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The back-end of the Nuclear Fuel Cycle 1- The physics of transmutation for radioactive waste minimisation

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The back-end of the Nuclear Fuel Cycle
1 - The physics of transmutation for radioactive waste minimisation

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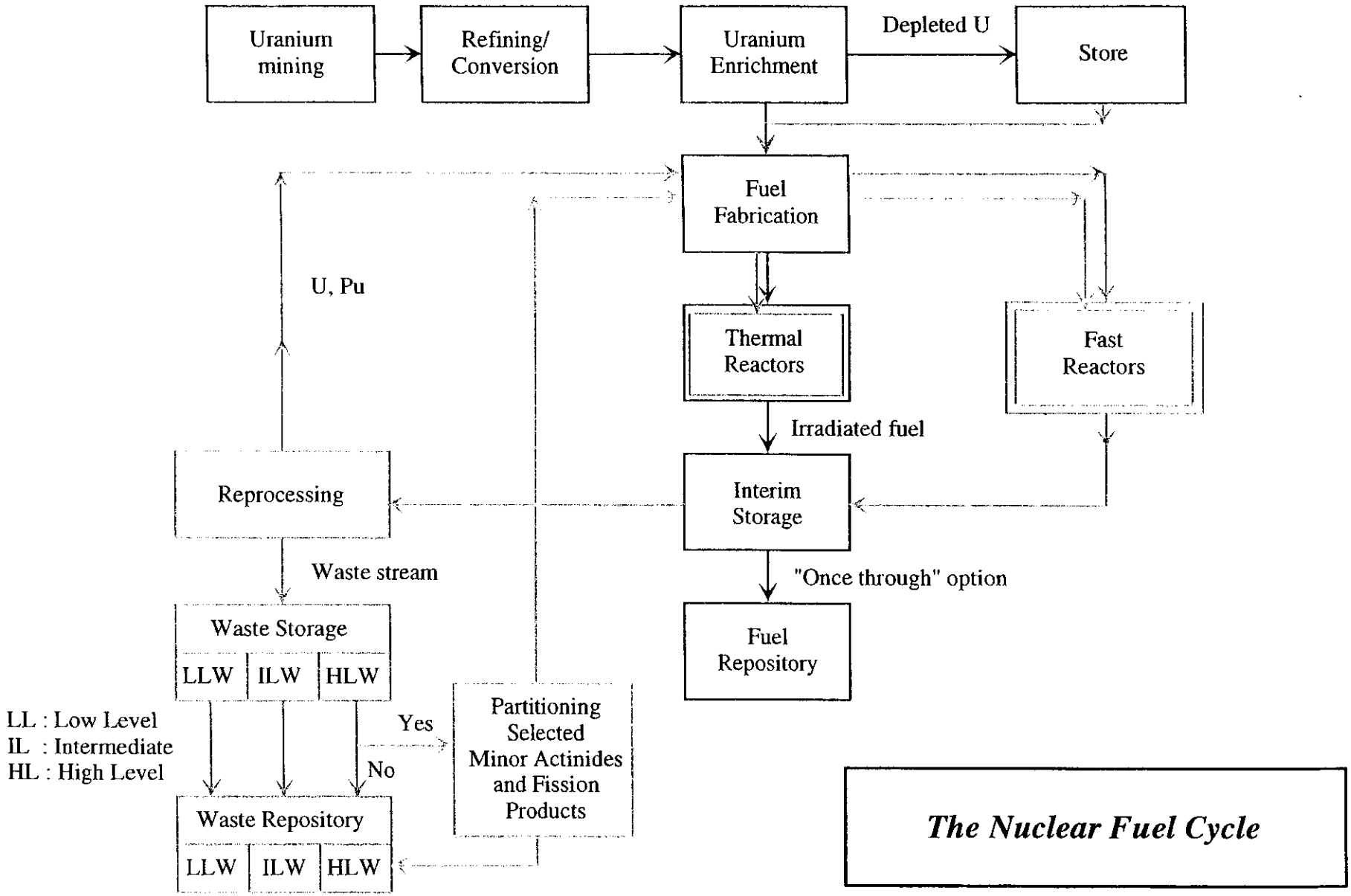
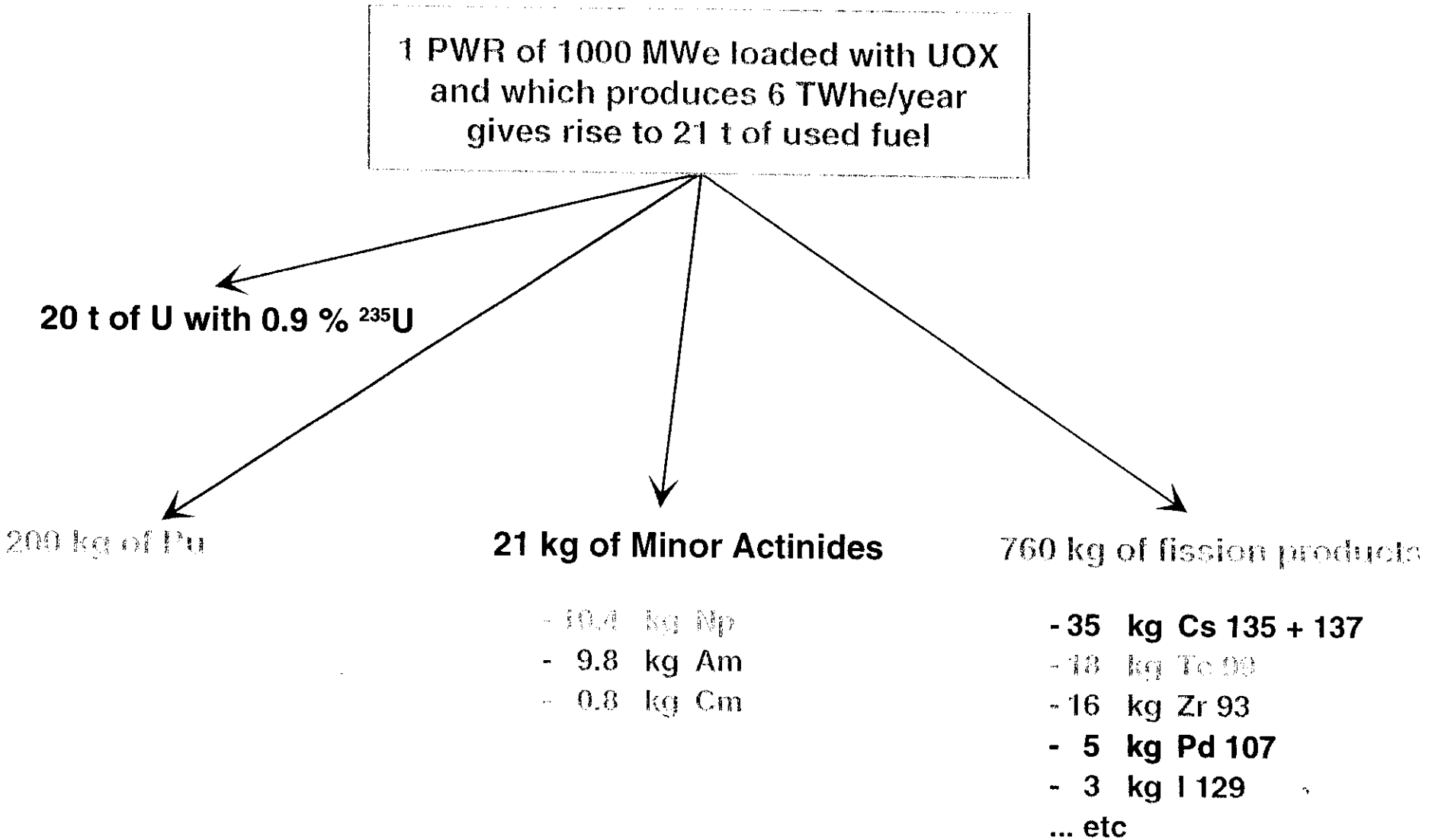
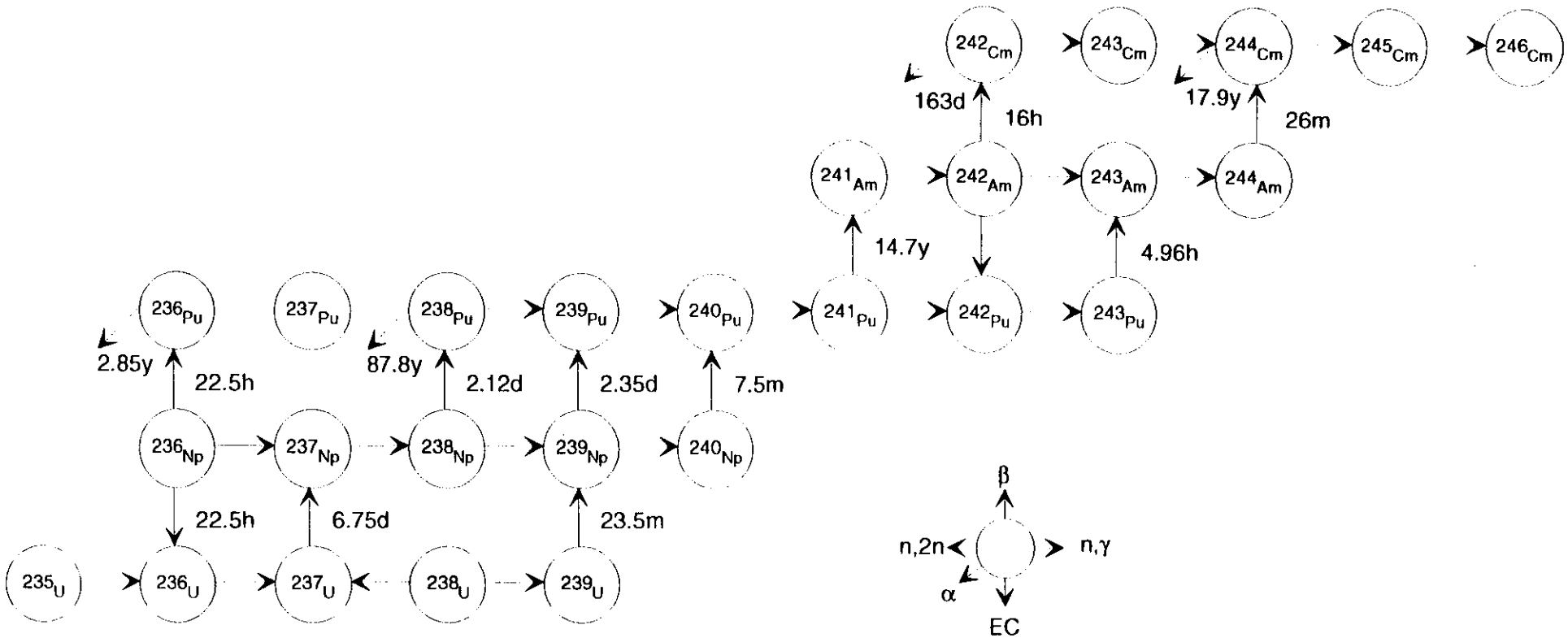

Uranium mining
Refining/ Conversion
Uranium Enrichment
Depleted U
Store
Fuel Fabrication
U, Pu
Thermal Reactors
Fast Reactors
Irradiated fuel
Reprocessing
Interim Storage
Waste stream
"Once through" option
Waste Storage
LLW
ILW
HLW
Fuel Repository
LL : Low Level
IL : Intermediate
HL : High Level
Yes
No
Partitioning Selected Minor Actinides and Fission Products
Waste Repository
LLW
ILW
HLW
The Nuclear Fuel Cycle


1 PWR of 1000 MWe loaded with UOX
and which produces 6 TWhe/year
gives rise to 21 t of used fuel
20 t of U with 0.9 % 235U
200 kg of Pu
21 kg of Minor Actinides
- 10.4 kg Np
- 9.8 kg Am
- 0.8 kg Cm
760 kg of fission products
- 35 kg Cs 135 + 137
- 18 kg Tc 99
- 16 kg Zr 93
- 5 kg Pd 107
- 3 kg I 129
... etc

$$\frac{dN_j}{dt} = \left(-\sigma_{aj}\,\Phi - \lambda_j\right) N_j + \sum_{k \neq j} \left(\sigma^i_{pk}\,\Phi + \lambda^i_k\right) N_k$$

$$\Phi = \Phi\left[\underline{N}(\vec{r}), \underline{\sigma}(\vec{r},E), \text{Power}\right]$$



ACTINIDE TRANSMUTATION CHAIN

The equation of nuclide balance in the equilibrium state is given by the following equation (Sekimoto et al.)

$$-\left(\lambda_i + \phi\sigma_{a,i} + r_i\right)n_i + \sum_j \lambda_{j\to i} n_j + \phi \sum_k \sigma_{k\to i} n_k + s_i = 0 \qquad (1)$$

where most notations are conventional. The coefficient r_i is the discharge constant of the ith nuclide. The term s_i is the supply rate of the ith nuclide from external source.

NATURAL DECAY OF ACTINIDES

ISOTOPES	EMISSION	HALF-LIFE	DAUGHTER ISOTOPES
Np 237	α	**2.1 x 10^6 years**	Pa 233 (27 d) U 233 (1.6 10^5 years) Th 229
Np 239	β^-	2.3 days	Pu 239
Am 241	α	432 years	**NP 237**
Am 242 g	(β^- (0.83)	16 hours	Cm 242
	(β^+ (0.17)		Pu 242
	isom. trans	152 years	Am 242 g
Am 243	α	**7380 years**	Np 239 → Pu 239
Cm 242	α	163 days	Pu 238
Cm 244	α	18 years	Pu 230
Cm 245	α	**8500 years**	Pu 241 → Am 241 → Np 237
Pu 236	α	2.8 years	U 232 (72 years), ... , Tl 208
Pu 238	α	87 years	U 234 (2.4 x 10^5 years), Th 230
Pu 239	α	24000 years	U 235
Pu 240	α	6540 years	U 236
Pu 241	β^-	14 years	Am 241
Pu 242	α	3.7 10^5 years	U 238

Isotope	Period	E (W/g)	neutrons/g/s		E_γ (keV)	γ ray	Dose coeff. (Sv/Bq)	
			FS	(α,n)			ICRP-30	ICRP-68
^{232}Th	1.40 10^{10} y	2.34 10^{-9}			1.2		7.40 10^{-7}	2.3 10^{-7}
^{231}Pa	3.28 10^{4} y	1.25 10^{-3}			38.7	27 keV (11%) 303 keV (2%) 300 keV (2%)	2.89 10^{-6}	7.1 10^{-7}
^{232}U ^{208}Tl	69.8 y 3.06 mn	0.620	1.05	1.85 10^{4}	1.7 3385.2	 2615 keV (100%) 583 keV (85%) 861 keV (13%)	3.44 10^{-7} —	2.9 10^{-7} —
^{233}U	1.59 10^{5} y	2.46 10^{-4}	4.19 10^{-6}		1.2		7.20 10^{-8}	5.0 10^{-8}
^{234}U	2.46 10^{5} y	1.57 10^{-4}	4.06 10^{-3}	3.44	1.5		7.20 10^{-8}	4.9 10^{-8}
^{235}U	7.04 10^{8} y	5.21 10^{-8}	6.51 10^{-4}	7.68 10^{-4}	167.8	186 keV (57%) 144 keV (11%)	6.80 10^{-8}	4.6 10^{-8}
^{236}U	2.34 10^{7} y	1.54 10^{-6}	4.04 10^{-3}	2.45 10^{-2}	1.2		6.70 10^{-8}	4.6 10^{-8}
^{238}U	4.47 10^{9} y	7.47 10^{-9}	1.89 10^{-2}	8.74 10^{-5}	1.3		6.70 10^{-8}	4.4 10^{-8}
^{237}Np ^{233}Pa	2.14 10^{6} y 27.0 d	1.80 10^{-5} 1.51 10^{-6}	≈ 10^{-4}	0.322	33.5 215.8	14 keV (56%) 86 keV (12%) 312 keV (39%) 98 keV (15%)	1.06 10^{-6} —	1.1 10^{-7} 8.8 10^{-10}
^{238}Pu	87.7 y	0.499	2.28 10^{3}	1.58 10^{4}	1.5		1.00 10^{-6}	2.3 10^{-7}
^{239}Pu	2.41 10^{4} y	1.70 10^{-3}	2.61 10^{-2}	46.4	0.7		1.16 10^{-6}	2.5 10^{-7}
^{240}Pu	6.56 10^{3} y	6.21 10^{-3}	931	170	1.4		1.16 10^{-6}	2.5 10^{-7}
^{241}Pu	14.4 y	2.85 10^{-3}					2.36 10^{-8}	4.7 10^{-9}
^{242}Pu	3.74 10^{5} y	1.03 10^{-4}	1.63 10^{3}	2.58	1.3		1.16 10^{-6}	2.4 10^{-7}
^{241}Am	433 y	0.100	0.796	3.17 10^{3}	28.2	60 keV (36%) 14 keV (36%)	1.20 10^{-6}	2.0 10^{-7}
^{242m}Am	141 y	1.67 10^{-3}			5.0		1.14 10^{-6}	1.9 10^{-7}
^{243}Am ^{239}Np	7.36 10^{3} y 2.36 d	5.66 10^{-3} 4.65 10^{-4}		177	57.0 182.2	75 keV (66%) 15 keV (51%) 106 keV (27%) 104 keV (25%) 100 keV (16%) 278 keV (14%) 61 keV (13%) 228 keV (11%)	1.19 10^{-6} —	2.0 10^{-7} 8.0 10^{-10}
^{242}Cm	163 d	107	1.72 10^{7}	4.18 10^{6}	1.4		3.54 10^{-8}	1.3 10^{-8}
^{243}Cm	30.0 y	1.56		6.09 10^{4}	133.2	104 keV (24%) 228 keV (11%) 278 keV (14%) 100 keV (15%)	7.86 10^{-7}	2.0 10^{-7}
^{244}Cm	18.1 y	2.50	1.01 10^{7}	8.84 10^{4}	1.3		6.00 10^{-7}	1.6 10^{-7}
^{245}Cm	8.50 10^{3} y	4.98 10^{-3}			93.8	104 keV (30%) 100 keV (18%) 175 keV (10%) 117 keV (7%)	1.20 10^{-6}	3.0 10^{-7}
^{246}Cm	4.73 10^{3} y	8.90 10^{-3}	≈ 7 10^{6}	<< FS	3.0		1.19 10^{-6}	2.9 10^{-7}
^{247}Cm	1.60 10^{7} y	2.53 10^{-6}			302.8	403 keV (69%) 278 keV (3%)	1.11 10^{-6}	2.7 10^{-7}
^{248}Cm	3.40 10^{5} y	4.66 10^{-4}	≈ 3 10^{7}	<< FS	579.1	579 keV (100%)	4.40 10^{-6}	1.1 10^{-6}

Physical parameters of some isotopes

Waste radiotoxicity :

- Two notions :

  - ♦ Source of potential radiotoxicity :

    - ■ it does not consider storage barriers,
    - ■ abnormal evolution of storage packages / sites.
      ⇒ Minor actinides dominate.

