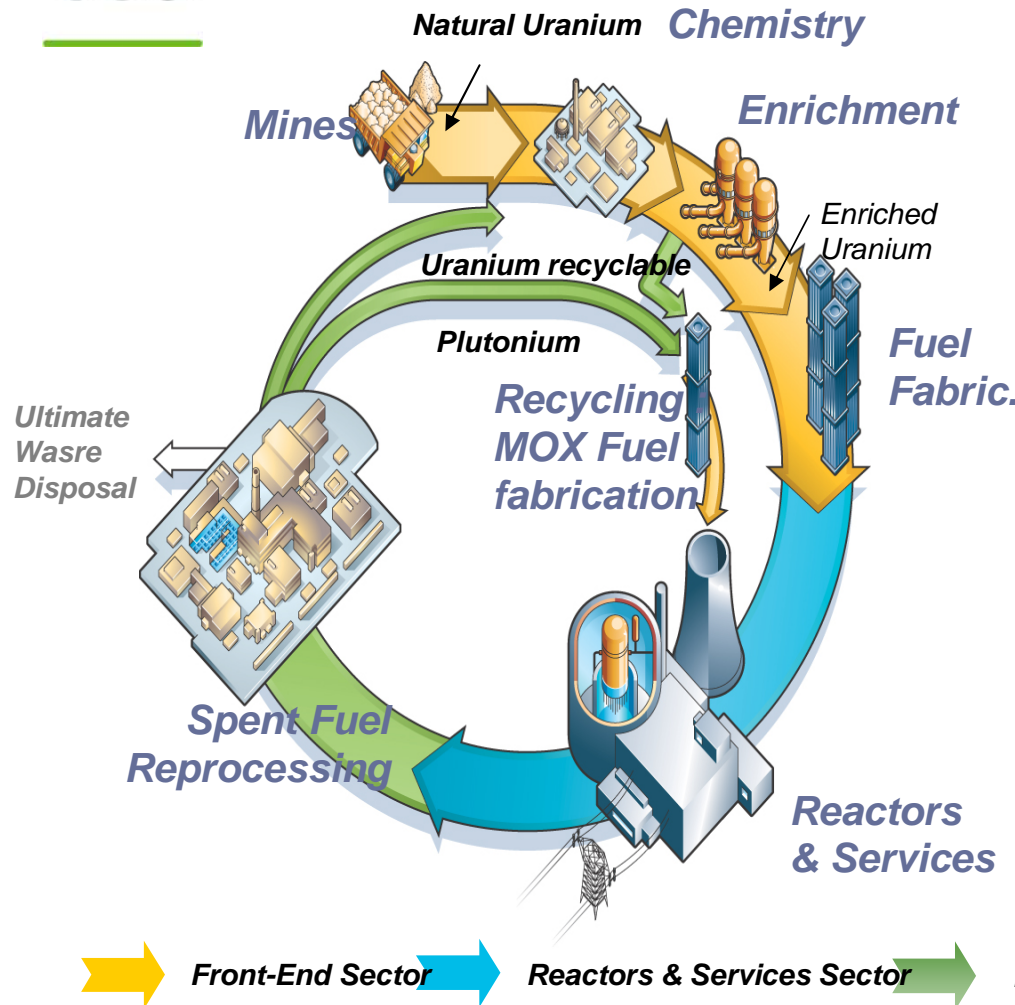
Reprocessing : the principle



Recycling Uranium and Plutonium



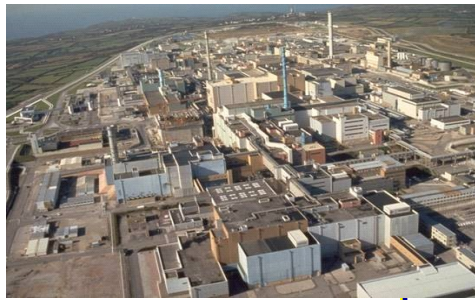
Closing the Fuel cycle ... an industrial reality



The exemple of the french situation :

- 58 PWRs → 400 TWh annually
- ~ 78 % of french electric production
- Fuel processing : more than 25 years of experience
- 1100 t_{HM} /yr of spent fuel discharged from the French PWRs
- 850 t_{HM} /yr of domestic spent fuel reprocessed + foreign
- Until now: ~ 20 000 t_{HM} spent fuel reprocessed and more than 1500 t_{HM} MOX fuel recycled

Fuel Reprocessing and recycling



LA HAGUE
UP2-UP3 PLANT
>21000 t processed

MARCOULE



MELOX PLANT
>1000t manufactured

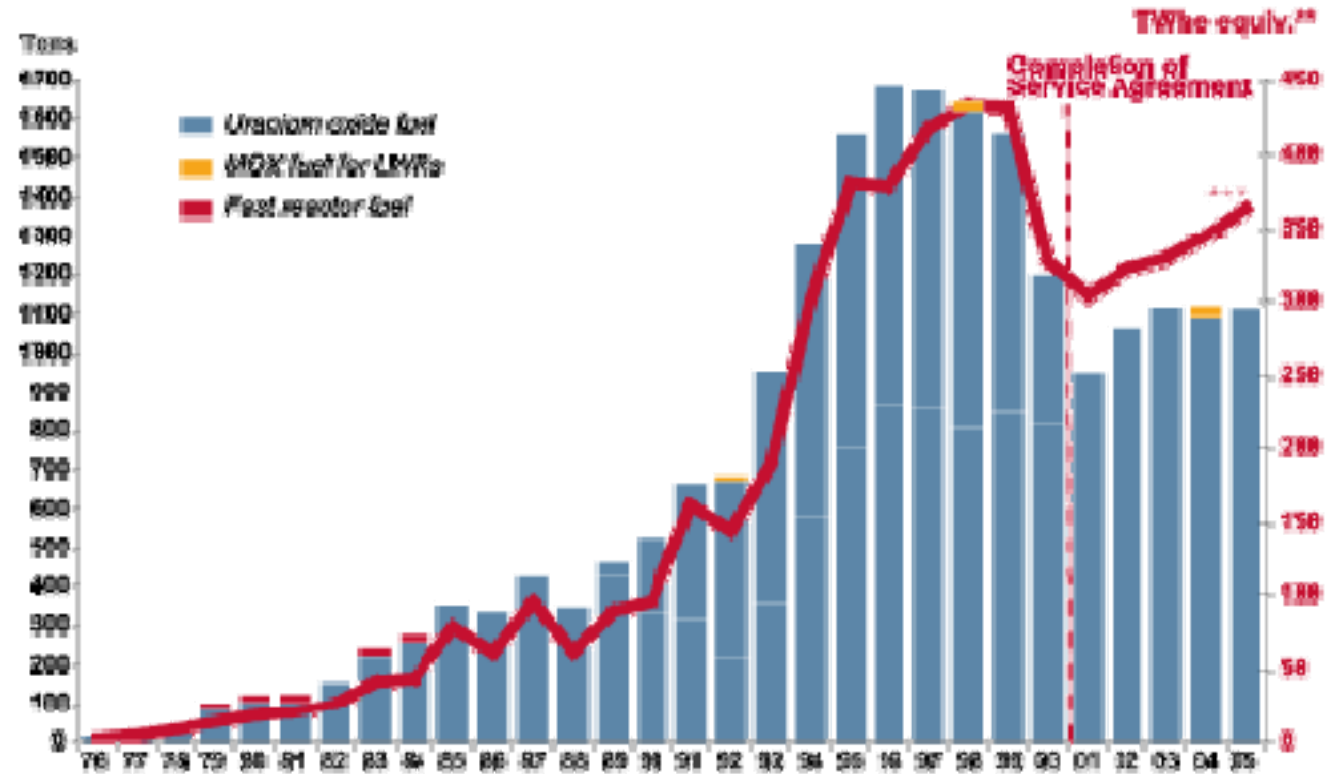
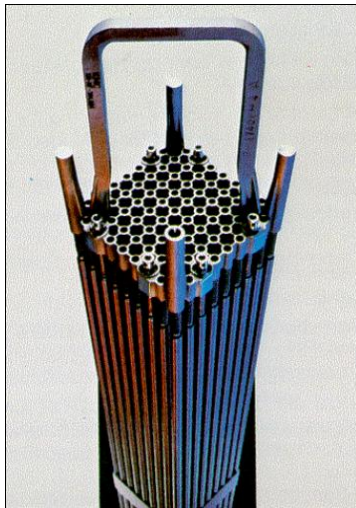
La Hague Reprocessing plants