  - ♦ Residual radiotoxicity :
    What goes back to the biosphere in scenarios of normal evolution of storage packages / sites, after solution and migration in the geological environment.
    ⇒ Some long-lived fission products dominate

RADIOTOXICITY

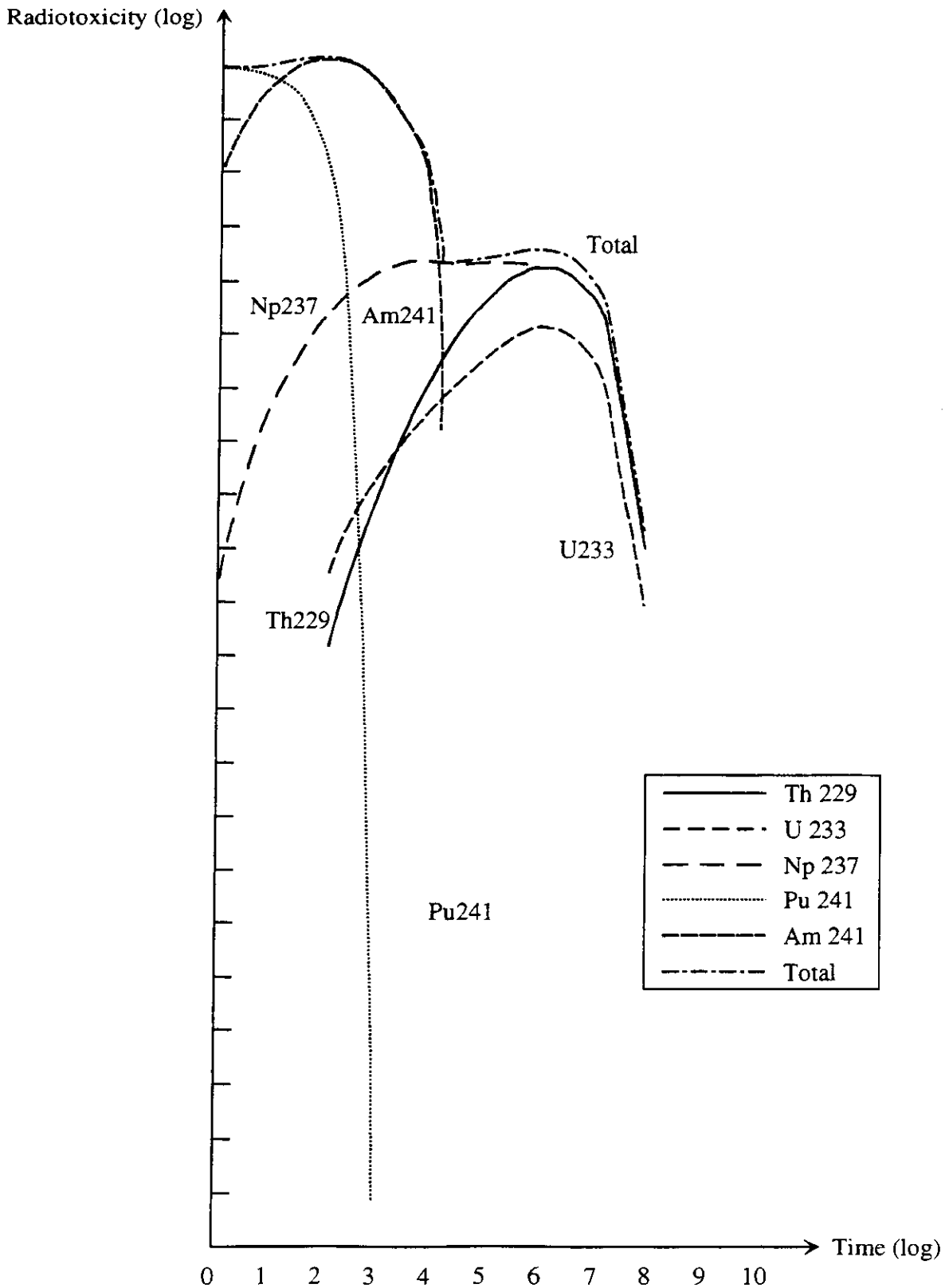
To each isotope it is associated an "ingestion danger" coefficient : (in Sv/Bq).

For a unit mass of a given isotope, the radiotoxicity R(t) at an instant t is given by :

$$R(t) = a.\sum_i \frac{Q_i(t)\, D_i}{Q_o(o)\, MT_i} \qquad (Sv/\ g)$$

- where "a" is a constant (= 1.322 x 106) ;
- the sum is over the "father" isotope and its progenies ;
- $Q_O(o)$ initial number of "father" nuclei ;
- $Q_i(t)$ number of nuclei of isotope i at t ;
- M molar mass of "father" isotope ;
- D_i "ingestion danger" coefficient (Sv/Bq) of isotope i.

RADIOTOXICITY (INGESTION)
DUE TO 1 g of Pu-241



MAJOR PROBLEMS for PARTITIONING/ TRANSMUTATION STRATEGIES

- Spent fuel radioactive nuclides inventory as a starting point.

  Example : PWR / with UOX fuel burn-up 33 GWd/t.

- Radiotoxicity Source term and its evolution with time :

  - The source characterizes in a simple way the potential toxicity of wastes, and also some accidental scenarios like intrusion in a repository.

  - On the contrary in scenarios of normal evolution of a repository, the concentrations at the outlet are dependent on the source term, but also on solubility, migration etc of nuclides, and then on the geological environment (residual toxicity).

	10^3 y	10^4 y	10^5 y	10^6 y
TOTAL (URT + Pu + Np + Cm + Fp) Sv/TWhe	$3.1\ 10^8$	$7.7\ 10^7$	$4.2\ 10^6$	$5.2\ 10^5$
Components (%)				
URT	/	/	6	28
Pu	90	97	88	50
Np	/	/	1.3	13
Am	9.2	2.5	2.7	6.8
Cm	0.3	0.4	/	/
Long Lived Fission Products	$6.0\ 10^{-4}$	$2.4\ 10^{-3}$	$3.2\ 10^{-2}$	$9.6\ 10^{-2}$
URANIUM COMPONENTS (Sv/TWhe)				
Depleted U	$2.4\ 10^4$	$3.5\ 10^4$	$1.4\ 10^5$	$5.7\ 10^5$
U from reprocessing (URT)	$2.1\ 10^4$	$4.8\ 10^4$	$2.2\ 10^5$	$1.4\ 10^5$
Mill tailings	$7.2\ 10^5$	$6.6\ 10^5$	$2.6\ 10^5$	$6.5\ 10^1$

Components of the radiotoxicity source term
Evolution with time (Sv/TWhe)
(PWR fuel irradiated at 33 GWd/t)

MAIN LONG-LIVED FISSION PRODUCTS CONTRIBUTION (%) TO FP RADIOTOXICITY SOURCE TERM

	10^3 y	10^4 y	10^5 y
Se 79 T = 6.5 10^4 y	6	5	3
Zr 93 T = 1.5 10^6 y	12	12	14
Tc 99 T = 2.1 10^5 y	36	37	36
Sn 126 T = 1.0 10^5 y	24	24	17
I 129 T = 1.6 10^7 y	16	16	22
Cs 135 T = 2.3 10^6 y	6	6	8

- As far as the source term of potential radiotoxicity, it is dominated by actinides

- As far as residual toxicity, solubility and migration properties, variable in different geological environments, would underline the contribution of some long-lived fission products : e.g. I-129, Cs-135 or Tc-99.

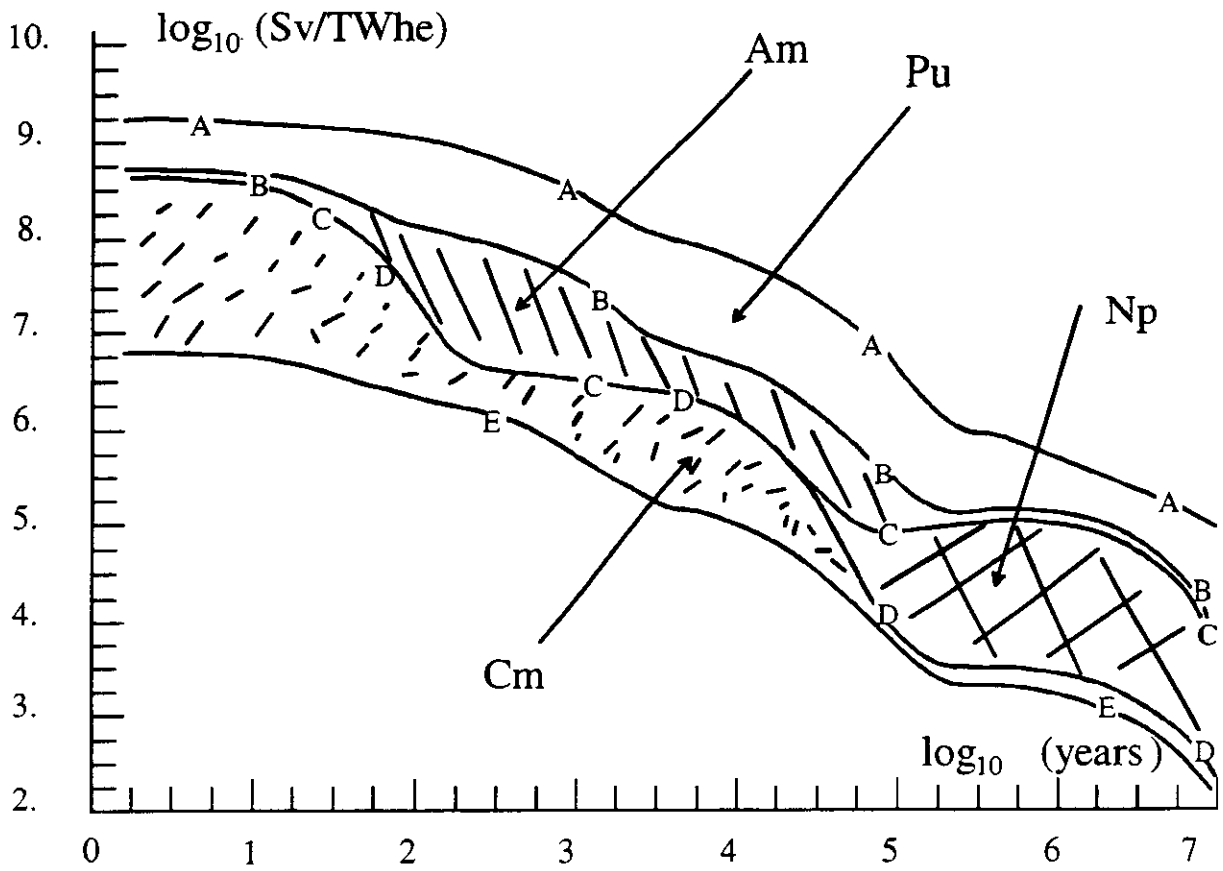
As far as the source of potential radiotoxicity :

- The plutonium contribution is dominant.

If the Plutonium contribution is set equal to the losses in reprocessing ($\rightarrow$ 0.1 %), then :

- Am contribution is dominant between ~ 10^2 y and ~ $10^4 \div 10^5$ y.
- Np becomes relevant only from ~ 10^5 y.
- Cm (which is important for $t \leq 10^2$ y) is significant, by means of daughter isotopes, at $t \approx 10^4$ y.

Potential radiotoxicity to wastes



A : open cycle
B : 0.1 % Pu ; 100 % AM to wastes
C : 0.1 % Pu ; 1 % Am ; 100 % Np, Cm to wastes
D : 0.1 % Pu ; 1 % Am, Np ; 100 % Cm to wastes
E : 0.1 % Pu ; 1 % Am, Np, Cm to wastes

Radiotoxicity reduction : Theoretical cases
(i.e. one does not say what is done with the materials after partitioning)

THE PLUTONIUM ISSUE

« Transmutation » of minor actinides is meaningful only if the Pu issue has been clearly defined.

Also, the impact of Pu utilisation on the fuel cycle has to be assessed, and this has much to do with MA production and possible needs to « transmute » them.

Mass flow to storage, in kg/TWhe, for a nuclear park (400 TWhe, 60 GWe) (Loss at reprocessing : 0.1 % for Pu)

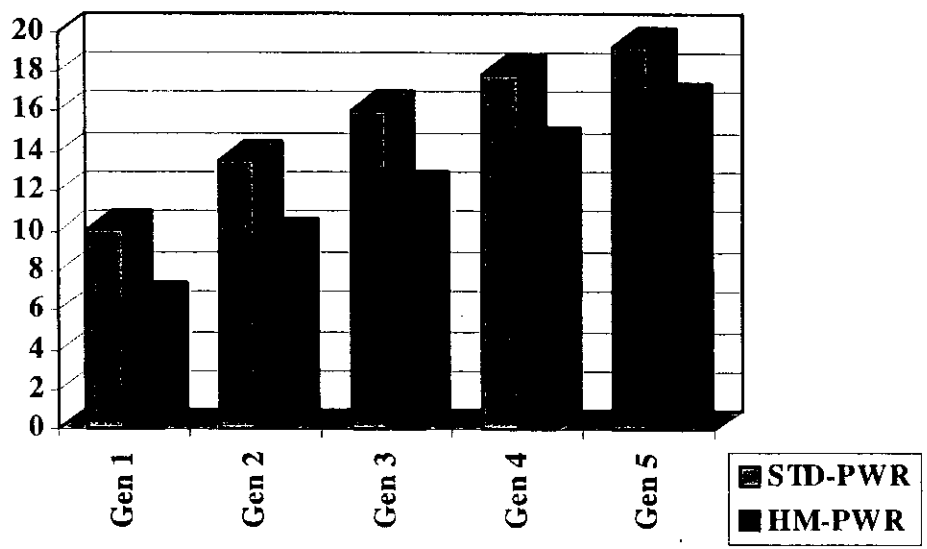
Fraction of reactors in the park / Element	100 % PWR UOX (open cycle)	100 % Fast Reactors (FR)	32 % FR + PWR (UOX)	18 % FR + PWR (UOX & MOX)	100 % PWR MOX with enriched U support
Pu	28.1	0.16	0.09	0.08	0.05
Np	1.7	0.50	1.2	1.3	1.5
Am	1.3	3.2	4.3	4.6	4.5
Cm	0.26	0.34	0.75	1.0	2.25

⇒ In principle, the flexibility of Fast Neutron Reactors offers a wide range of options in order to manage resources for a sustainable future.

⇒ PWRs and BWRs, offer the possibility to foresee the use of Plutonium in relatively standard reactors over most of the next century.

⇒ The Pu vector degradation with a multiple recycling can be such that a theoretical limit for total Pu content is reached, the limit being a positive value for the moderator void reactivity coefficient.

Evolution of Total Pu Content with Recycle Generation (w/o)



However, several physics approaches have been proposed to allow an extension of Pu multirecycling in PWRs :

This is the case of the « MIX » (Pu with a ^{235}U enriched support) concept.

High-moderator-to-fuel ratio PWR lattices (HM-PWR) offer another way to increase the Pu multirecycling potential of PWRs.