History

- 1966 – 1976 : **UP2, 400t/y, UNGG Fuel**
- 1976 – 1994 : **UP2/HAO, 400 t/y LWR Fuel**
- 1990 - ..., : **UP3, 800 t/y, LWR Fuel**
- 1994 - ..., : **UP2/800, 800 t/y, LWR Fuel**

La Hague plants production (treated fuels)



(*) Excluding gas-graphite fuel
 (**) Annual consumption in France is close to 600TWh
 (***) Electricity generated with the treated metal fuel

UP2/HAO Feedback

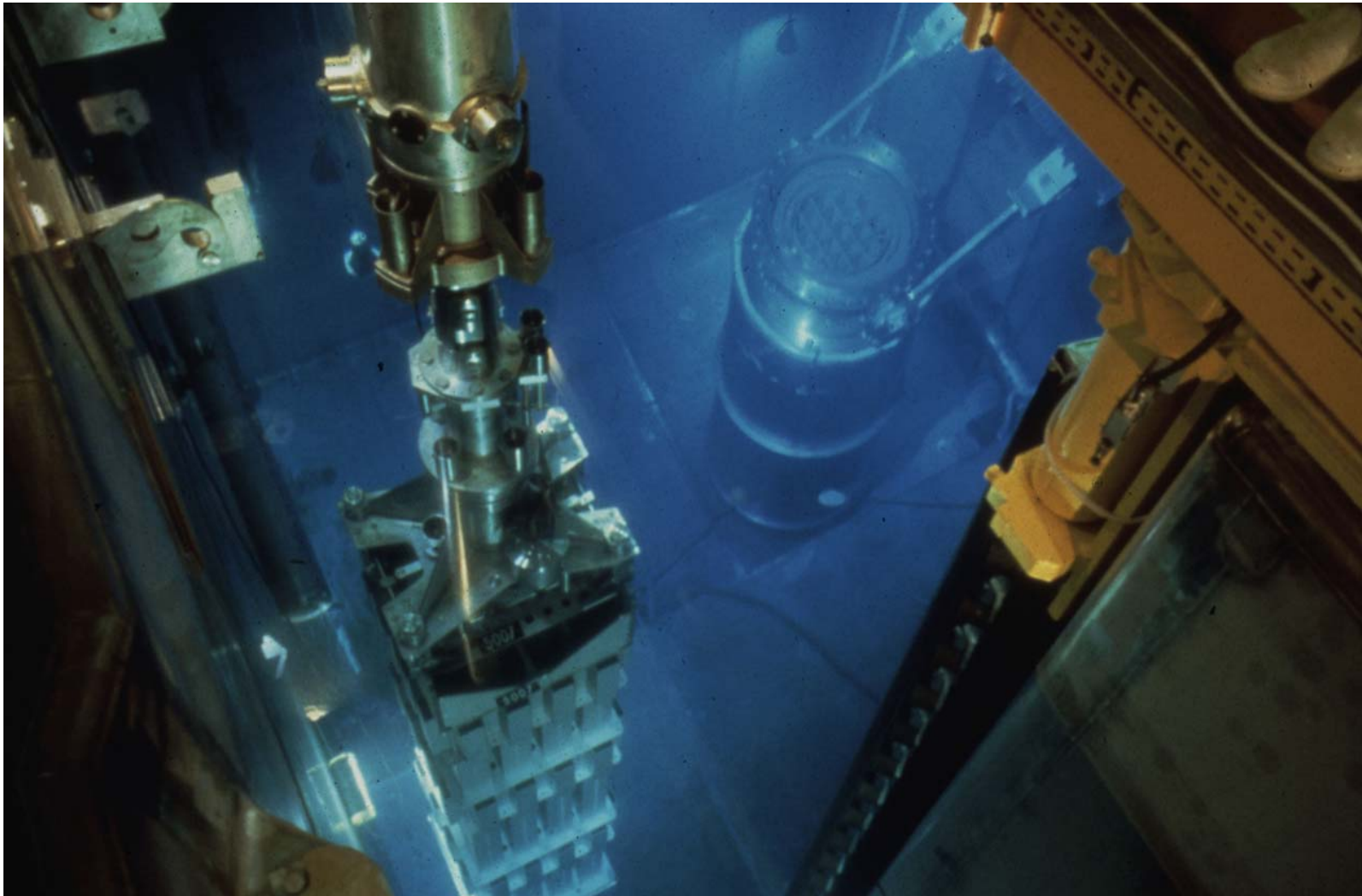


Many difficulties at the beginning of the operation

- **Bad estimation of hard-linked to the reprocessing of LWR fuel**
 - *Technological gap from the UNGG to the LWR spent fuel*
- **French decision to launch a major R & D plan for the UP3 process**
 - *From the head end process to the waste conditioning*
 - *Clean Plant*
 - *Continuous dissolution*
 - *The increasing of the Burn Up of the LWR fuel*
 - *The LWR MOX Spent Fuel Treatment*

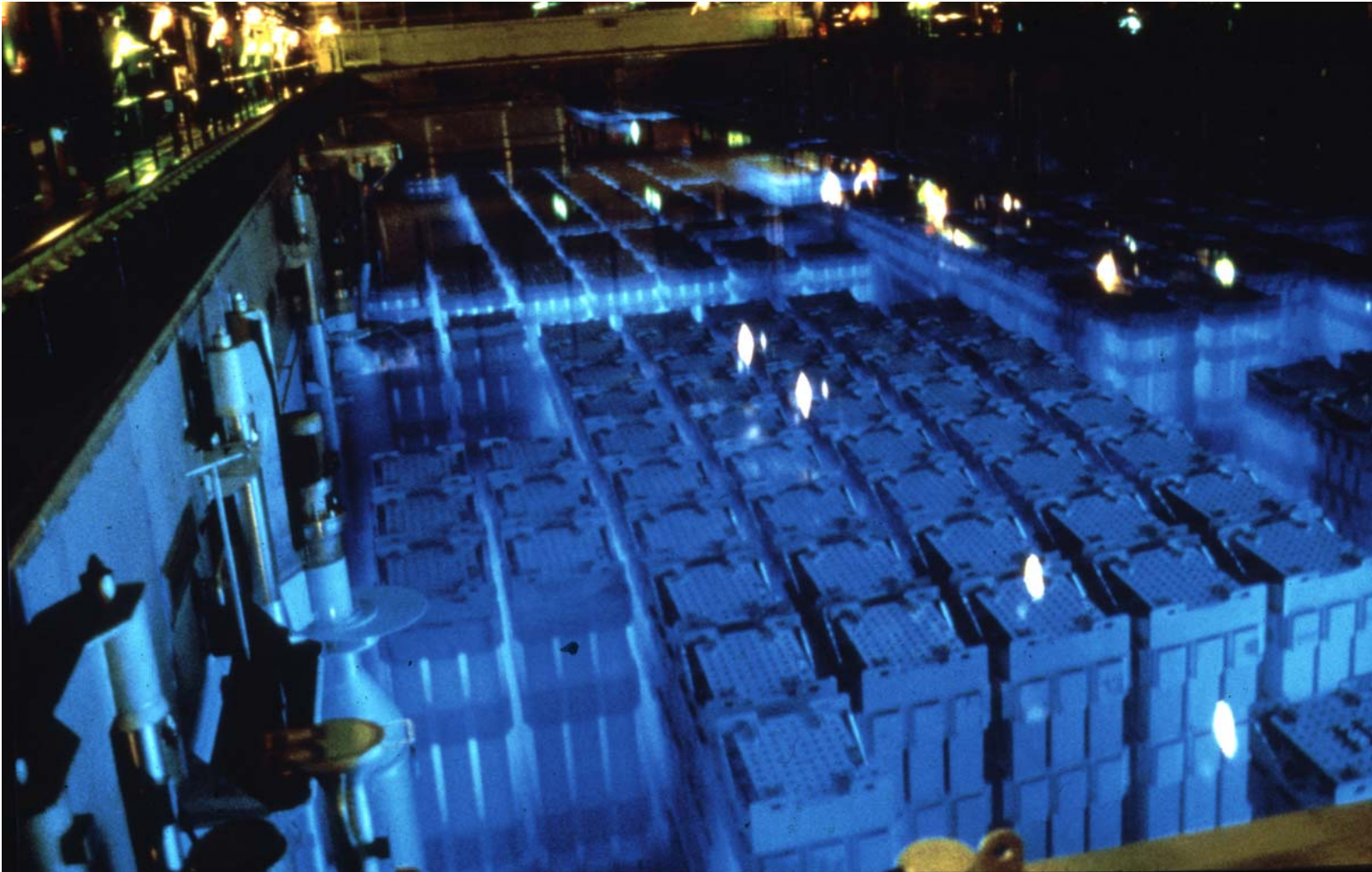
Closing the Fuel cycle ... an industrial reality

cea



Closing the Fuel cycle ... an industrial reality

cea



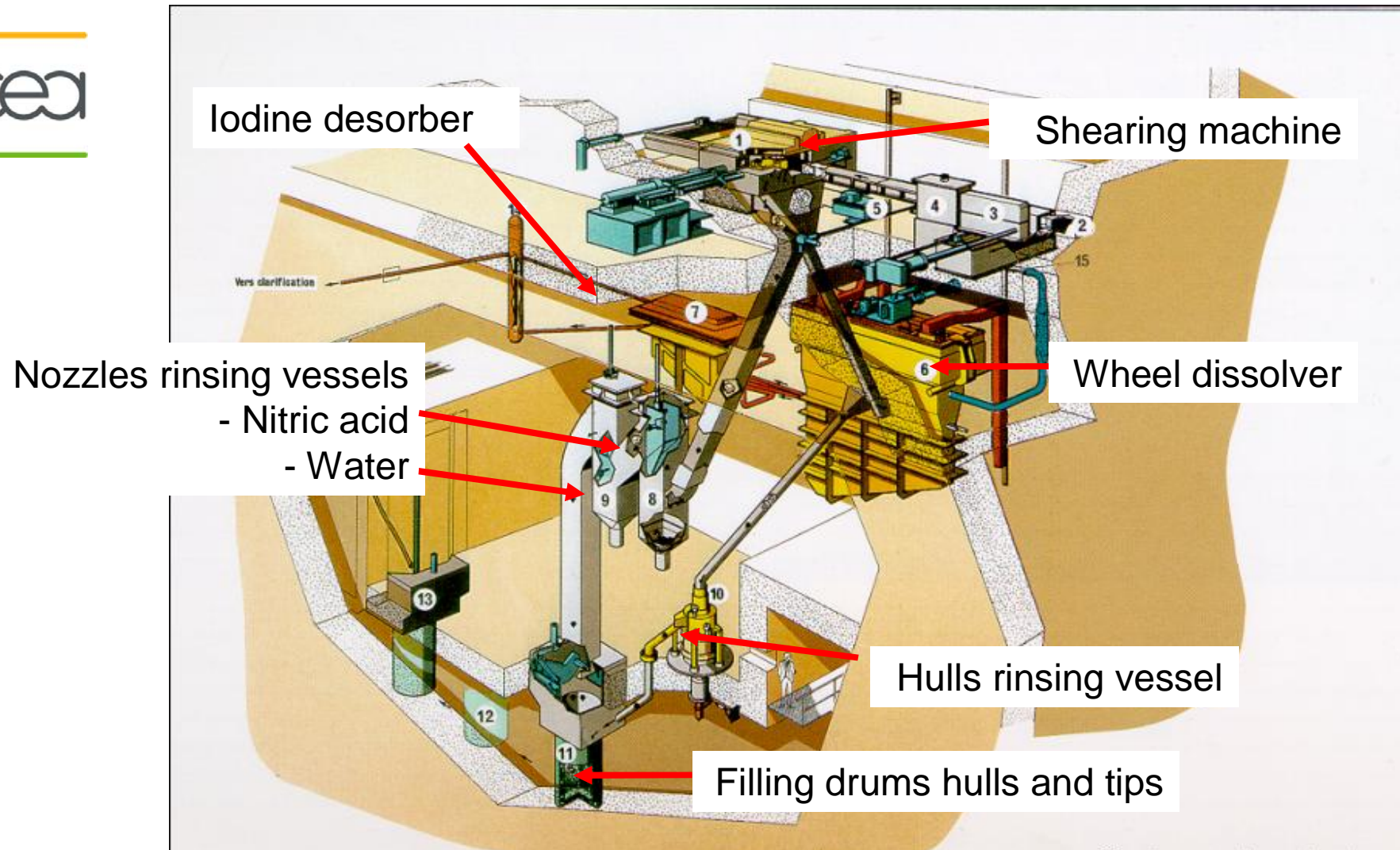
Exemple of La Hague cooling pool



- **Dimensions :**
 - L : 50 m
 - l : 16 m
 - P : 9 m(about 7200 m³ water)