However, the increase of MA production has to be accounted for :

MA production (tons/year) in a 60 GWe power park at equilibrium

	UOX-PWR in open cycle	MOX-PWR	HM-PWR	MIX-PWR
MA (total)	1.5	4.1	3.6	3.3
Np	0.8	0.7	0.7	0.6
Am	0.6	3.0	2.4	1.8
Cm	0.1	0.5	0.5	0.9

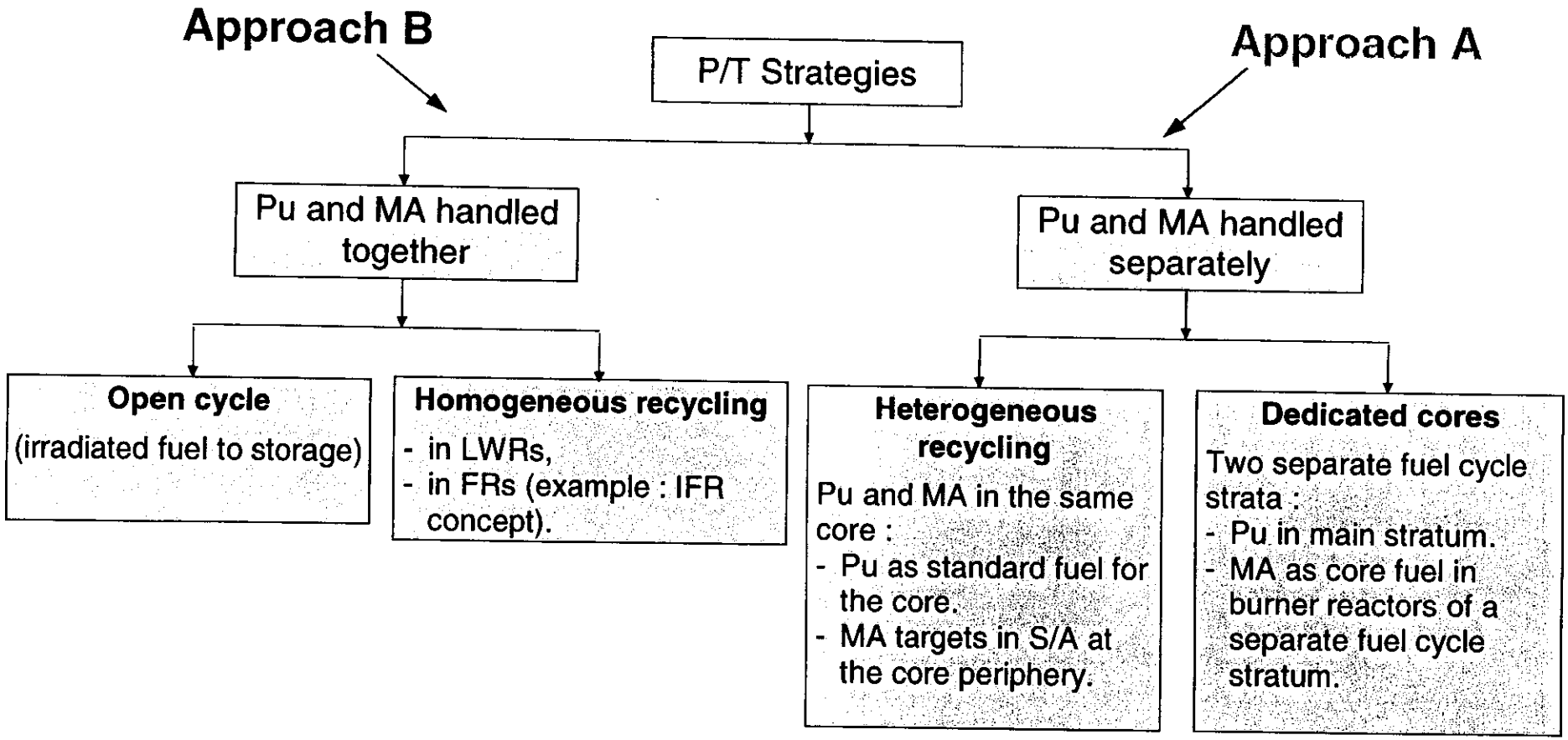
* For MA transmutation :
  - Homogeneous recycling (e.g. as in the IFR concept).
  - Heterogeneous recycling (e.g. Am + Cm targets in special S/A at the periphery of a fast reactor core).

  However :
  - Special interest in the « double strata » approach and the comparison of critical/subcritical cores.
  - Dedicated cores (MA-based fuels).

* For LLFP transmutation :
  - Hierarchy (I-129, Tc-99, Cs-135, Se-79 etc.) to be established.
  - Mostly : transmutation by « LSD » (Leakage with Slowing Down) method - Applicable to critical/subcritical systems.

* Role of pyrochemistry being revisited in most countries.

Pu and MA management in P/T strategies



Remark : If LLFP management is required, they can in principle be handled in the different scenarios as targets to be irradiated at the periphery of the different core types.

Transmutation in a fission reactor

Physics aspects to be accounted for :

→ Destruction rate by fission (which accounts for fission of isotope i and of isotopes i + 1, i + 2 ... obtained by successive captures).

→ Neutron balance : is there a sufficient number of neutrons available for transmutation ?

→ There is build-up of higher mass isotopes : what are the consequences on the fuel cycle ?

→ If one introduces isotopes to be transmuted, are there consequences on the reactor performances ?

Average cross-sections

$$\bar{\sigma} = \int \sigma(E)\,\phi(E)\,dE \Big/ \int \phi(E)\,dE \quad (b)$$

Isotope	ϕ : Thermal n reactor (PWR)			ϕ : Fast n reactor (FR)		
	σ_f	σ_c	α	σ_f	σ_c	α
Np-237	0.52	33	63	0.32	1.7	5.3
Np-238	134	13.6	0.1	3.6	0.2	0.05
Pu-238	2.4	27.7	12	1.1	0.58	0.53
Pu-239	102	58.7	0.58	1.86	0.56	0.3
Pu-240	0.53	210.2	396.6	0.36	0.57	1.6
Pu-241	102.2	40.9	0.40	2.49	0.47	0.19
Pu-242	0.44	28.8	65.5	0.24	0.44	1.8
Am-241	1.1	110	100	0.27	2.0	7.4
Am-242	159	301	1.9	3.2	0.6	0.19
Am-242m	595	137	0.23	3.3	0.6	0.18
Am-243	0.44	49	111	0.21	1.8	8.6
Cm-242	1.14	4.5	3.9	0.58	1.0	1.7
Cm-243	88	14	0.16	7.2	1.0	0.14
Cm-244	1.0	16	16	0.42	0.6	1.4
Cm-245	116	17	0.15	5.1	0.9	0.18
U-235	38.8	8.7	0.22	1.98	0.57	0.29
U-238	0.103	0.86	8.3	0.04	0.30	7.5

Qualitative indications on the capture-to-fission ratios are favourable for multiplying systems with a fast neutron spectrum.

However a more quantitative approach is needed to understand the transmutation mechanisms (for each isotope) :

- impact of other isotopes build-up during the transmutation of a specific radioactive isotope into stable isotope(s),

- competition among nuclear reactions and radioactive decay,

- impact of the multiplying system type (critical or subcritical).

To investigate the potential of a multiplying (critical or subcritical) system to transmute isotope J (actinide or long-lived fission product), one can use the notion of neutron consumption D_J of isotope J at equilibrium :

D_J = number of neutrons (normalized to one fission) necessary to transform isotope J (an all the products of successive reactions starting from J) into stable isotopes.

D_J can be evaluated with a simple algorithm.

→ $D_J < 0$ means "neutron production"

→ $D_J > 0$ means "neutron consumption"

The algorithm for the calculation of neutron consumption of initial nucleus J, producing incineration and all kinds of decays in the J-th family : $J \rightarrow J1 \rightarrow J2 \rightarrow \ldots \rightarrow JM$

(M : number of generations inside this family) :

$$D_J = \sum_{J1_i} P_{J \rightarrow J1_i} \left(R_{J1_i} + \sum_{J2_k} P_{J1_i \rightarrow J2_k} \left(R_{J2_k} + \ldots \right) \right)$$

where :

$P_{JN_r \rightarrow (JN+1)_s}$: probability of the $JN_r \rightarrow (JN+1)_S$ transmutation, normalized to the total rate of all possible JN_r process.

R_M : neutron loss of surplus as a result of the appearance of M.

If the initial FUEL consists of a set S of different J-nuclei :

$$D_{FUEL} = \sum_J \varepsilon_J D_J \text{ (sum over the S set)}$$

ε_J : fraction of the J-family in the fission product.

LWR $\phi = 10^{14}$ n/cm^2 sec

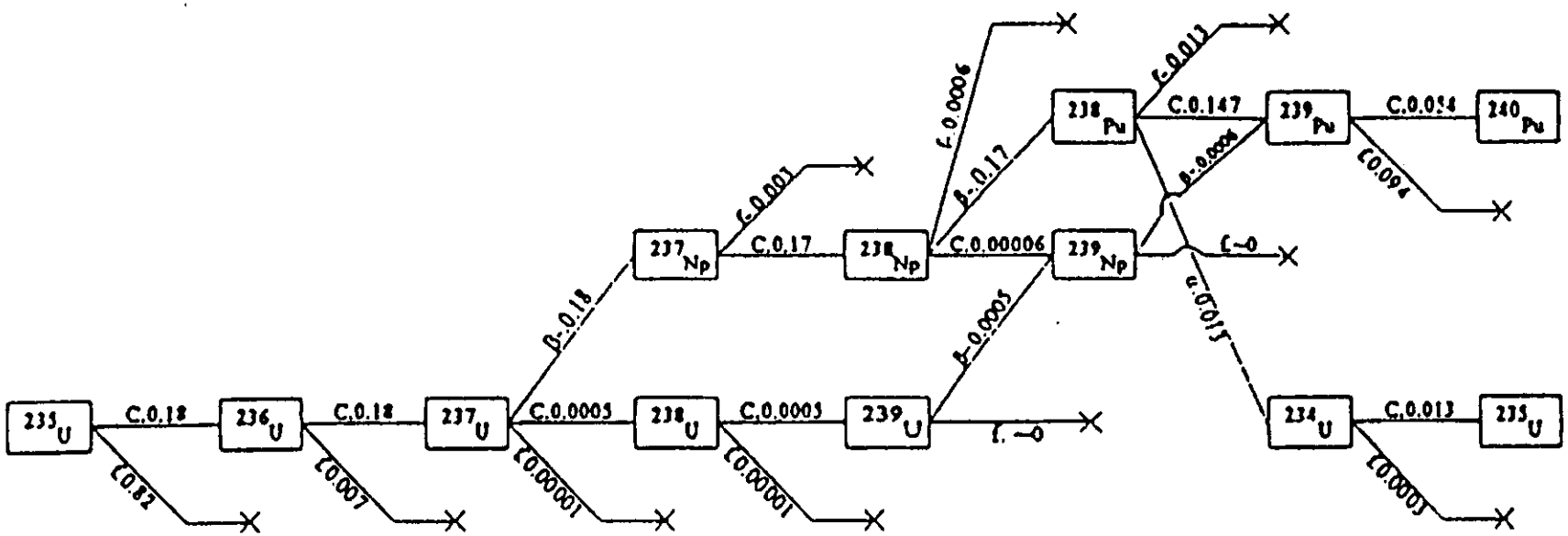
^{235}U

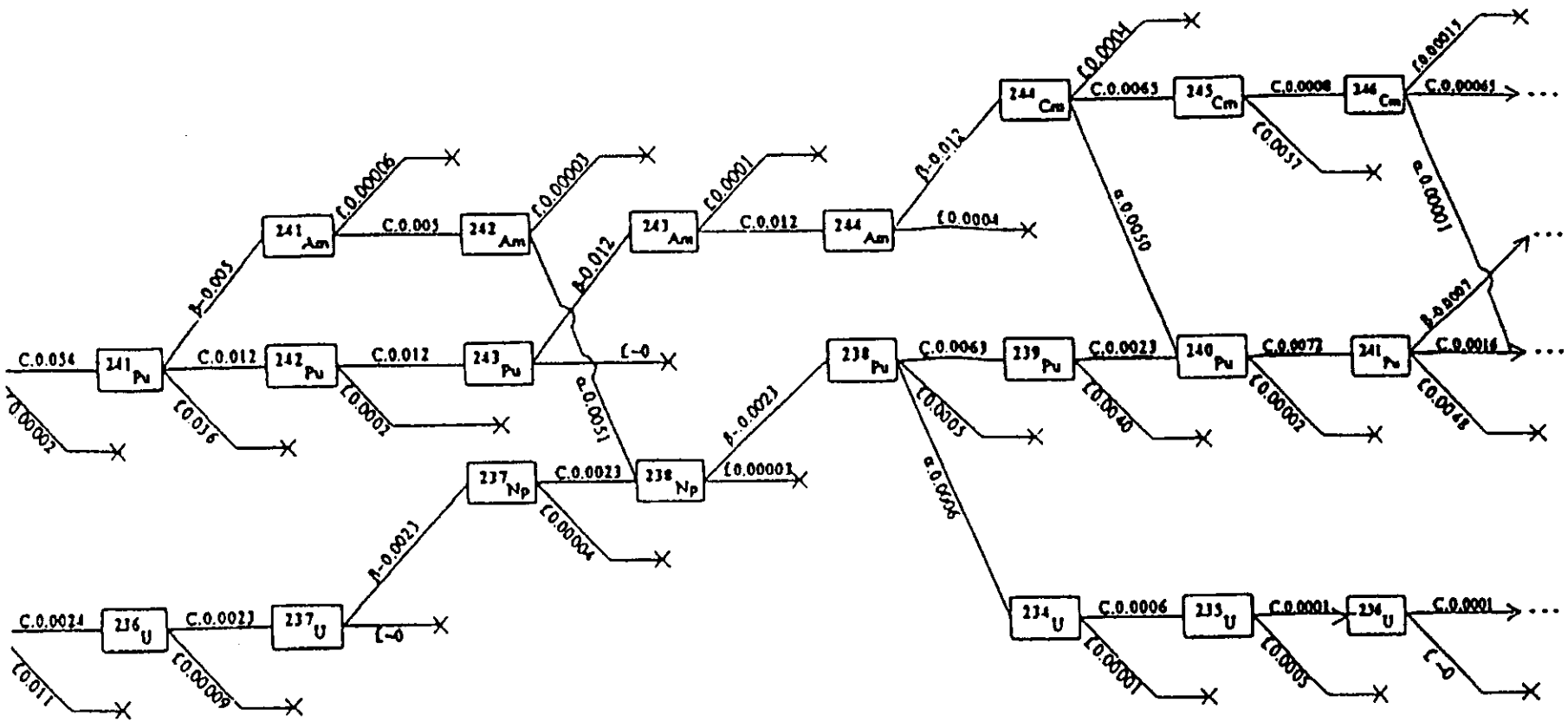
(z o.p - means : z-process has the probability equals to o.p)

z = f : fission

z = c : capture

z = β^+, β^-, α : decays




241 Pu
242 Pu
243 Pu
241 Am
242 Am
243 Am
244 Am
244 Cm
245 Cm
246 Cm
238 Pu
239 Pu
240 Pu
241 Pu
237 Np
238 Np
236 U
237 U
234 U
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C.0.054
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C.0.0008
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C.0.0006
C.0.0001
C.0.0001
β-0.005
β-0.012
β-0.012
β-0.0023
β-0.0023
α-0.0051
α-0.0050
α-0.0006
β-0.0007
C.0.0016
C.0.00002
C.0.036
C.0.0003
C.0.00006
C.0.00003
C.0.0001
C.0.0004
C.0.0004
C.0.0051
C.0.00001
C.0.0013
C.0.00065
C.0.0005
C.0.0040
C.0.00002
C.0.0048
C.0.00004
C.0.00001
C.0.0005
C.0.011
C.0.00009

THE PHYSICS OF TRANSMUTATION

"Transmutation" means excess neutron availability in the reactor core neutron balance.

The eventual "excess" G (expressed in neutrons/fission) is given by :

$$G = -\sum \varepsilon_J D_J - (CM + L) \qquad \text{(critical system)}$$

$$G = S_{ext} - \sum \varepsilon_J D_J - (CM - L) \qquad \text{(external source driven system)}$$

where : CM + L are neutrons (per fission) lost in parasitic captures or leakage
D_J is the neutron consumption (per fission) to transmute isotope J (down to fission products)
ε_J is the fraction of isotope J in the fuel of the system

Since CM + L is generally equal to ≈ 0.3 n/fission (for most systems) one can assess what system can allow "transmutation".

A positive G value can be obtained with fast reactors and practically any fuel. A positive G can be obtained with thermal neutron spectra only in very few cases, unless one is, e.g. increasing the enrichment (case of a PWR).

These simple physics considerations indicate that one can envisage the transmutation of any actinide (Am-241, Am-243, Np-237, etc ...) in a fast neutron spectrum. The effect of the flux level is less important (with a few exceptions, e.g. Np-237).

However, other types of spectra can be envisaged, but there is a « price » to pay. For example, in a PWR spectrum, one has to increase the enrichment in U-235, to get extra neutrons, to allow transmutation.

APPLICATION OF THE GENERIC PHYSICS FEATURES OF TRANSMUTATION

In general, the physics analysis shows that :

1. Transmutation rate of fissionable nuclides (MA, Pu, etc.) does not depend neither on neutron flux level, nor on neutron spectrum, but is direct function of fission rate (or power). It means, that if one fixes the power, all transmuter types (fast, thermal, superthermal with or without elevated flux) have the same transmutation rate.
   It comes from the fact that, for **ACTINIDES**, **TRANSMUTATION** means **FISSION**.
   Fast spectrum gives the best condition for transmutation due to an excellent neutron balance (i.e. neutron availability).

2. Long-Lived Fission Products LLFP-transmutation demands a significant neutron surplus because "**LLFP-TRANSMUTATION** means **CAPTURE**" and the rate of LLFP-transmutation is proportional to neutron surplus production rate.
   Again, fast spectrum produces many more "extra" neutrons/fission (which are available for LLFP).

CONCEPTUAL SCHEMES FOR TRANSMUTATION IN REACTORS

- Homogeneous/heterogeneous modes :
  - homogeneous : nuclei to be transmuted are mixed to the fuel
  - heterogeneous : nuclei to be transmuted are put in special subassemblies

- Once-through/multirecycling
  - only one irradiation (transmutation $\rightarrow$ 100 %)
  - multiple irradiations (transmutation << 100 %)

- « Dispersal »/ « Dedicated »
  - « dispersal » : nuclei « dispersed » in a large fraction of power park
  - « dedicated » : nuclei « concentrated » in few specially designed reactors, to minimize their fraction in the power park.

Both LWRs and FRs allow the **homogeneous** recycling of Np and Am. General features about core performances are as follows :

- The reactivity loss over the cycle is reduced (which is a positive feature).
- The temperature coefficients (and boron effectiveness) become worse (from a safety point of view)
- In particular, coolant void reactivity effects both for LWRs and FRs, become less negative (or more positive).
- For LWRs, there is the need of over-enrichment (which is due to the tight neutron economy).
- The maximum allowable fraction of MA in the fuel is 5 ÷ 10 % of total heavy isotopes for FRs, and 1 ÷ 2 % for LWRs.

As far as the **heterogeneous** recycling, an interesting option in particular for Am is represented by targets irradiated at the periphery of the core, to have a minimum perturbation to power distributions and reactivity coefficients.

Mass inventories after 1 recycle, for different types of recycling modes, both in fast reactors (FR) and PWRs

	FR of EFR type (1500 MWe) Homogeneous recycling (MA content : 2.5 %)	FR of EFR type Heterogeneous recycling in radial blankets (MA content : 40 %)	PWR-MOX with $V_m/V_F = 3$ (content : 1 %) BU=47 GWd/t	PWR-UOX Heterogeneous recycling in the core BU=42 GWd/t
Np				
Consumption (Kg/TWhe)	10	13	11	15
Consumption rate (%)	60	60	45	38
Fission rate (%)	27	24	9	3
Am				
Consumption (Kg/TWhe)	9	14	10	8
Consumption rate (%)	45	60	42	70
Fission rate (%)	18	22	6	13

Consequences on physics parameters of the fuel cycle

Type of recycling		HOMOGENEOUS			HETERO-GENEOUS
Actinide content in the fuel		2.5 % Np	2.5 % Am	2.5 % Cm	40 % Am + MgO
FABRICATION	Power	× 1	+ 71 %	× 24	× 14
	Activity	× 1	+ 3 %	× 2	× 19
	Gamma source	× 1	× 4	× 12	× 55
	γ-dose at 1 m	× 4	× 76	× 470	× 1680
	Neutron source	× 1	+ 40 %	× 1700	× 7
End of irradiation	Decay heat	+ 2 %	+ 1 %	- 4 %	÷ 3
	Activity	+ 0.3 %	+ 1 %	- 2 %	÷ 5
	Gamma source	- 0.7 %	- 1 %	- 3 %	÷ 3
	γ-dose at 1 m	+ 6 %	+ 2 %	- 1 %	÷ 2
	Neutron source	- 5 %	× 4	× 6	/
End of irradiation + 5 years	Decay heat	+ 80 %	× 3	× 6	× 32
	Activity	+ 5 %	+ 13 %	+ 30 %	× 5
	Gamma source	+ 1 %	+ 5 %	+ 7 %	+ 43 %
	γ-dose at 1 m	- 1 %	- 2 %	- 2 %	÷ 4
	Neutron source	- 6 %	× 4	× 8	/

Am TRANSMUTATION IN A FAST REACTOR

A special case of interest is the transmutation of Am. It seems, on the basis of the previous discussion that, to avoid large consequences on the reactor and on the fuel cycle, one should focus on the heterogeneous mode strategy.

However during eventual multirecycling, large amounts of Cm are produced. For the Cm one can imagine different strategies :

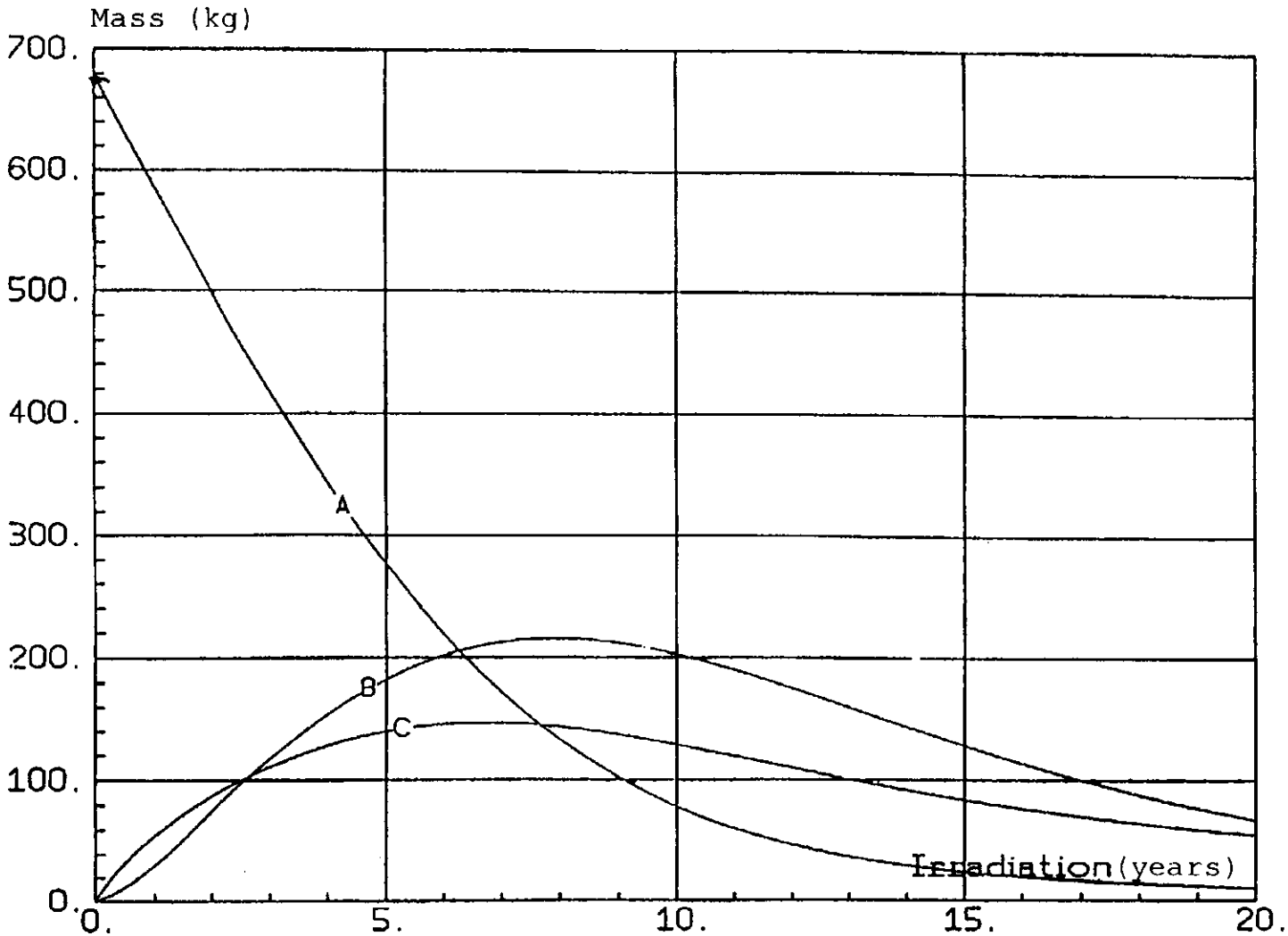
- Temporary storage, waiting for the decay to Pu (60 - 100 y).
- Recycle as for Am - In that case higher mass MA production is to be expected.
- Reduction of its production.

For that last purpose, one should try to fission the original Am up to 90 - 95 %.

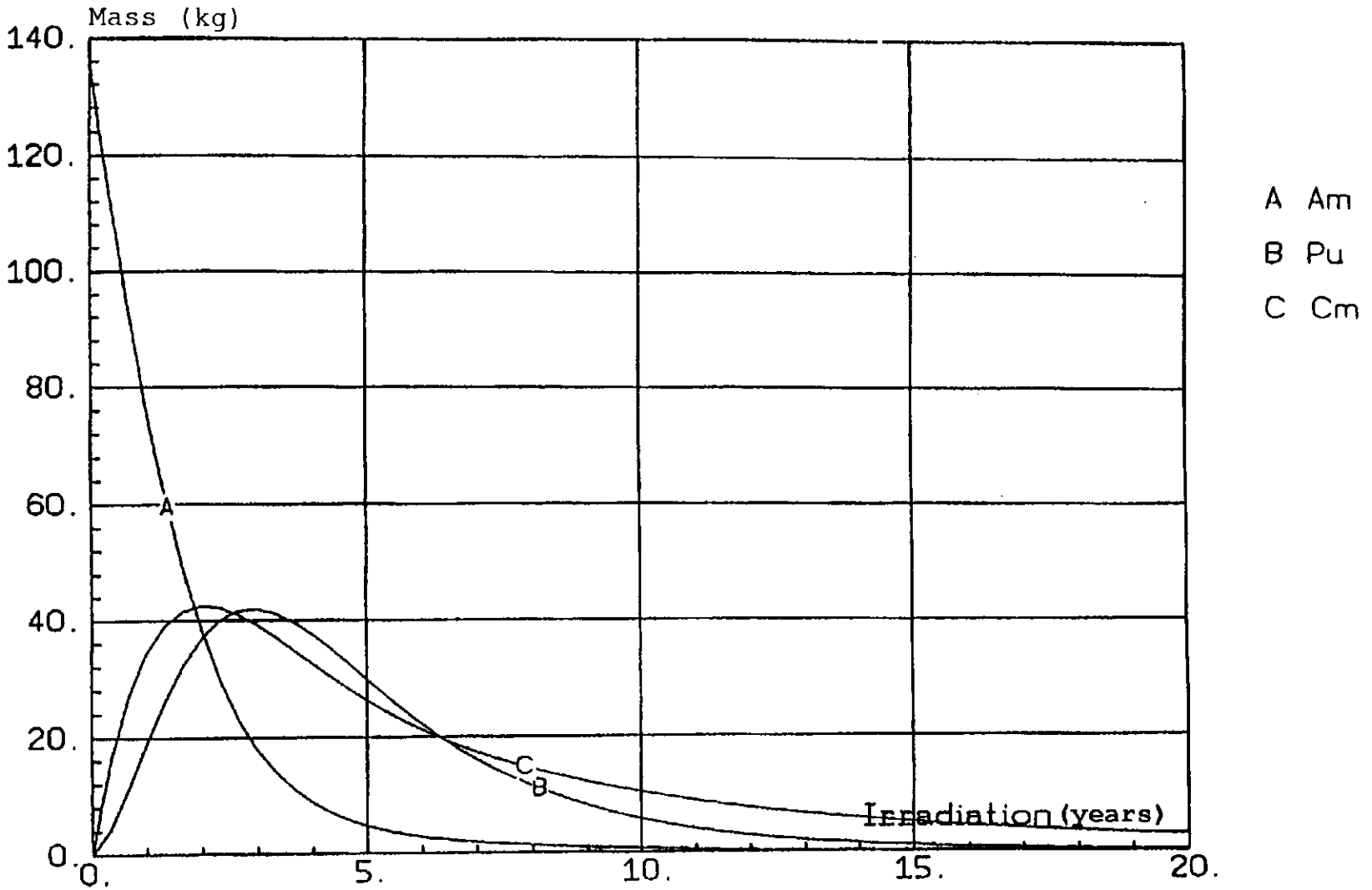
This means a very long irradiation in a high flux at the periphery of a FR, possibly in a thermalized spectrum environment. However, high damages to cladding (> 200 DPA NRT, displacement per atom, according to NRT model) are to be expected and the promising option of the "once-through" irradiation of Am targets in a fast reactor has to be optimized.

Results of a parametric study to optimize the target S/A in the Phenix reactor

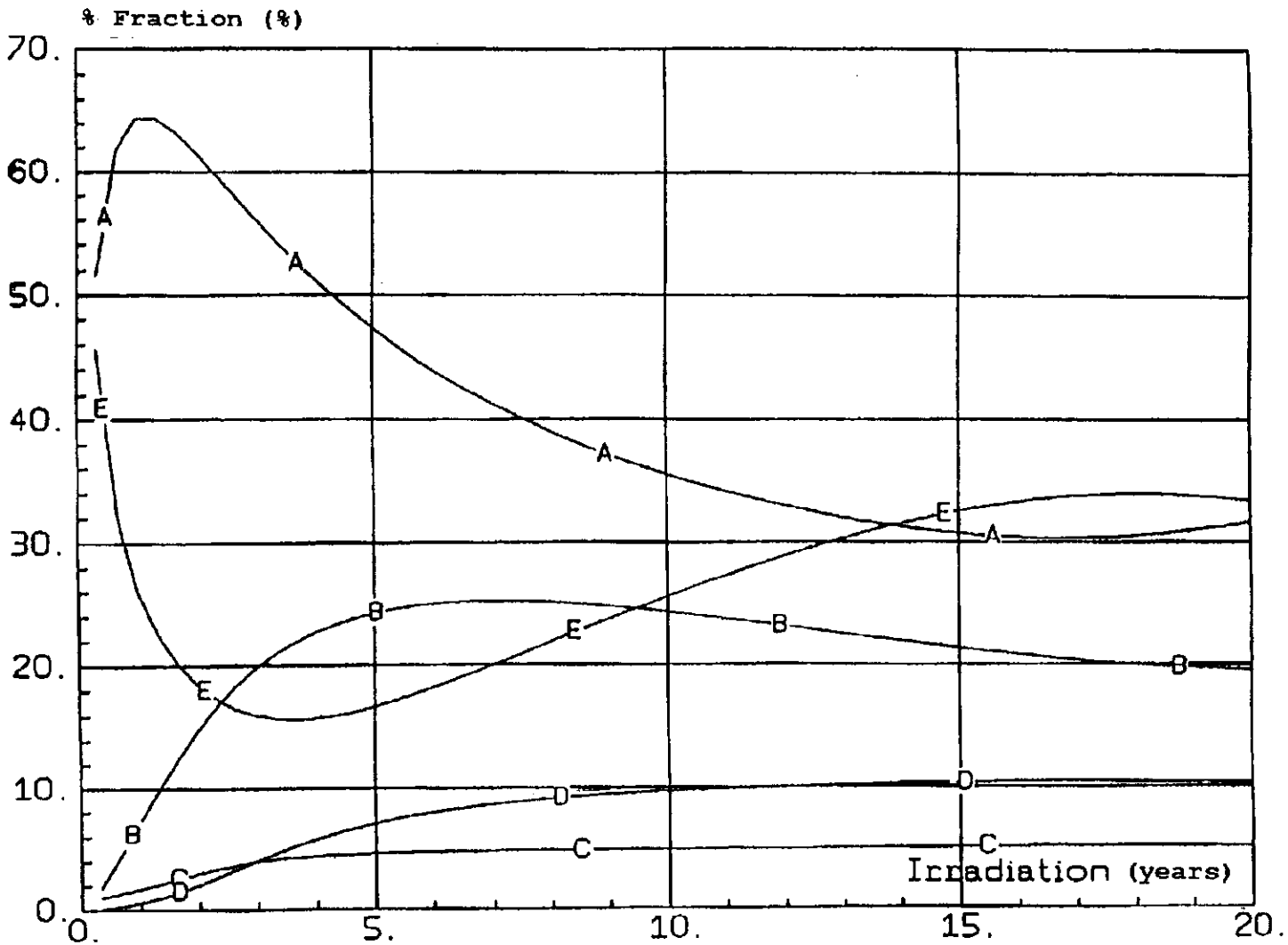
AmO_2 (%) (target)	25	10	5	5
CaH_2 (%) (moderator)	75	90	95	73
Al_2O_3 (%) (inert matrix)	0	0	0	22
	10 effective full power years			
Am consumption (%)	88	97	100	99
Am fission (%)	39	63	89	87
Am consumption (kg/TWhe)	31	13	6.3	6.3
Damage (dpa)	153	165	126	139
	15 effective full power years			
Am consumption (%)	96	99	100	100
Am fission (%)	64	84	96	95
Am consumption (kg/TWhe)	21	8.4	4.2	4.2
Damage (dpa)	236	239	178	198
	20 effective full power years			
Am consumption (%)	98	100	100	100
Am fission (%)	80	92	98	98
Am consumption (kg/TWhe)	16	6.3	3.1	3.1
Damage (dpa)	304	307	230	257



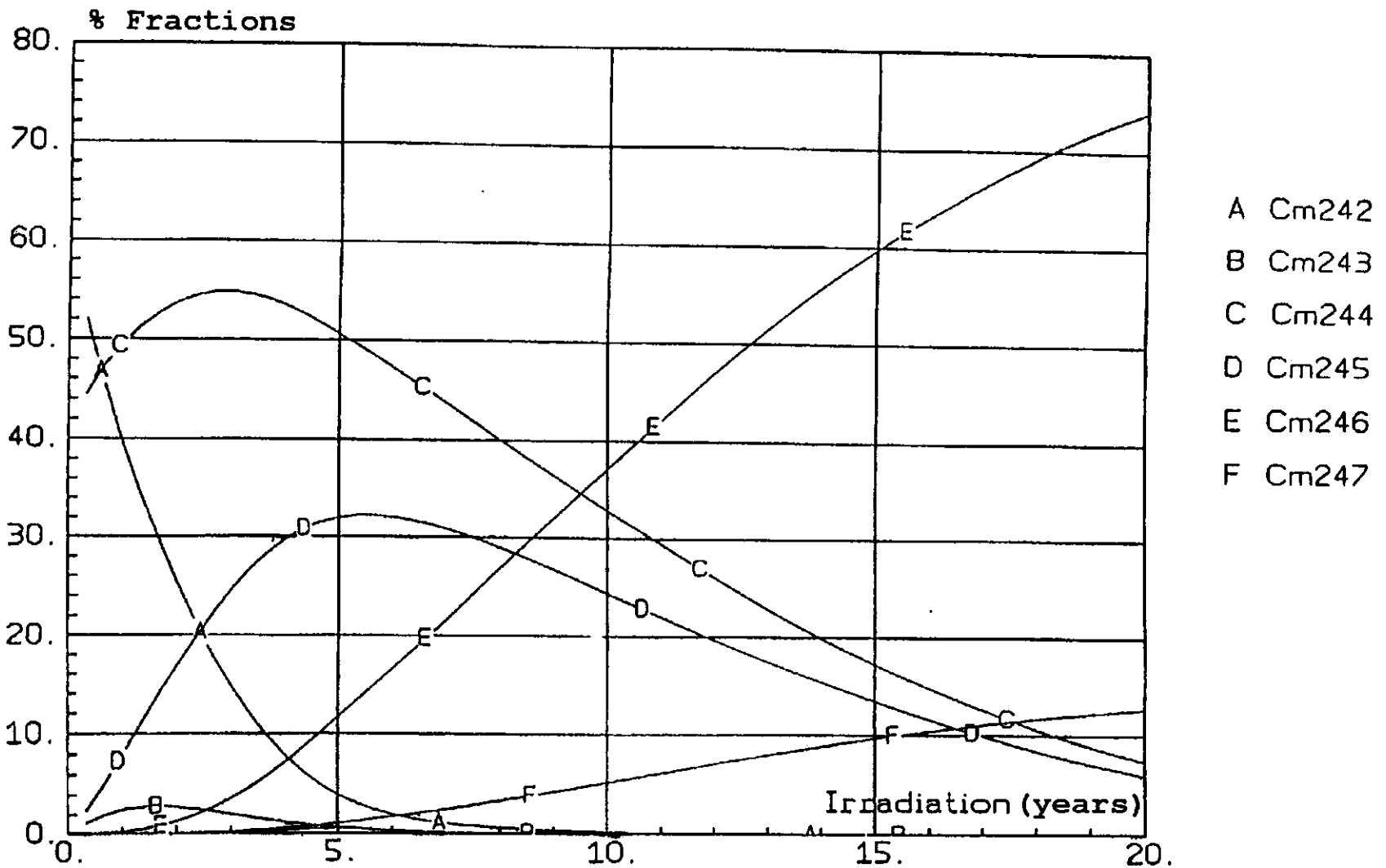
IRRADIATION of Am in Moderated S/A
25 % AmO_2 + 75 % CaH_2