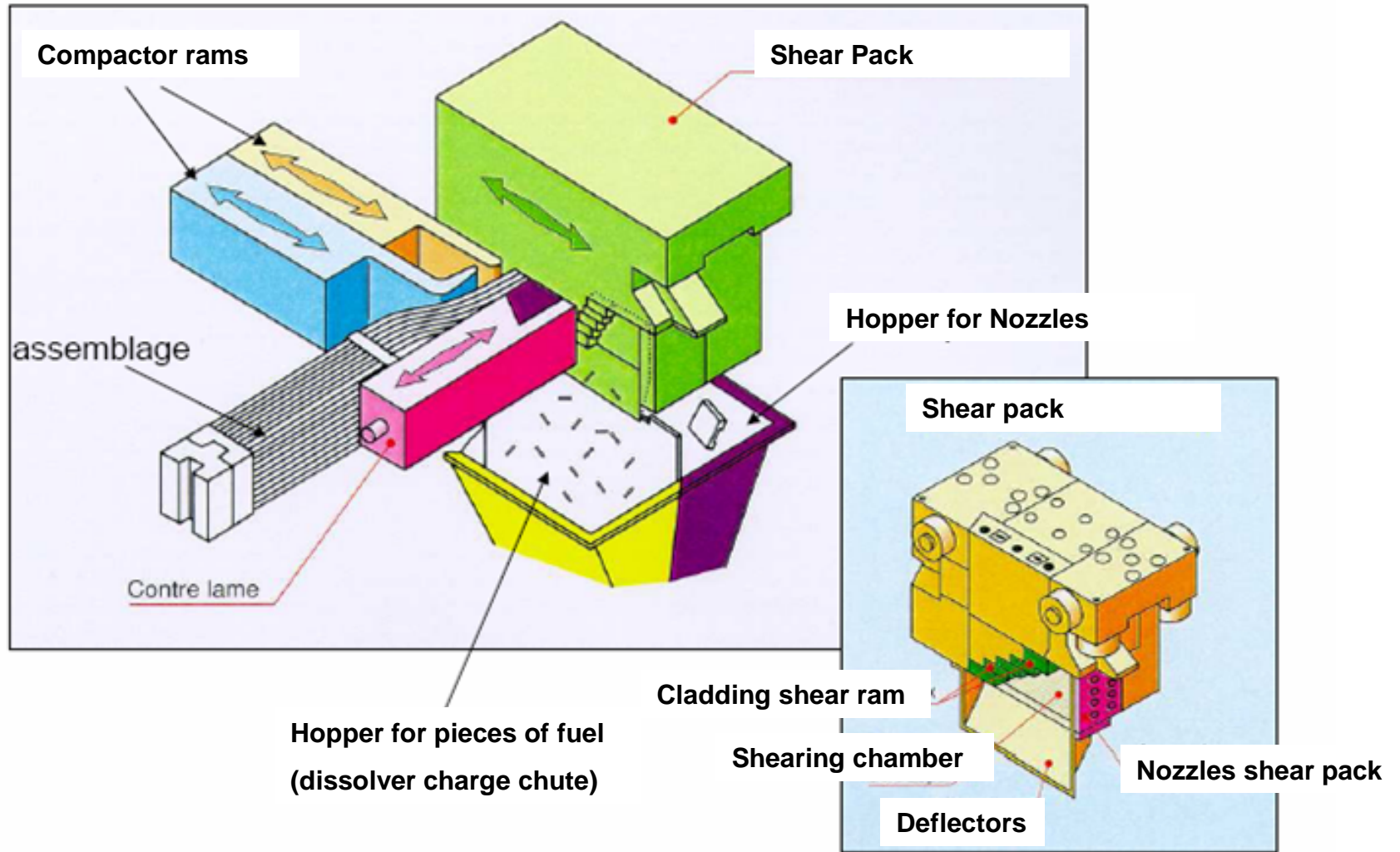
- **Storage capacity :**
 - 730 baskets, each bearing :
 - . 9 PWR assemblies
 - . or 16 BWR assemblies(~ 4000 t, ~ 200 reactor.years)

UP3 Dissolution Line Mock-Up



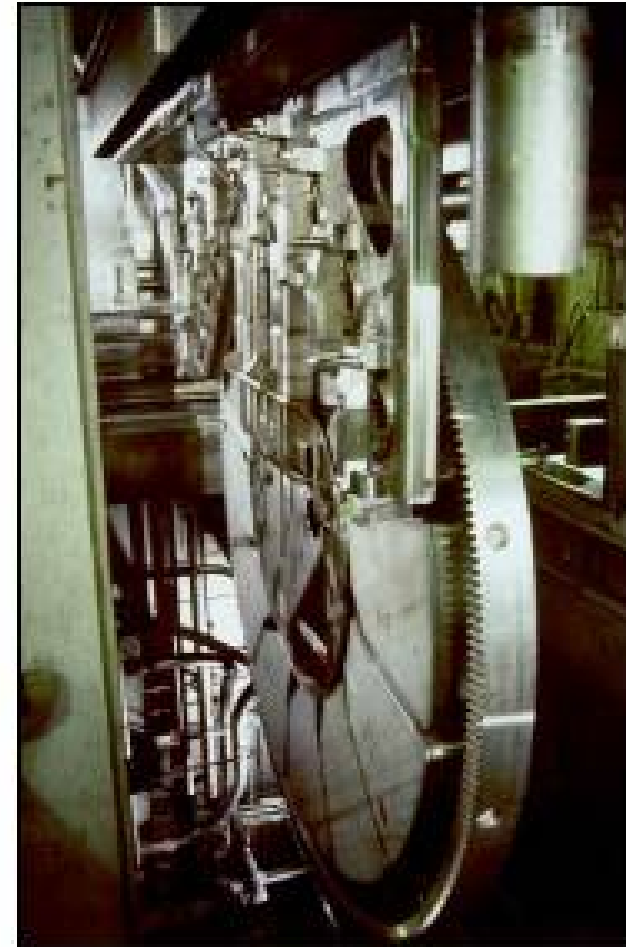
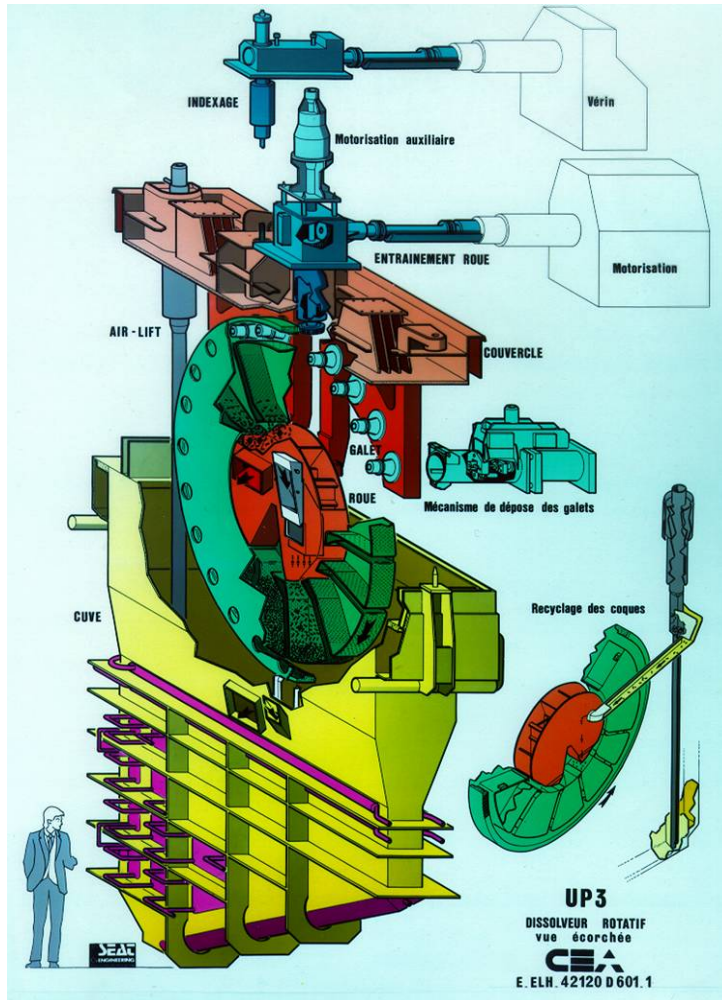
Mechanical shearing machine