IRRADIATION OF Am IN MODERATED S/A
5 % AmO_2 + 22 % Al_2O_3 + 73 % CaH_2



IRRADIATION OF Am IN MODERATED S/A
(case 5 % AmO_2 + 95% CaH_2)



IRRADIATION OF Am IN MODERATED S/A
(case 5 % AmO_2 + 95% CaH_2)

Am and Cm heterogeneous mode transmutation

Reduction of radiotoxicity (with respect to open cycle)

Time after disposal (years)	10^2	10^3	10^4
Open cycle	1	1	1
Pu recycling (100 % Am and Cm to wastes, 0.1 % Pu losses)	3	2.5	4.5
Pu recycling and Am targets irradiation			
Theoretical maximum reduction (Am = 0, 100 % Cm to wastes)	20	23	15
With cumulative fission rate = 90 %	12	17	10
= 95 %	16	20	13
Pu recycling and (Am + Cm) targets irradiation			
Theoretical maximum reduction (Am, Cm = 0)	490	400	390
With cumulative fission rate = 90 %	40	45	30
= 95 %	72	81	55

Am and Cm homogeneous mode transmutation

Reduction of radiotoxicity (with respect to open cycle)

Time after disposal (years)	10^2	10^3	10^4
Open cycle	1	1	1
Pu recycling (100 % Am and Cm to wastes, 0.1 % Pu losses)	3	2.5	4.5
Pu + Am recycling (homogeneous mode)			
Theoretical maximum reduction (Am = 0, 100 % Cm to wastes)	20	23	15
Actual recycling (with 1 % Am losses)	7	10	6
Pu + Am + Cm recycling (homogeneous mode)			
Theoretical maximum reduction (Am, Cm = 0)	490	400	390
Actual recycling (with 1 % Am, Cm losses)	90	100	120
Actual recycling (with 0.1 % Am, Cm losses)	270	335	310

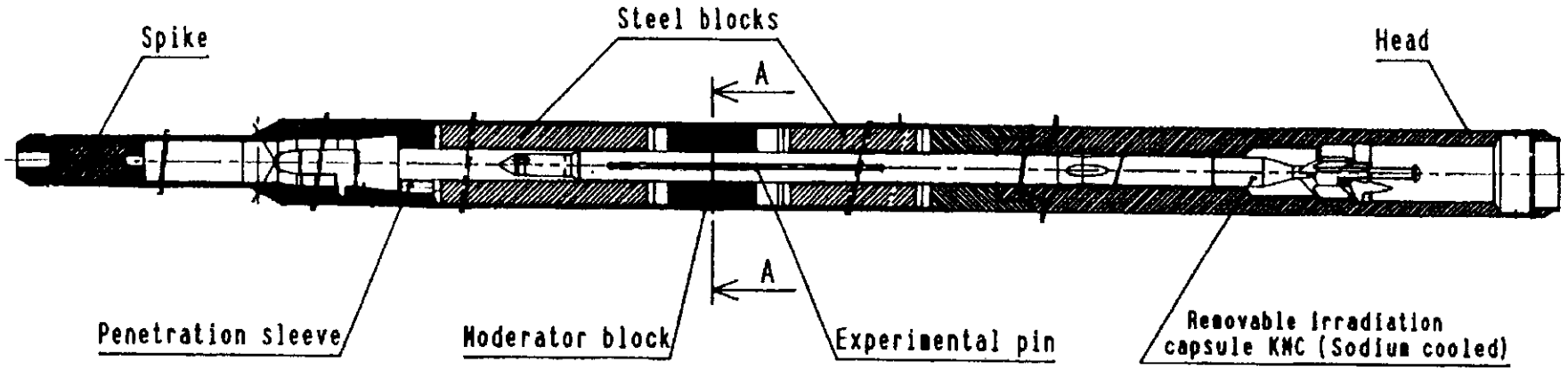
Influence of Cm for target fabrication

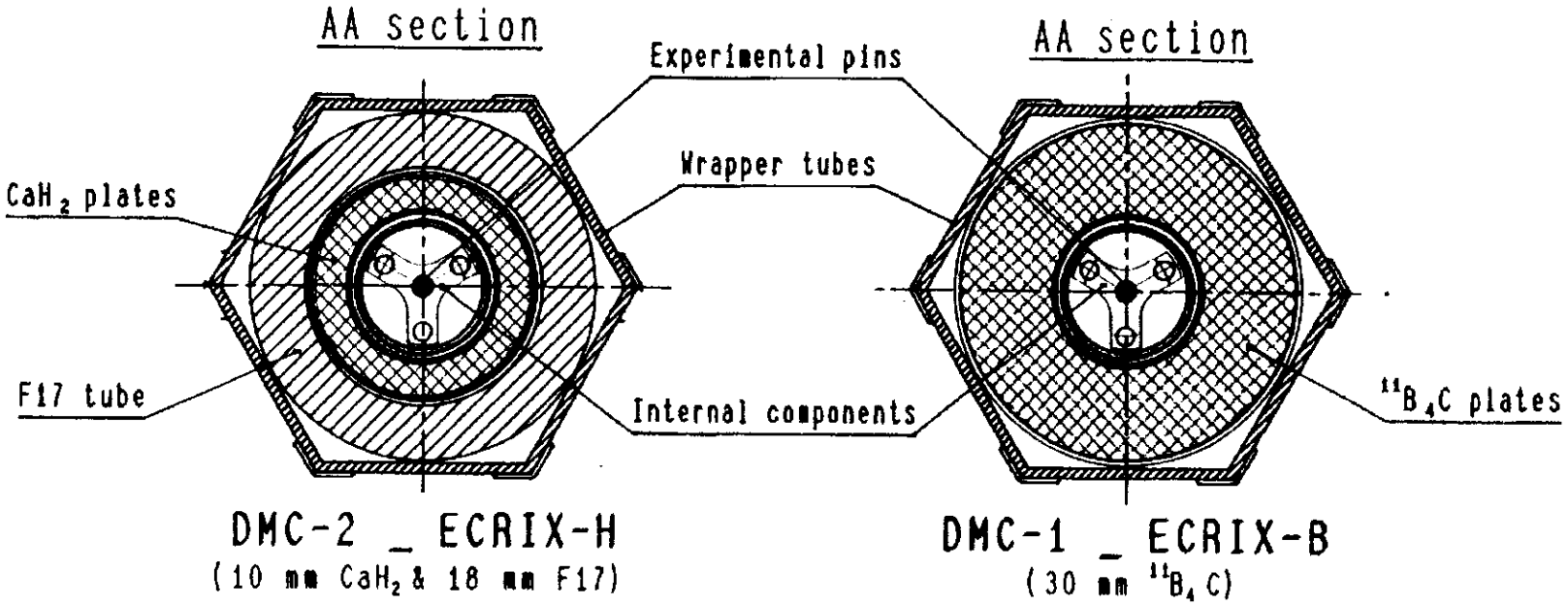
	100% Am	90% Am + 10% Cm	80% Am + 20% Cm
Heat	1	× 2.3	× 3.6
γ dose at 1 m	1	× 1.5	× 2
Neutronic source	1	× 120	× 240

Few Experiments have been performed up to now to validate the target concept/materials.

One example : the ECRIX experiment planned at PHENIX (under preparation)

REMOVABLE IRRADIATION CAPSULE MODERATOR ASSEMBLY DMC-1 ($^{11}B_4$ C) & DMC-2 (CaH_2)





As far as existing experimental validation, a special case is the SUPERFACT experiment performed in PHENIX, in the '80s.

SUPERFACT : EXPERIMENT IN PHENIX

COMPOSITION OF HOMOGENEOUS AND FRESH NEPTUNIUM - AND AMERICIUM BASED FUELS*

Pin Number and Type	Composition[a] required	Measured After Fabrication
8 and 16, standard	$(U_{0,75}\ Pu_{0,25})\ O_{2-x}$	$(U_{0,718}\ Pu_{0,282})\ O_{1.983}$
7 and 13, 2 % neptunium	$(U_{0,74}\ Pu_{0,24})\ O_{2-x}$	$(U_{0,741}\ Pu_{0,244}\ Np_{0,0150})\ O_{1.973}$
4 and 16,2 % americium	$(U_{0,74}\ Pu_{0,24}\ Am_{0,02})\ O_{2-x}$	$(U_{0,745}\ Pu_{0,237}\ Am_{0,0184})\ O_{1.957}$
5 and 15, 45 % neptunium	$(U_{0,55}\ Np_{0,45})\ O_{2-x}$	$(U_{0,552}\ Np_{0,4482})\ O_{1.996}$
6 and 14, 20 % neptunium and 20 % americium	$(U_{0,60}\ Np_{0,20}\ Am_{0,20})\ O_{2-x}$	$(U_{0,596}\ Np_{0,2118}\ Am_{0,1918})\ O_{1.926}$

* Pellet diameter = 5.42 mm ; height = 7 mm

[a] As a percent of weight for actinides

SUPERFACT : IRRADIATION CONDITIONS IN PHENIX

Pin Number and Type	Burnup (at. %)		Linear Power (W/cm)
	CEA Calculation	ITU Measurement	CEA Calculations Beginning and End of Life
8 and 16, standard	8.5	/	430 370
7 and 13, 2 % neptunium	6.8	6.4	380 325
4 and 16, 2 % americium	6.8	6.4	380 325
5 and 15, 45 % neptunium	4.6	4.5	206 283
6 and 14, 20 % neptunium and 20 % americium	4.3	4.5	174 273

SUPERFACT EXPERIMENT

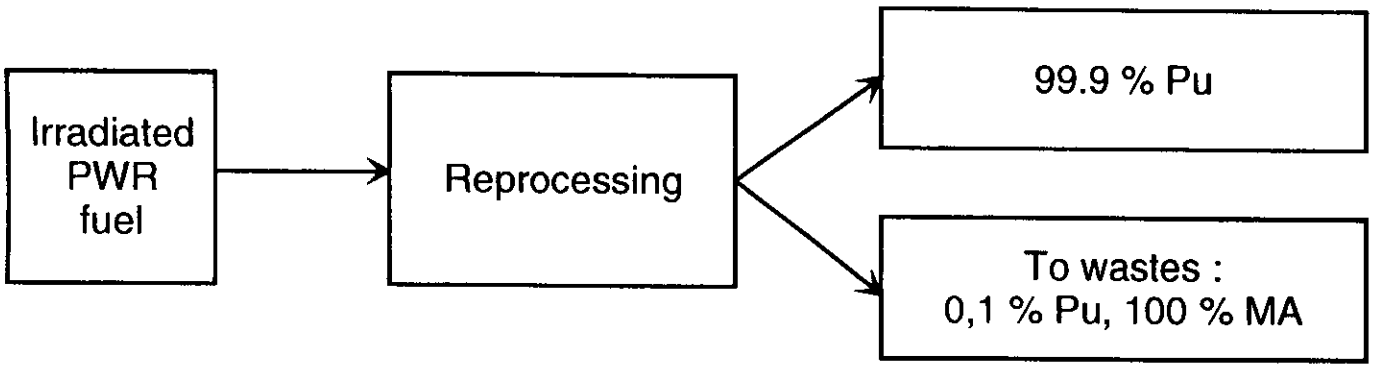
Minor Actinide	Neptunium			Americium	
Percentage of MA in the fresh fuel	2	20	45	2	20
Transmutation rate (%) measurement	-	-	27	27 to 30	27 to 32
Transmutation rate (%) calculation	24 to 26	27.4	27.5	27.3	31.6

* Percent

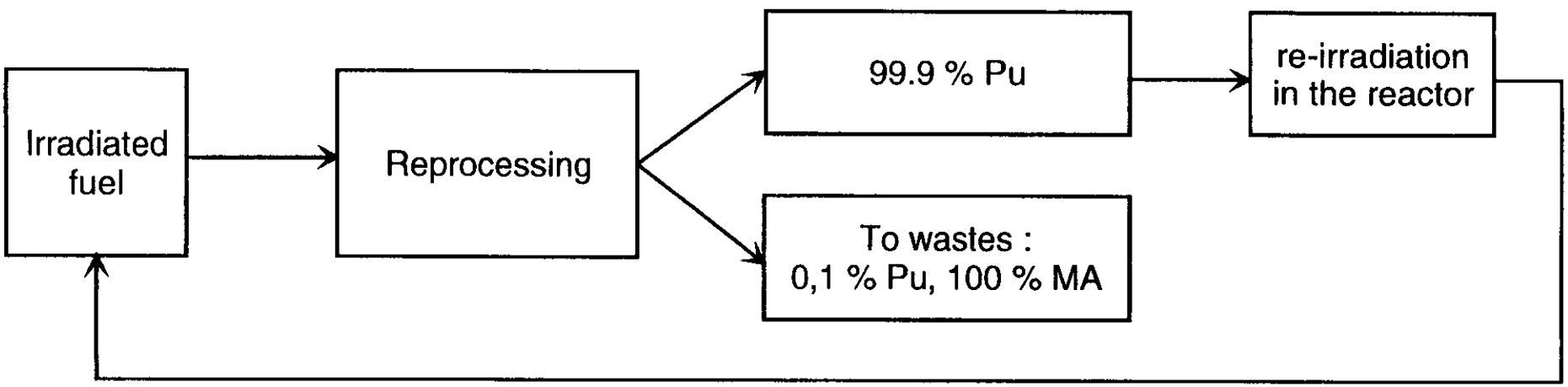
Radiotoxicity reduction in a power Park

For example :

In theory :

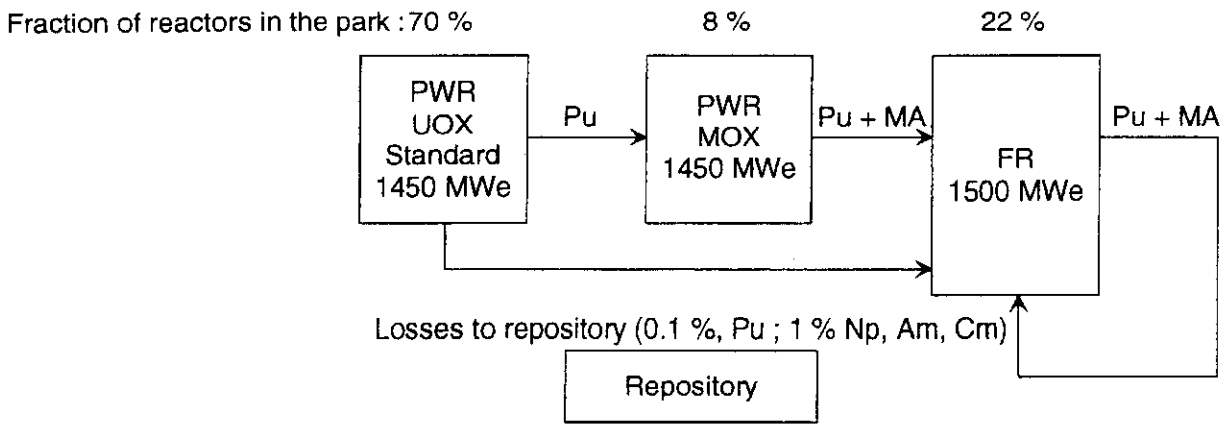


In reality :



Scenarios with P/T, at equilibrium

Example of a reactor park (60 GWe), with which it is possible to reach equilibrium i.e. consumption = production of Pu, Am, Np :

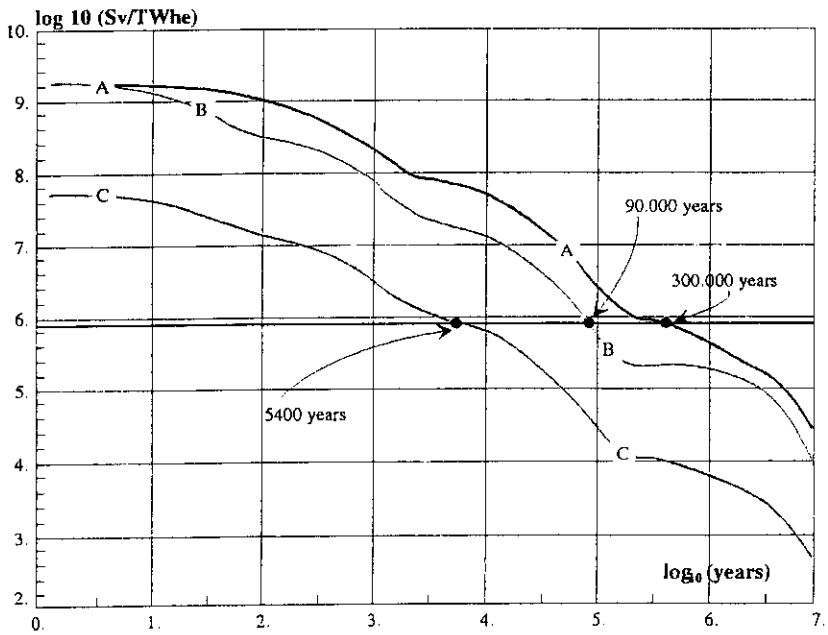


Np : homogeneously recycled in the core : $(UPuNp)O_2$ fuel

Am : heterogeneous recycling (targets at the periphery of the core : AmO_2 + inert matrix)

Cm : separated and left to decay → Pu, then recycled together with the rest of Pu.

Radiotoxicity as a function of time



A : Open cycle

B : Recycling only of Plutonium. 0.1 % Pu ; 100 % Am, Np, Cm, etc ... to the Repository

C : Scenario with P/T. 0.1 % Pu ; 1 % Am, Np, Cm losses to the Repository

Long-Lived Fission Product (LLFP) Transmutation

In a transmutation process, fission products, unlike transuraniums, can not produce supplementary neutrons when put in the neutron field. That is why this is a strictly neutron consumption process. Neutron consumption is the most important parameter, if one wants to assess the potential of transmutation in a given nuclear system.

The rate of transmutation of a J-nucleus can be characterised by the time T_J^{transm} needed to incinerate half of initial mass which is function of the cross-section $\sigma_{n,\gamma}^J$ (barns) and of the neutron flux Φ (n/cm^{2}s) :

$$T_J^{transm} = \frac{\ln 2}{\sigma_{n,\gamma}^J \Phi \times 3.16 \times 10^7} \text{ years}$$

Transmutation of the toxic fission products in nuclear reactors and subcritical systems has a sense if rates of nuclear interactions with neutrons are much higher than rates of natural decays, which are defined by decay life-time $T_{1/2}$. That is, transmutation under neutron flux can be reasonable if $T_{1/2} >> T_J^{transm}$.

PHYSICS PARAMETERS OF THE MAJOR LLFP

Isotope	Period (y)	Decay mode	Thermal power (W/Bq)	Dose (ingestion) (Sv/Bq)	Fraction in an irradiated fuel (g/t) [(a)]
^{14}C	5.7×10^{3}	β	1.6×10^{-14}	5.7×10^{-10}	1.3×10^{-1}
^{36}Cl	3.0×10^{5}	β^-, β^+	4.4×10^{-14}	8.2×10^{-10}	1.6×10^{0}
^{79}Se	6.5×10^{4}	β	6.5×10^{-15}	2.3×10^{-9}	4.7×10^{0}
^{90}Sr	2.9×10^{1}	β	2.8×10^{-14}	3.9×10^{-8}	5.0×10^{2}
^{90}Y	7.3×10^{-3}	β	1.5×10^{-13}		1.3×10^{1}
^{93}Zr	1.5×10^{6}	β	2.6×10^{-15}	4.2×10^{-10}	9.8×10^{1}
^{99}Tc	2.1×10^{5}	β	1.4×10^{-14}	3.4×10^{-10}	8.2×10^{2}
^{107}Pd	6.5×10^{6}	β	1.4×10^{-15}	3.7×10^{-11}	2×10^{2}
^{126}Sn	1×10^{5}	β	4.2×10^{-14}	5.1×10^{-9}	2.0×10^{1}
^{126}Sb	3.4×10^{-2}	β	5.0×10^{-13}		6.9×10^{-6}
^{129}I	1.6×10^{7}	β	1.3×10^{-14}	7.4×10^{-8}	1.7×10^{2}
^{135}Cs	2.3×10^{6}	β	9×10^{-15}	1.9×10^{-9}	1.3×10^{3}
^{137}Cs	3.0×10^{1}	β	3.2×10^{-14}	1.4×10^{-8}	1.1×10^{3}
^{137m}Ba	4.9×10^{-6}	β	1.1×10^{-13}		1.7×10^{-4}
^{151}Sm	9.0×10^{1}	β	3.2×10^{-15}	9.1×10^{-11}	1.6×10^{1}

(a) UOX of a PWR (3.5 %, 33 Gwd/t)

Parameters of long lived nuclei to be eventually transmuted in a fast (E_n (neutron energy) = 0.2 MeV) and thermal (E_n = 1 eV) spectra with standard flux levels : $\Phi = 10^{15}$ (n/cm^{2}s) and $\Phi = 10^{14}$ (n/cm^{2}s) respectively

Isotopes, J	$\sigma^J_{n,\gamma}$ (barns)		T1/2 (years)	T_J^{transm} (years)		Recommendation to transmutation
	fast spectrum	thermal spectrum		fast spectrum	thermal spectrum	
^{79}Se	0.03	0.1	6.5×10^4	7.3×10^2	2.2×10^3	*questionable*
^{90}Sr	0.01	0.14	29	2.2×10^3	1.6×10^3	*non-transmutable*
^{93}Zr	0.03	0.28	1.5×10^6	730	790	*transmutable*
^{94}Nb	0.04	2.2	2.0×10^4	5.5×10^2	1×10^2	*questionable or transmutable*
^{99}Tc	0.2	4.3	2.1×10^5	110	51	*transmutable*
^{107}Pd	0.5	0.3	6.5×10^6	44	730	*transmutable*
^{126}Sn	0.005	0.05	1×10^5	4.4×10^3	4.4×10^3	*questionable*
^{129}I	0.14	4.3	1.6×10^7	160	51	*transmutable*
^{135}Cs	0.07	1.3	2.3×10^6	310	170	*transmutable*
^{137}Cs	0.01	0.02	30	2.2×10^3	1.1×10^4	*non-transmutable*
^{151}Sm	0.7	700	89	31	0.3	*non-transmutable or questionable*

There are at least 5 long lived isotopes : ^{99}Tc, ^{129}I, ^{93}Zr, ^{107}Pd, ^{135}Cs which can be transmuted in a fast spectrum much faster than their natural decay.

For three isotopes : ^{90}Sr, ^{137}Cs and ^{151}Sm, natural decay is similar or much faster than transmutation and it is more reasonable to put them into interim storage for decaying.

As for "questionable" isotopes : ^{79}Se, ^{126}Sn and ^{94}Nb, they are rather long lived, however, due to small cross-sections, their transmutation will go slowly.

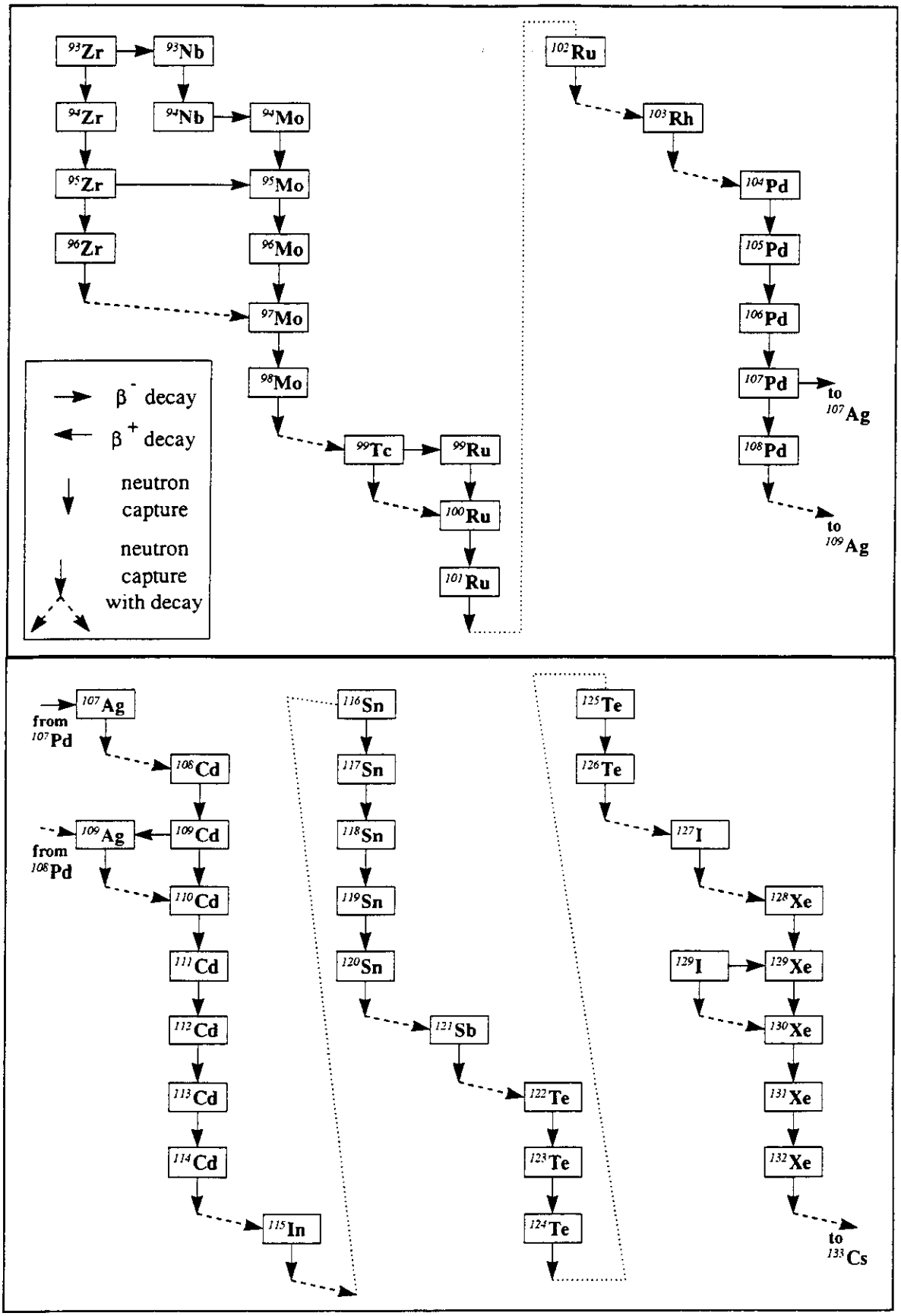
However, the yield of these isotopes is limited indicating that total toxicity is rather modest.

If one wants to transmute the particular J nucleus together with all its "*daughters*", it is useful to adopt the following definition of LLFP families to be transmuted :

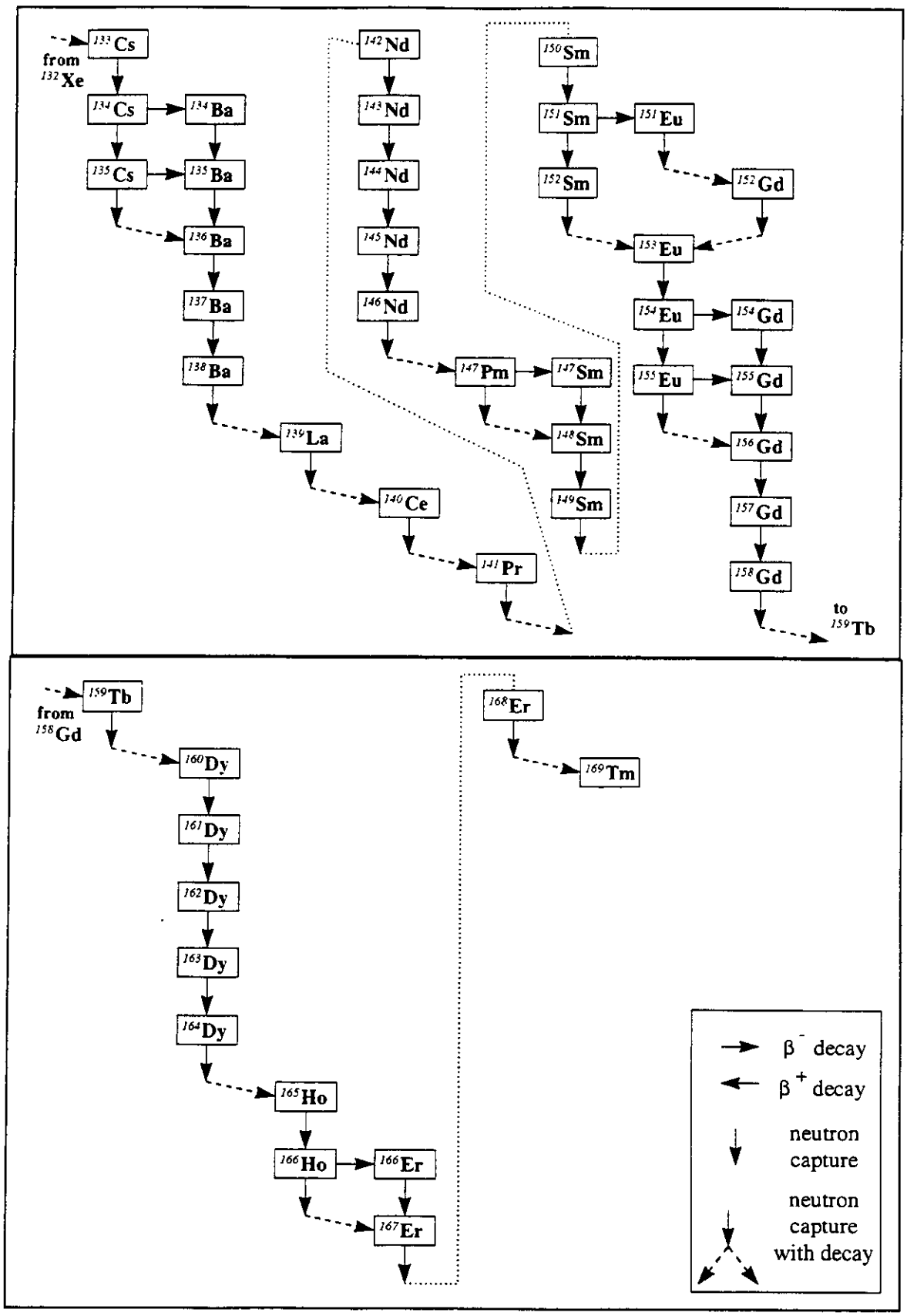
DEFINITION : *J-family is a set of nuclei (including the initial nucleus J) which have been produced both by the transmutation in a neutron flux Φ of the J nucleus as well as of its subsequent products, and by decay of nuclei.*

The overall neutron consumption D_J^* of the LLFP family (J-family) can be defined as the total number of neutrons which have to be spent to incinerate all this family.

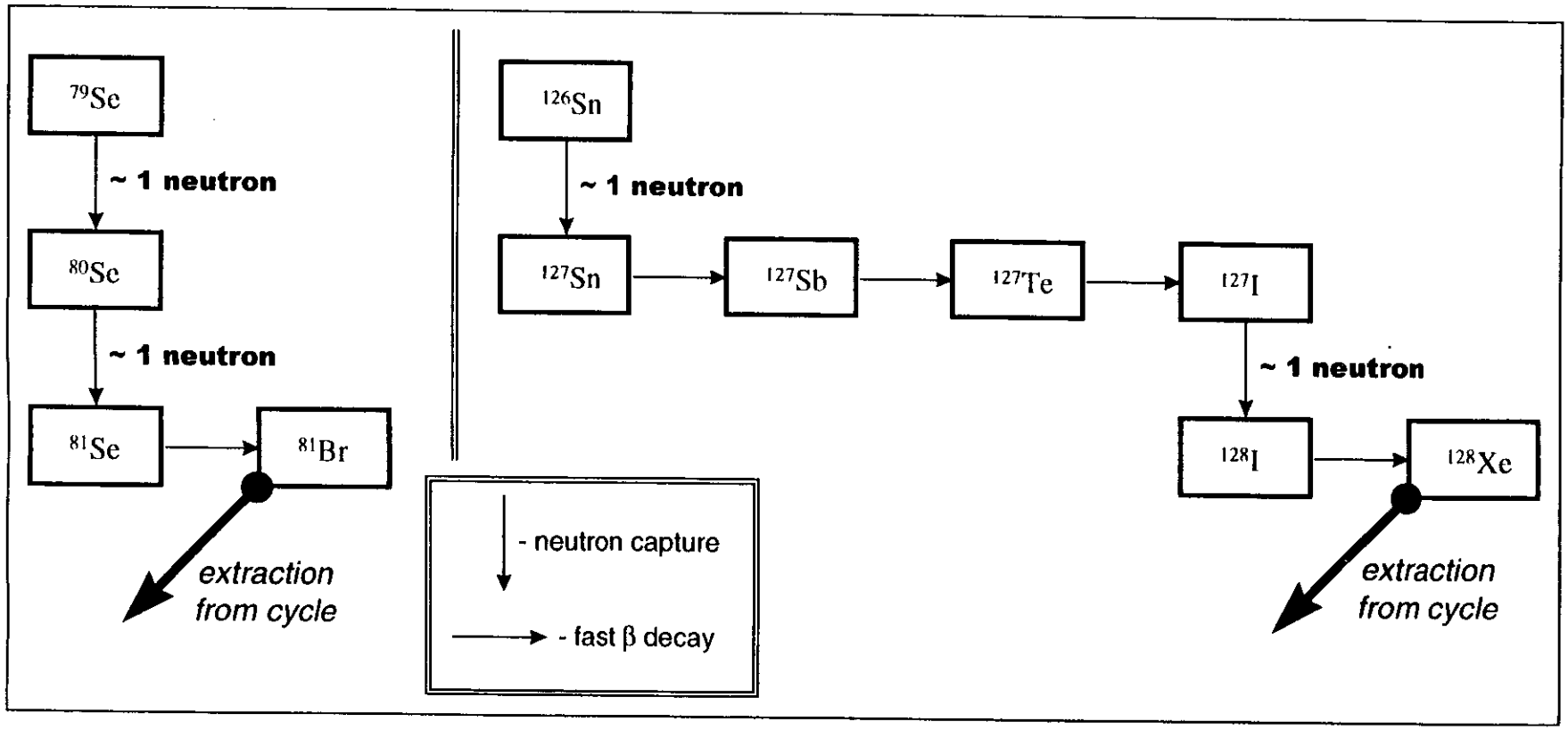
Removal of stable and short lived fission products can be considered as a single sink of long-term radioactive fission products.



LLFP transmutation scheme



LLFP transmutation scheme *(continuation)*



Transmutation schemes for ^{79}Se and ^{126}Sn isotopes.