Operating mock-up of UP3 shearing tools



Continuous Wheel Dissolver

UP3 Dissolver vessels

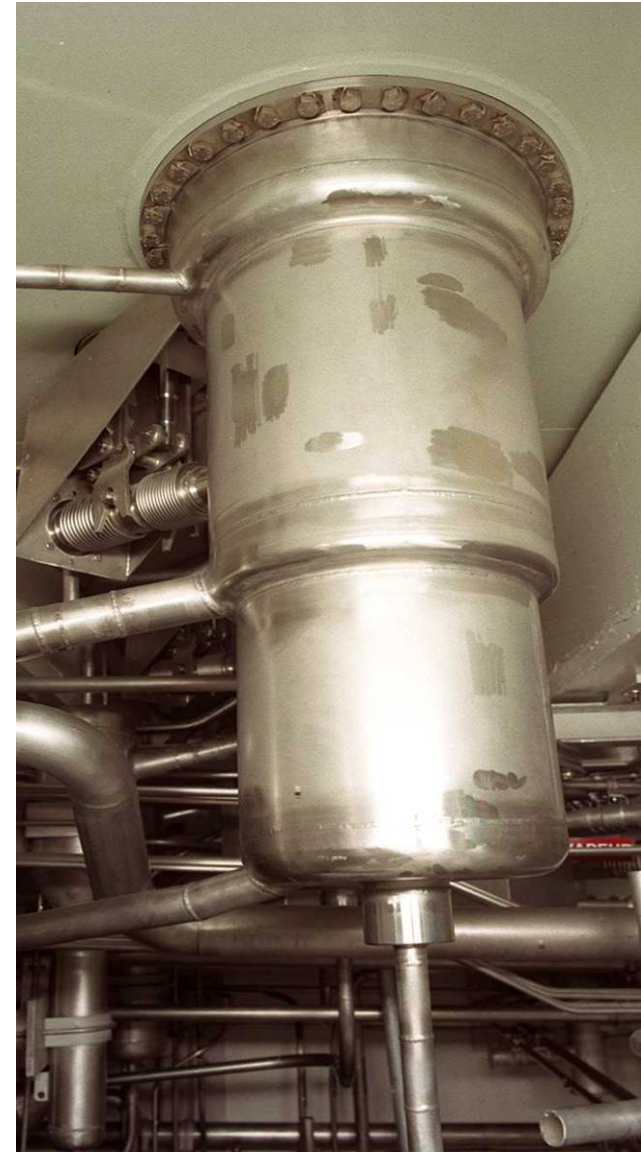


UP3 and UP2-800/R4 : Pulsed columns



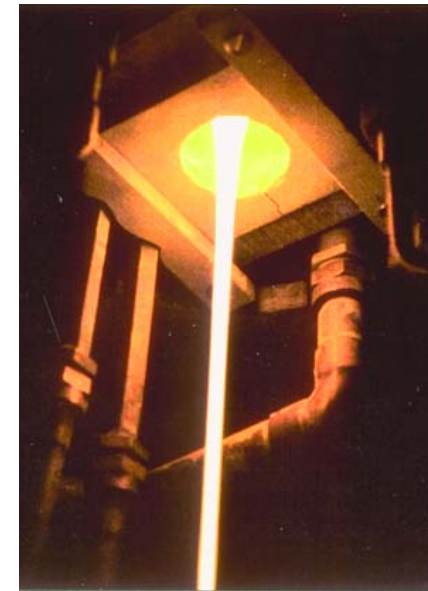
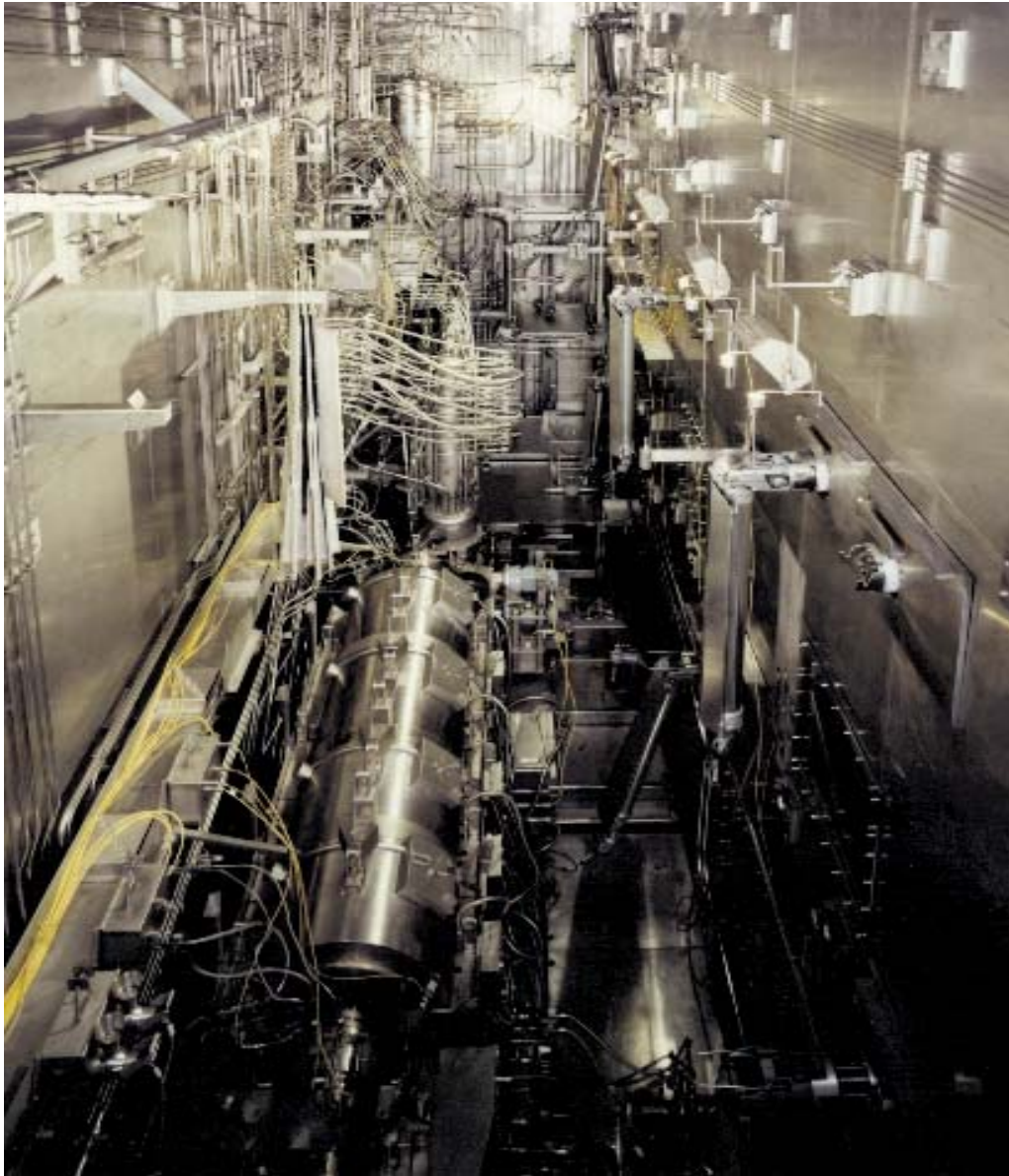
UP2-800/R4 : Centrifugal contactors

cea



La Hague Vitrification line

cea



Feedback of UP3 and UP2-800 operation



At the Beginning : UOX from 10 to 30 GWd/tHM

Today : UOX 45 GWd/tHM

Tomorrow : 60 GWd/tHM

And also : 72,6 t of LWR MOX irradiated from 30 to 53 MWd/HM in industrial conditions (Demonstration Campaigns)