Overall neutron consumption (D_J^*) of "transmutable" and "questionable" isotopes together with their yields (Y_J) per 1 fission in LWR (UOX) after 5 years of cooling time. Time interval between reprocessing steps - 3 years (removable nuclides : all fission products except all isotopes of Zr, Tc, Pd, I, Cs, Sn, Nb, Se)

Transmutable and questionable isotopes (J)	D_J^* neutron/transmutation	Y_J nuclei/fission in NP
^{93}Zr	2.01	0.050
^{99}Tc	1.01	0.055
^{107}Pd	2.04	0.015
^{129}I	1.008	0.009
^{135}Cs	1.002	0.017
^{126}Sn	~2	0.0012
^{94}Nb	0.985	6.3×10^{-7}
^{79}Se	~2	0.0004

Overall neutron consumption (D_J^*) of "transmutable" nuclides (including all isotopes) together with its yields (Y_J) per 1 fission in LWR (UOX) after 5 years of cooling time. Time interval between fission products reprocessing - 3 years (removable nuclides : all fission products excluding all isotopes of Zr, Tc, Pd, I, Cs)

Transmutable nuclides, J	D_J^* (neutron/transmutation)	Y_J (nuclei/fission in NP)
all Zr	2.03	0.26
all Tc	1.01	0.055
all Pd	3.22	0.095
all I	1.01	0.011
all Cs	0.58	0.13

It is helpful to use two types of neutron consumption definitions, depending on units choice - D_J^* (neutron/transmutation) and D_J (neutron/fission). D_J can be obtained as the product of D_J^* and of the yield of J nuclide per fission, Y_J. If a LLFP transmuter is fed constantly by a group of nuclides, then the D value of this group is the sum of $D_J^* Y_J$ of the group components J.

For example, taking into account LLFP yield, one can calculate the total neutron consumption needed to incinerate all "transmutable" and "questionable" long lived isotopes of fission products :

$$D(^{93}Zr, ^{99}Tc, ^{107}Pd, ^{129}I, ^{135}Cs, ^{126}Sn, ^{94}Nb, ^{79}Se) = \sum_J D_J^* Y_J \cong 0.22 \text{ (neutron / fission in NP)}$$

This value defines the total neutron consumption for incineration of all LLFP presented above, if preliminary isotope separation of fission product in LWR discharge has been realised.

To incinerate all Tc, I, Cs <u>without isotopic separation</u>, one needs about 0.15 (neutron/fission in NP).

<u>Isotopic separation</u> of ^{99}Tc, ^{129}I and ^{135}Cs allows to reduce this neutron consumption to 0.08 (neutron/fission in NP).

To transmute elements such as Tc, I, Cs in a fast spectrum transmuter one needs to know the neutron surplus G available and to the fraction (f) of these transmuters in a Nuclear Power Park (NPP) :

$$f \times G \geq 0.15 \text{ neutron/fission in NPP}$$

with the hypothesis that the yield of Tc, I, Cs of a transmuter is similar to the yield in the standard LWR and that the LLFP transmutation takes place in these transmuters only.

For transmutation of ^{99}Tc, ^{129}I and ^{135}Cs this demand can be much smaller :

$$f \times G \geq 0.08 \text{ neutron/fission in NPP}$$

Taking into account a "standard" value of the neutron parasitic capture (CM) and the neutron leakage (L) as CM+L = 0.3 neutron/fission (which is valid for a fast reactor of an intermediate size and traditional composition), one get for the neutron surplus (for example in a subcritical system) :

$$G = -\sum_J \varepsilon_J \times D_J - (CM + L) + \mu,$$

where ε_J is a fraction of J-nucleus in fuel, μ is a neutron spallation source ($\mu \approx 0.15$ neutron/fission if $k_{eff} = 0.95$).

$$G_{TRU} = 1.11 - 0.3 + 0.15 = 0.96 \text{ neutron/fission}$$
$$G_{TRPu} = 0.72 - 0.3 + 0.15 = 0.57 \text{ neutron/fission}$$

For example, if the fast spectrum TRU transmuter (G = 0.96 neutron/fission) is used both for the TRU transmutation and Tc, I, Cs incineration, then one can assess the fraction f :

$$f \geq \frac{0.15 \text{ neutron / fission}}{G = 0.96 \text{ neutron / fission}} \approx 0.16$$

EVALUATION OF TRANSMUTATION RATES

The maximum rate of transmutation (*RT*, measured in number of transmutations per fission) of any isotope in a given transmuter can be easily evaluated if the neutron surplus *G* available for LLFP transmutation is known :

$$RT_J = \frac{G(\text{neutron / fission})}{D_J^*(\text{neutron / transmutation})}$$

One can then calculate the maximum rate R_J^{max} of transmutation of a given isotope *J* per $GW_t \times$ year of a transmuter :

$$R_J^{max}(\text{kg / GWt} \times \text{year}) \approx 1.6 \times A_J \times RT_J$$

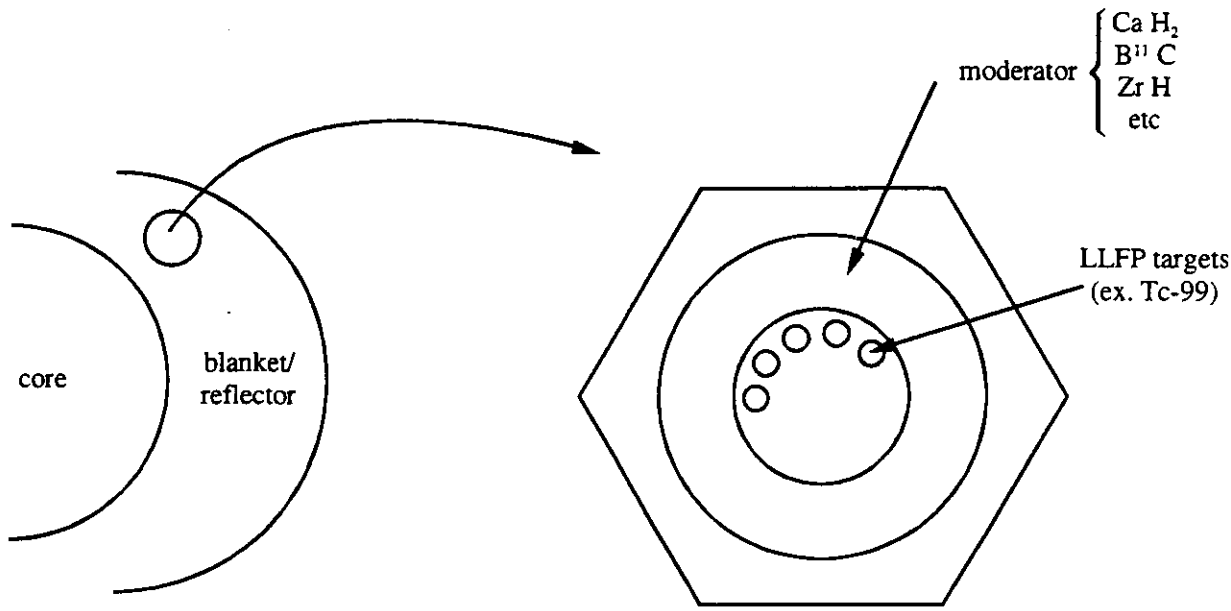
where A_J is atomic number of *J* isotope.

For example :

D_J^* is equal to 1.01 for ^{99}Tc and, in a fast spectrum subcritical transmuter based on TRU of LWR-UOX, $G = 0.96$. Then, $R_J^{max} \cong 160$ kg/GW_t×year if all neutron surplus will be spent for ^{99}Tc transmutation.

R_J^{max} defines the potential of a given transmuter to incinerate the *J* isotope of LLFP.

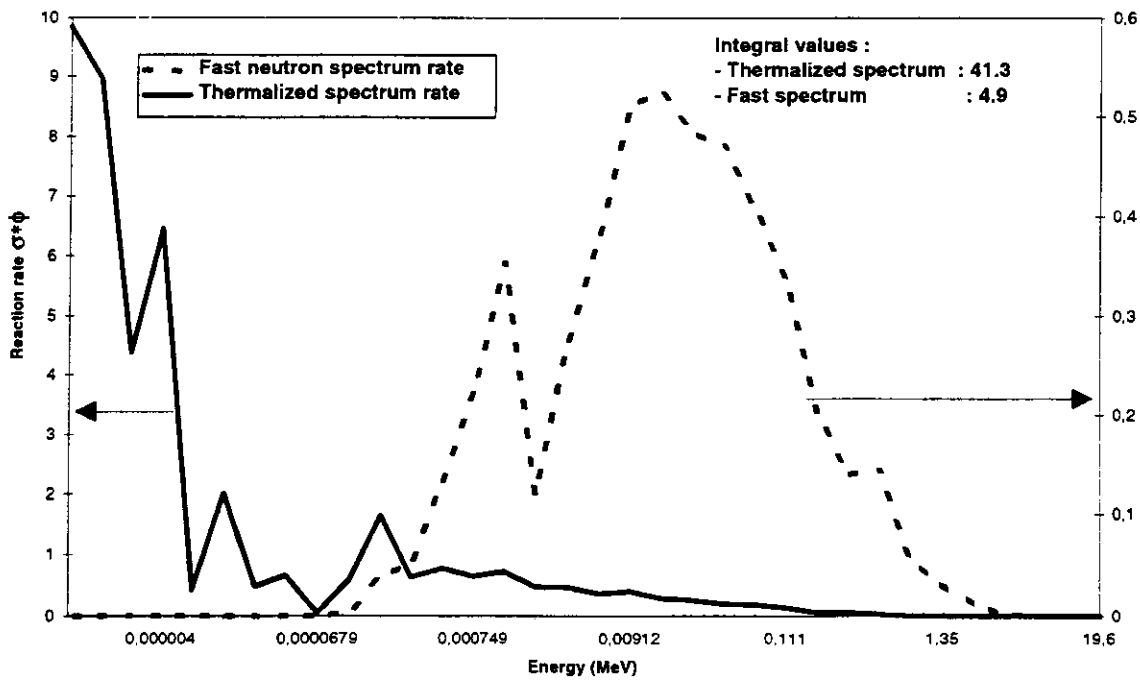
"Leakage with Slowing Down" (LSD) concept for LLFP transmutation



Transmutation in a "moderated" S/A :

1 - The neutron surplus available leaking out of the core, is used in the blanket/reflector.
2 - These neutrons are slown down to the required energy ("spectrum tailoring") using moderator (not-absorbing) materials in the specific S/A.

Example : Reaction rate of Tc-99



Transmutation of ^{99}Tc or ^{129}I

Exemple 1 : *In fast spectrum.*

	Masses (kg/TWhe)			σ_c	Φ	$T_{1/2}$ (transm)
	Loaded	Transmuted	%	barns	10^{15} n/cm^2/s	years
^{99}Tc	60.5	6.13	10.1	0.32	0.758	91
^{129}I	(26.0)	(3.96)	15.2	0.31	1.21	59

To be compared to productions :

$\rightarrow \approx 3$ kg/TWhe (^{99}Tc)

$\rightarrow \approx 0.7$ kg/TWhe (^{129}I)

Transmutation of ^{99}Tc

Exemple 2 : *In moderated spectrum.*

	Masses (kg/TWhe)			σ_c	Φ	$T_{1/2}$ (transm)
	Loaded	Transmuted	%	barns	10^{15} n/cm^2/s	years
1% ^{99}Tc	1.17	1.08	92	11.9	0.493	3.7
5% ^{99}Tc	5.86	3.62	62	5.85	0.372	10
10% ^{99}Tc	11.73	4.88	42	3.46	0.353	18
20% ^{99}Tc	23.46	6.19	26	2.19	0.317	32

To be compared to productions : ≈ 3 kg/TWhe (^{99}Tc) et ≈ 0.7 kg/TWhe (^{129}I)

PRESENT CONCLUSIONS

- FR, PWR transmutation potential well understood, both for homogeneous and heterogeneous recycling modes.

  Enough neutrons are available !

- Significant gains on the source of potential radiotoxicity.
- FR : better neutron economy, less consequences on the fuel cycle.
- Homogeneous recycling of Np looks promising.
- Heterogeneous recycling of Am in specific targets can be envisaged. However : mono- or multiple recycling ? (the problem of Cm production during Am transmutation).
- Cm : to be separated and left to decay is an option. A better option : targets with both unseparated Am + Cm, and high total fission rate ($\geq$ 95 %)
- Role of lanthanides to be investigated. How much is allowed ? (case of IFR).
- Fission Products transmutation : potential of S/A with moderator at FR core periphery, where FP target can be localised.
- Potential of accelerator-based devices for dedicated cores (MA fuels and FP targets).
- Partitioning issues are crucial (feasibility, efficiency).

PARTITIONING OF LLRN : WHAT PROCESSES ?

- Priority : Hydrometallurgy

  - Reprocessing experience :
    - potential for high performances of partitioning
    - low level of technological wastes
  - Target date 2006 : "Technological continuity" is privileged.

- Alternative options are explored ... ⇒ Pyrometallurgy

  - Potentially interesting (compactness, aptitude to handle specific objects, etc ...).
  - Interest renewed by innovative options for transmutation.

The situation today, The PUREX process

The PUREX process, which is universally employed in the irradiated fuel reprocessing industry, is a hydrometallurgical process based on the use of TBP, a solvent containing phosphorus.

Uranium and plutonium, whose stable oxidation degrees in nitric medium are VI and IV respectively, are co-extracted by TBP, and thus separated from the bulk of the fission products which remain in the aqueous phase. This is the basic principle of the PUREX process.

	DEGREES OF OXIDATION			
	III	IV	V	VI
U				●
Np			●	●
Pu		●		
Am	●			
Cm	●			

▒ extractible by TBP

□ not extractible by TBP

Uranium and plutonium are recovered with an industrial yield close to 99.9 % (including losses in secondary wastes).

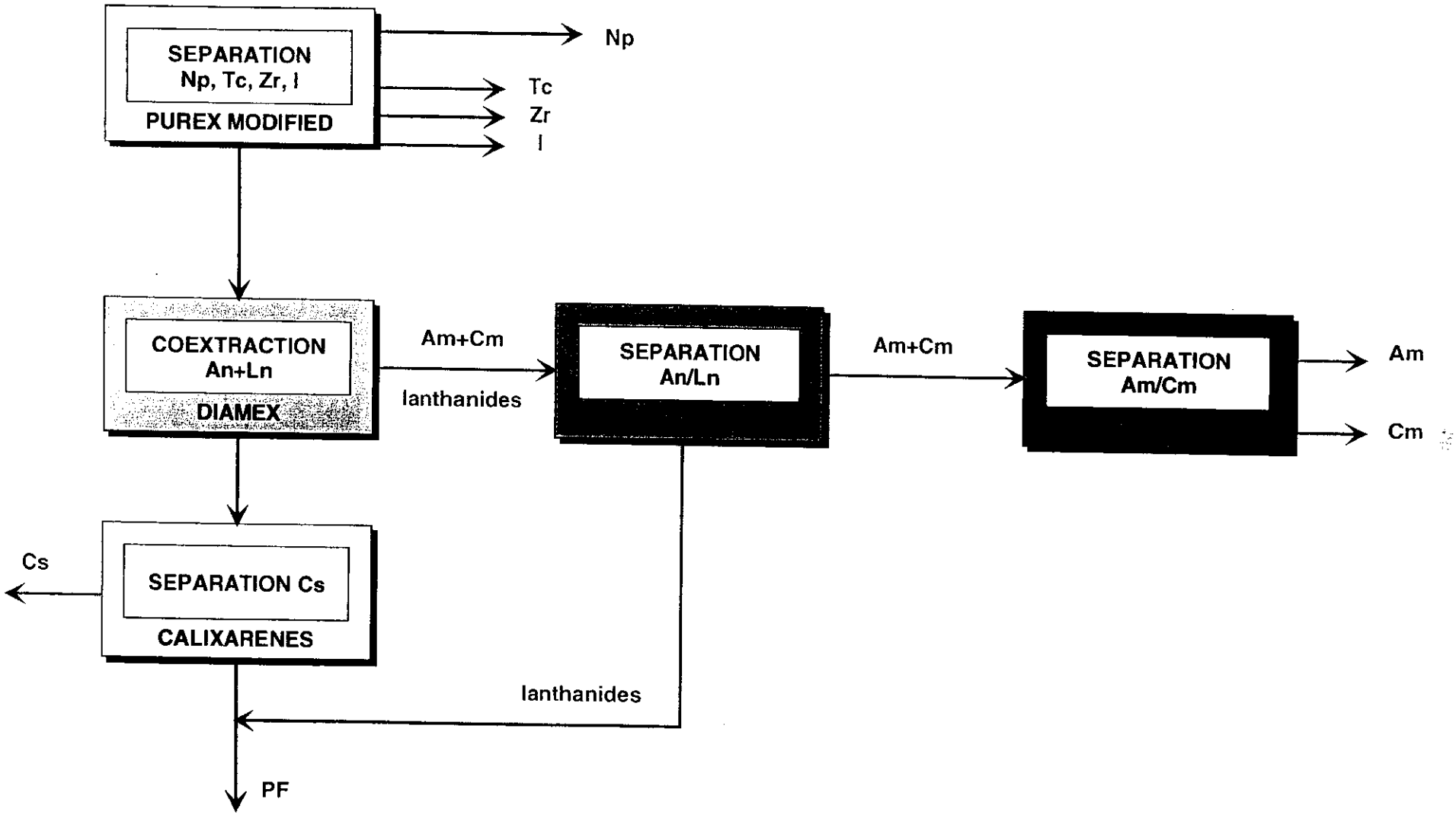
Separation of Americium and Curium

First step : Actinides (III) / Lanthanides (III) co-extraction
Several options, like the DIAMEX process developed at CEA
(Advantage : a totally organic and degradable compound, the CHON principle)