Feedback after 20 years of UP3 and UP2-800 operation (more than **22 000 tons** of LWR fuels **reprocessed**)

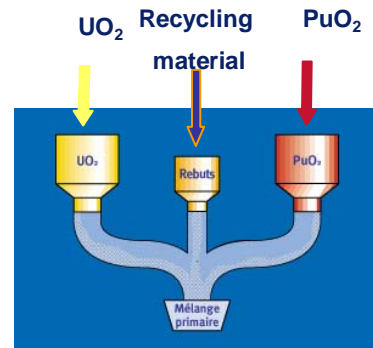
- Robust PUREX Process, large capacity, safety, ...***
- Flexible Plants***

Pu Recycling with MOX fuels : the MELOX plant

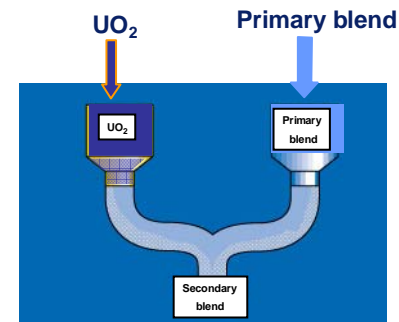
cea



Key advantages of the advanced MELOX high-throughput process



1 Preparation of primary blend



2 Preparation of secondary blend

- Powder blending is the **key to the MELOX process**.
- The MELOX process allows an on-line recycling of almost all scrap.
- More than 35 years of PWR and BWR operating experience have demonstrated the high quality of MOX fuel fabricated by the AREVA group.
- MOX fuel behavior in the reactor is similar to UO_2 fuel in normal and off-normal conditions.

The performance and reliability of the MELOX process are recognized worldwide

MELOX Fuel fabrication process



One MOX fuel assembly contains enough energy to supply a city of 100,000 with electricity for an entire year

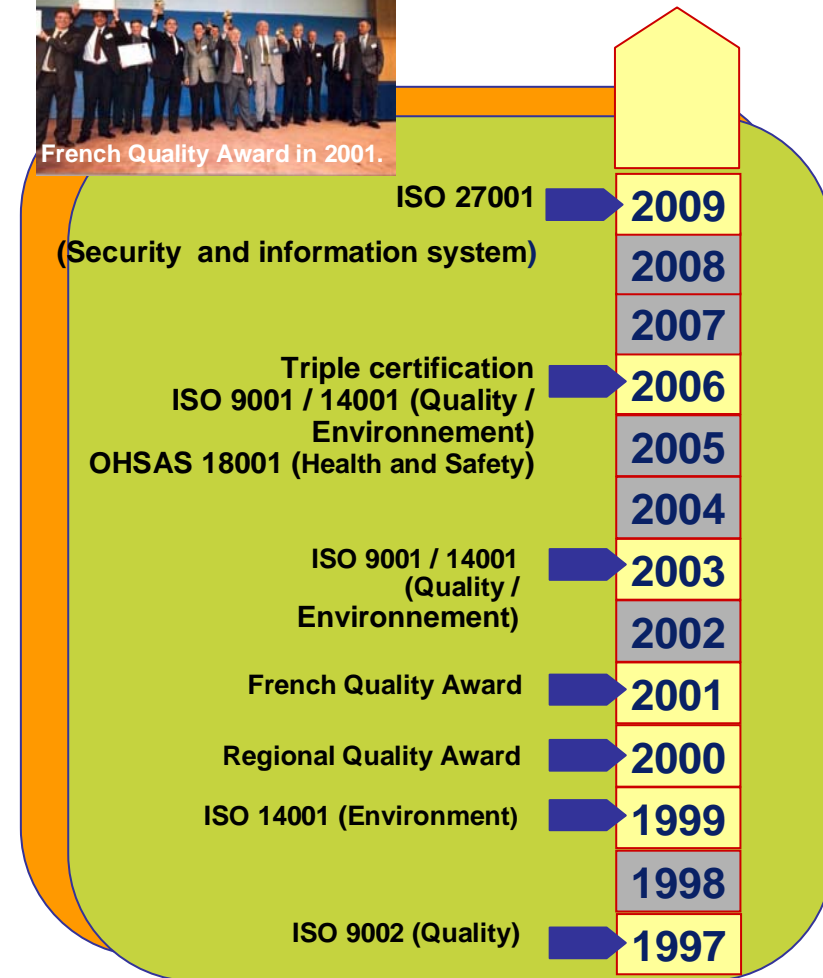
Total Quality Management at MELOX



- **Continuous improvement to achieve customer satisfaction**
- **Constant attention to successful environmental integration**



French Quality Award in 2001.



MELOX Plant safety



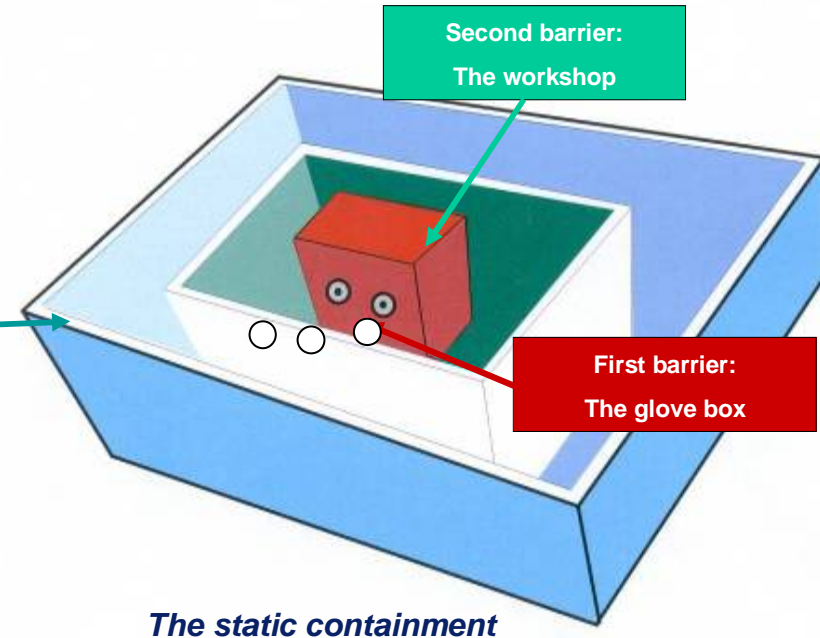
Prevention and control of nuclear materials dispersion and external exposure are based on:

- ◆ A confinement system using **three barriers and dynamic containment** (negative air pressure)

Third barrier:
The building

Second barrier:
The workshop

First barrier:
The glove box



- ◆ High level of plant automation

MELOX Plant design



▶ Major risk prevention has been integrated into facility design

- ▶ Fire
- ▶ Criticality
- ▶ Seismic measurements
- ▶ Thermal risks

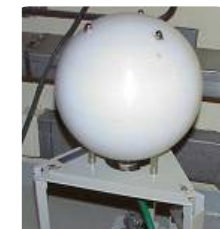
Seismic support



Thermocouple



CO₂ fire extinguisher



Neutron radiation detector



Fire door

MELOX: a continuous and effective system of safeguards

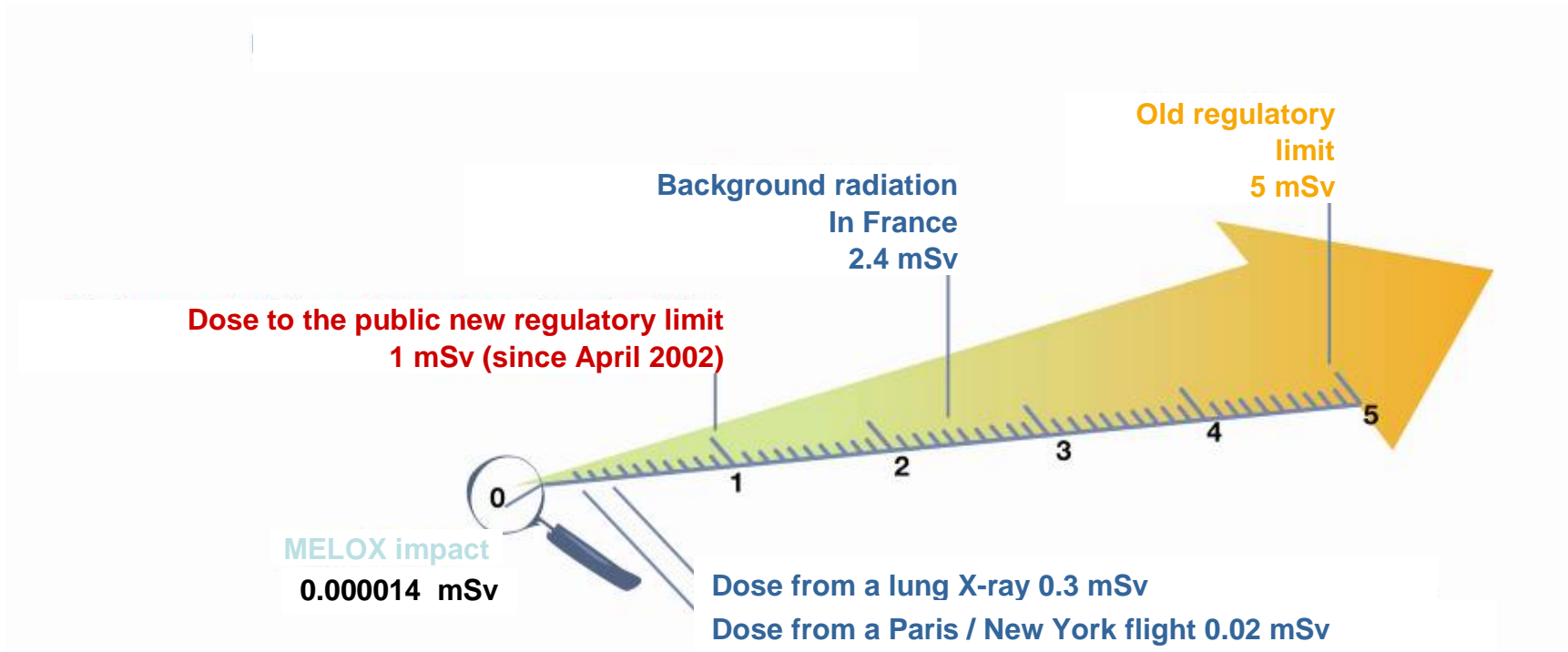


- **National and international organizations (Euratom, IAEA) monitor nuclear materials at AREVA MOX fabrication plants.**
- **Euratom worked with the French regulatory authorities and the plant operator during the MELOX design phase to develop the system of safeguards with the objective of “Continuous Inventory Verification”**
- **This system is specific to the plant characteristics:**
 - **Control of inputs/outputs**
 - **Independent, automatic measurements**
 - **Control of the annual inventory of nuclear materials**
 - **Sample analysis**
- **This system complies with IAEA requirements**

Radiological impact of the MELOX plant



- In year 2008, the **impact** of liquids and gaseous radioactive effluents of **MELOX plant** is about **0.000014 mSv**

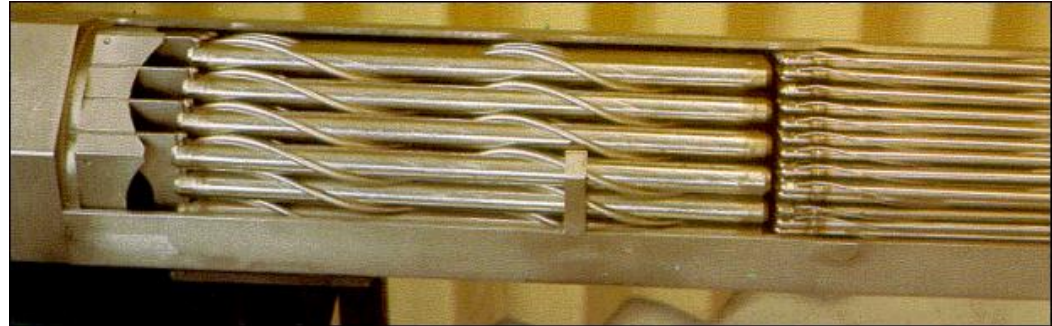
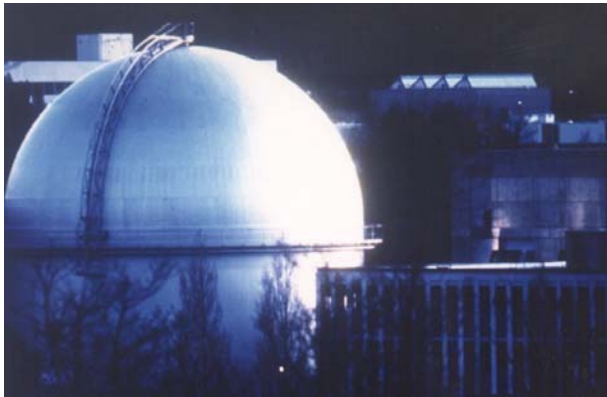


Maximum effluents impact allowed by the decree:
0.0017 mSv per year

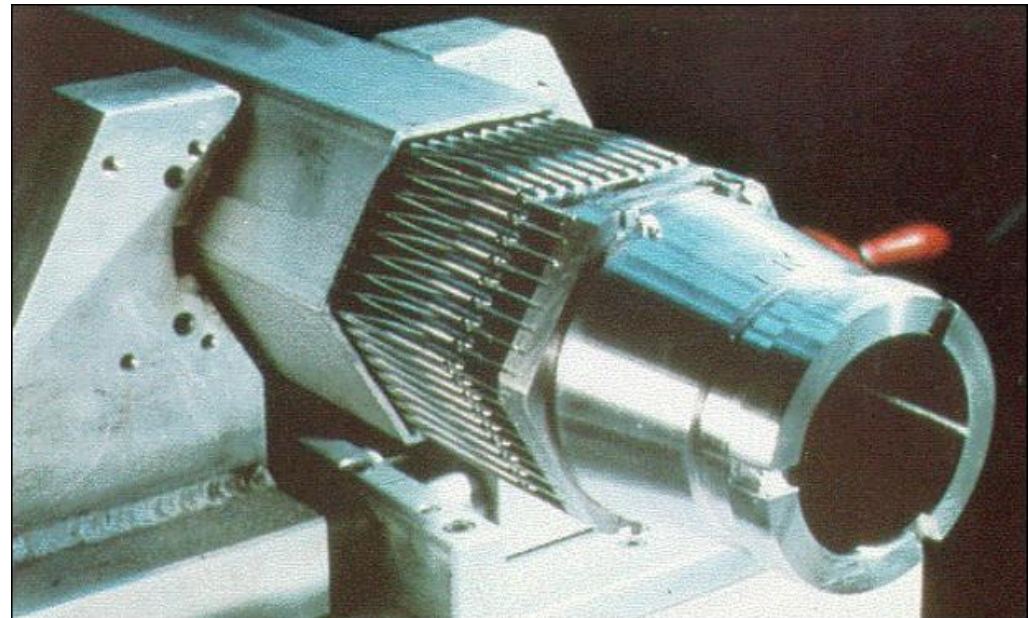
Reprocessing of SFR MOX FUEL : French experience



RAPSODIE Reactor
40 MWth
1967-1983

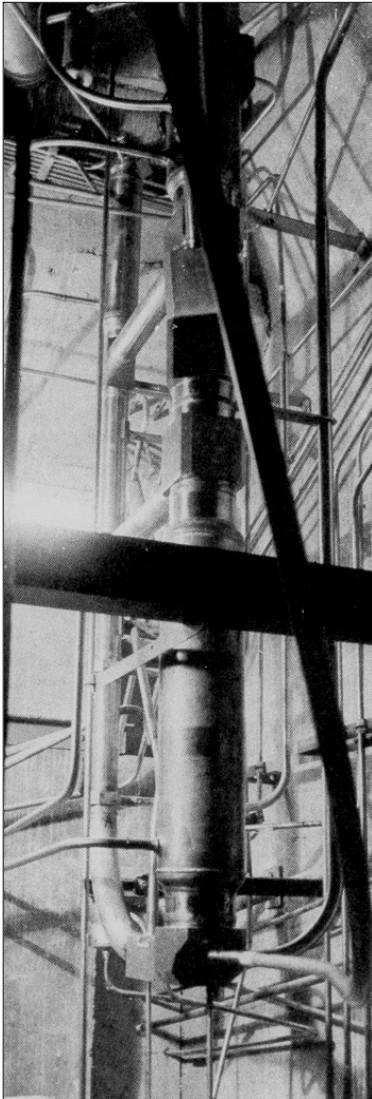


PHENIX Reactor
600 MWth / 250 MWe
1973-2009



Reprocessing of SFR MOX FUEL : French experience

cea



AT1 La Hague Facility (1969 – 1978)

First French facility for the reprocessing of SFR MOX FUEL (from Rapsodie and Phénix reactors)

:

- Capacity : **1 kg/d**
- Burn Up : **40 to 120 GWd/t** mixed oxide
- Cooling time : from **6 months to 2 years**
- ~1 t of MOX SFR spent fuel treated

Reprocessing of SFR MOX FUEL : French experience



UP2-400 La Hague Facility (1978 – 1984)

Reprocessing of ~11 t of PHENIX Fuel

- Burn Up : **20 to 90 GWd/t** mixed oxide
- Cooling time : from **16 months to 4 years**

Batch dissolution and extraction of U and Pu by dilution in UNGG fuel dissolution

Reprocessing of SFR MOX fuel : French experience



Marcoule Pilot Facility

- TOP Facility (1973 – 1983)

RAPSODIE, KNK and PHENIX Spent fuel

- ~ 8 t of MOX SFR Spent fuel
- Burn Up : **35 to 105 GWd/t** mixed oxide
- Cooling time : from **10 to 50 months**

- TOR Facility (1988 – 1997)

KNK and PHENIX Spent fuel

- ~ 7 t of MOX SFR Spent fuel
- Burn Up : **60 to 103 GWd/t** mixed oxide
- Cooling time : from **3 to 10 years**



What characterize the SFR MOX Fuel at the reprocessing

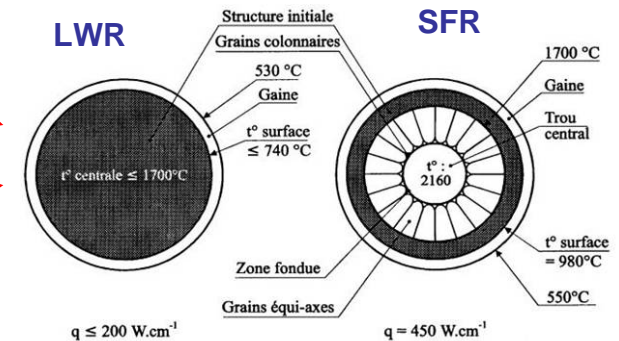


Fuel	Characteristics	LWR		MOX RNR
		UOX	MOX	
Composition before. irr.		UO ₂ enrichi	(U,Pu)O ₂	(U,Pu)O ₂
		²³⁵ U: 3 - 4,5%	Pu: 3 - 8,5%	Pu: 13 - 20%
Metallic Materials	Assembly	270 pins		200-300 pins
	Cladding	Zircaloy	Zircaloy	Stainless steel
	Others materials	Nozzles, grids, ...		SNP, nozzles, wrapper, spacewire, ...
	Ø pins (mm)	11 à 12	11 à 12	6 à 8
Irradiation	BU (MWd/t)	33000/60000	33000/45000	80000/120000
	Average temperature (°C)	850-1300°C)	850-1300°C	1700
Thermal power (KW)	Cooling 1 year	10	25	40
Composition after irr.	Plutonium	1	1 - 5	8 - 15
	Minors Act.	0,03 - 0,1	0,55	0,5
	FP	3,5	5	8 - 12
	Nobles metals	0,4	0,9	2 - 3

Solubility
Criticality

Corrosion Fe, Cr, ...

Technology



Pu-PF compounds

Glasses Incorporation

The french Act of June 2006, up to 2015 and 2020

⇒ **Key-points for waste management, and future R and D :**



- A national plan on radioactive materials and radioactive waste management (up-grading by Parliament every three years)
- A program on R & D with a time schedule to implement this plan
- A secured financing of radioactive waste management and R & D (dedicated fund)
- A step by step program of HLLL waste management, including the 3 **complementary** solutions :
 - Geological disposal for the final HL waste (dossier and Parliament decision in 2015, operation in 2025) ; **existing wastes will be disposed**
 - Intermediate storage for industrial flexibility
 - **MA Recycling :**
 - R & D in the framework of **Gen IV Systems**; international framework
 - interest (as consequences on the geological repository) to be proved with **a cost/benefit detailed analysis by 2012**
 - **2012 decision : demo by 2020** ; industrialization by 2040...