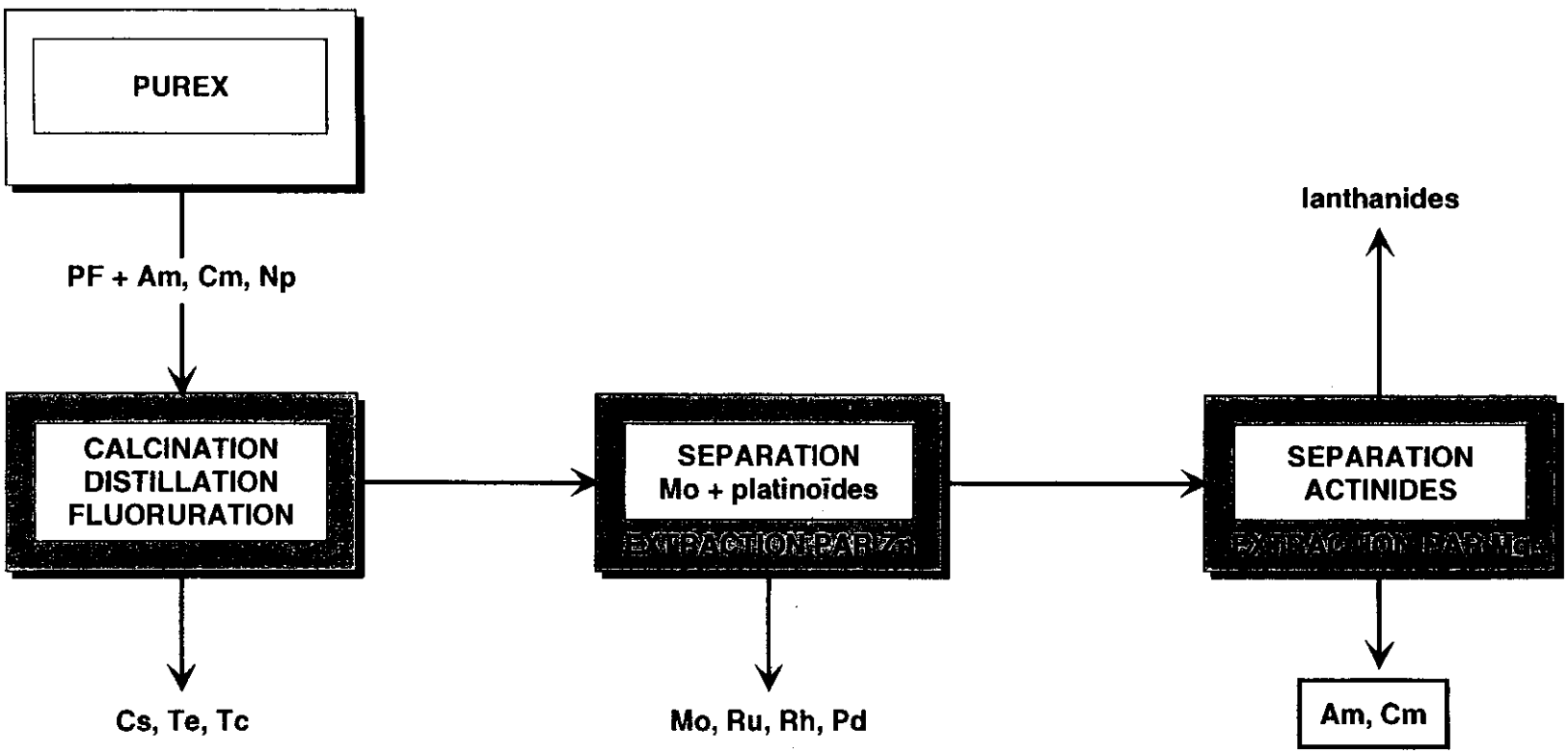
Second step : Actinides (III) / Lanthanides (III) separation
Active research underway

Third step : Americium/Curium separation

Example : The SESAME process, an electrochemical method to bring Am to oxidation IV or VI and then separate it in the oxidized state.

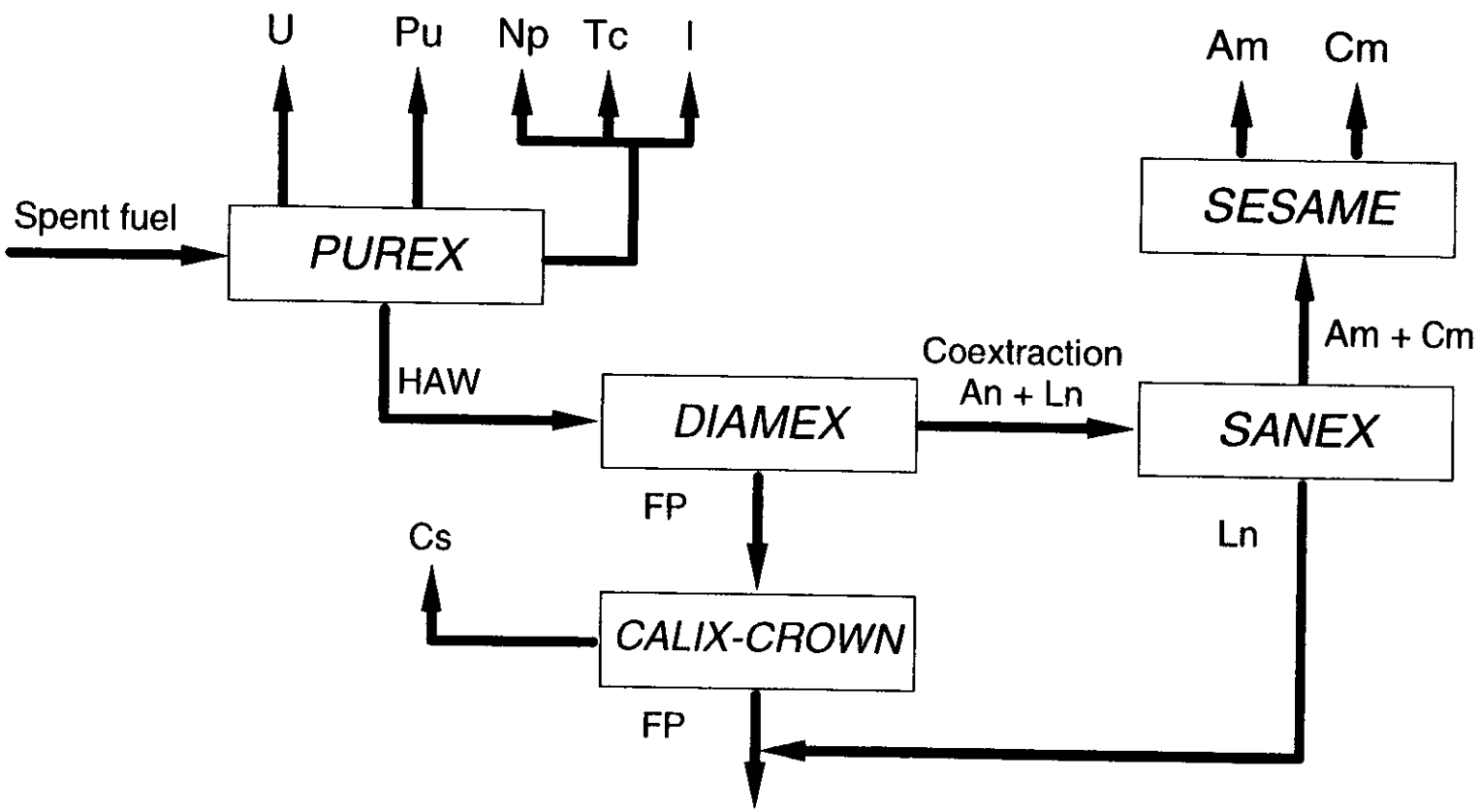


Flow sheet of pyrometallurgical process for separation of the minor actinides (1/2)



Flow sheet of pyrometallurgical process for separation of the minor actinides (2/2)

The Reference Flowsheet for the French SPIN Program



The role of pyrochemistry

- 4 major scenarios have been investigated :
  - use of « standard » critical reactors and use of homogeneous recycling (i.e. MA mixed within standard fuel in the core),
  - use of « standard » critical reactors and use of heterogeneous recycling (i.e. MA or LLFP targets in specific core/blanket locations),
  - use of dedicated reactors (critical or sub-critical) in a « double strata » approach of the fuel cycle,
  - finally, in the case of hybrids (ADS), molten salts fuel has also been considered in the studies.

For all these cases, the question is : why pyrochemistry ? What are the possible motivations for it ?

1. Homogeneous recycling of MA in a solid fuel

- Both PWRs and FRs have been considered.
- If one mixes MA with Pu (or U ?), consequences on the fuel cycle (doses, radiation sources etc) are minimized.
- An example : the IFR concept developed at ANL (USA). Interest of the concept : compactness, high efficiency, reduced delay in reprocessing, etc ...
- Main feature : a metal fuel is developed, which contains Pu and all MA (with some lanthanides), to be put in a very effective FR (safety, irradiation length etc). Transmutation in an "intrinsec" feature (a condition : high recovery of Am, Cm, etc ...).
- An alternative concept, based on oxide fuels, developed at RIIAR-Dimitrovgrad (DOVITA process).
- Also : work done at CRIEPI-Japan (and now at TUI-Karlsruhe).

2. Heterogeneous recycling of MA and LLFP

- MA targets (on inert support) with e.g. 20 % Heavy Isotopes fraction
  Targets → special subassemblies.
- Am and Cm should be handled.
- As far as recycling mode :
  - Multirecycling.
  - Monorecycling (« once-through »), if 90 ÷ 95 % cumulative fission rate is feasible.
- A large fraction (> 30 %) of the nuclear power park should be equipped.
- A large mass flow is foreseen (2 to 10 tons/year).
- Large impact on fuel cycle (fabrication, reprocessing).
- Interest of pyrochemistry : targets (with support) may be difficult to dissolve via PUREX - Also : highly active fuel to be dealt with (γ, α, etc).

3. Double strata scheme and dedicated reactors

- In the « second » stratum, dedicated critical or subcritical reactors should be envisaged, with innovative fuels (MA-based).
- A limited part (< 10 %) of the power park can be equipped with these reactors.
- Mass flow : ~ 5 ÷ 10 tons/y of MA (~ 2 t of Cm), with some adjunction of Pu (or enriched U).
- Strong impact on the dedicated fuel cycle.
- Pyrochemistry interest for handling innovative and very active fuels - Also : possibility to envisage a closed integral full cycle in the second stratum.
- An example : a pyrochemistry process based on nitride fuels is being investigated at JAERI - Japan.

4. Homogeneous Actinide and LLFP recycling in a (subcritical) molten salts reactor

Apt to "on-line" treatment (fission products elimination).

Low decontamination factors, very high irradiation times (burn-up).

To be foreseen as an option for the double strata concepts and mostly as a subcritical reactor (hybrid).

The back-end of the Nuclear Fuel Cycle

2 - Accelerator Driven Systems (ADS), physical principles and role for radioactive waste transmutation

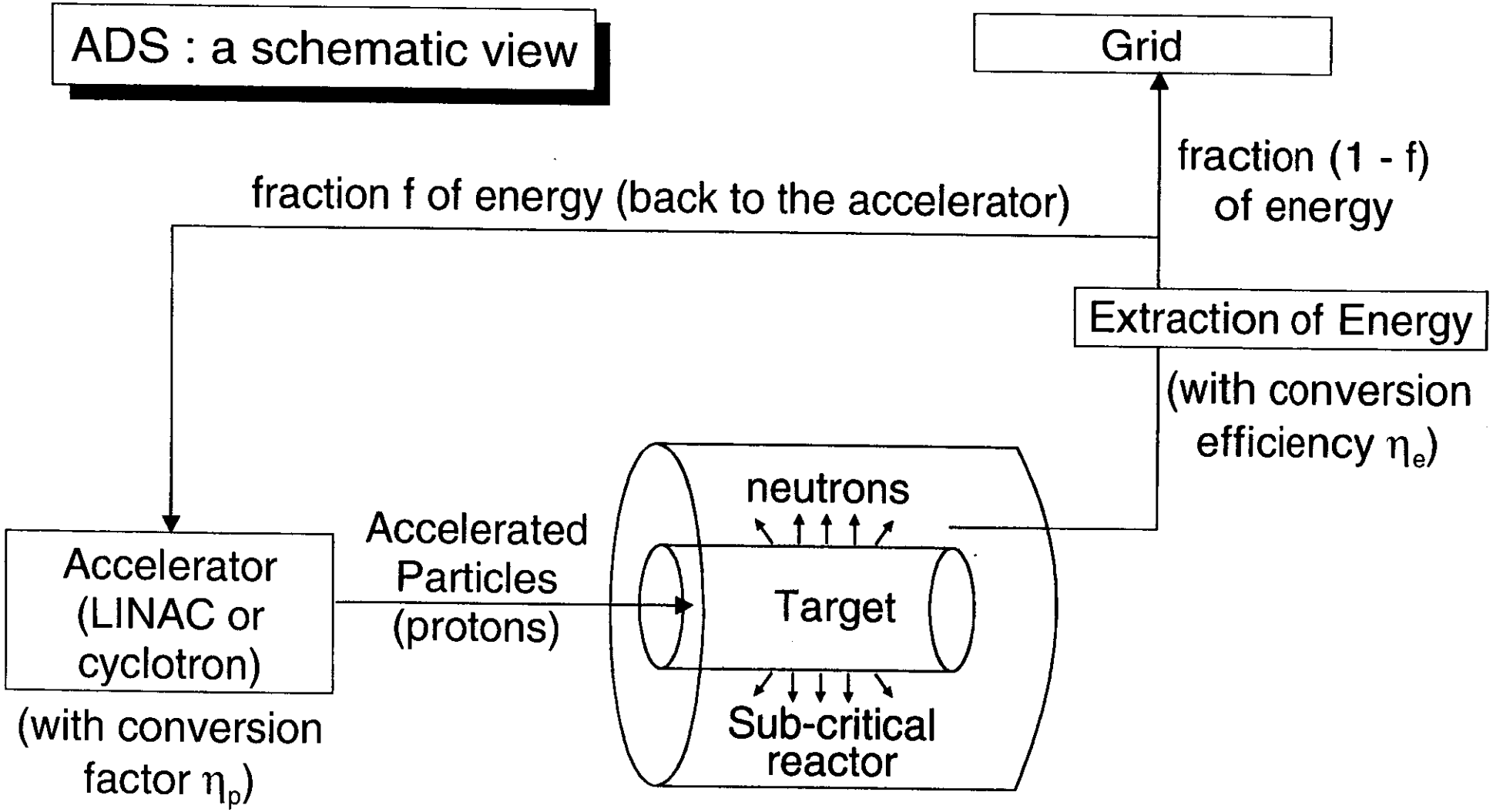
M. Salvatores

CEA/DRN/Cadarache

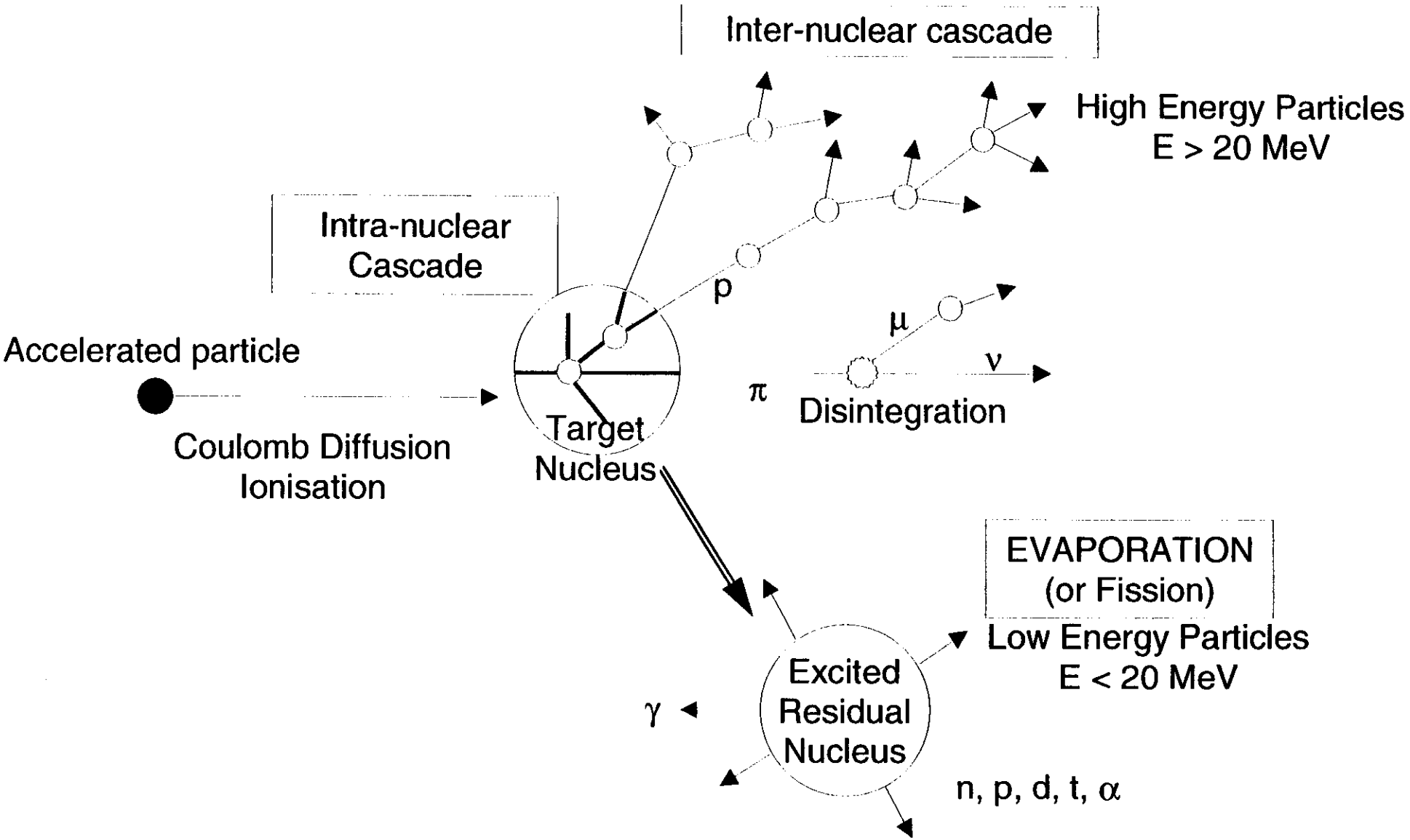
- Some generic features of accelerator-driven systems (ADS).
- Neutronics of a subcritical reactor.

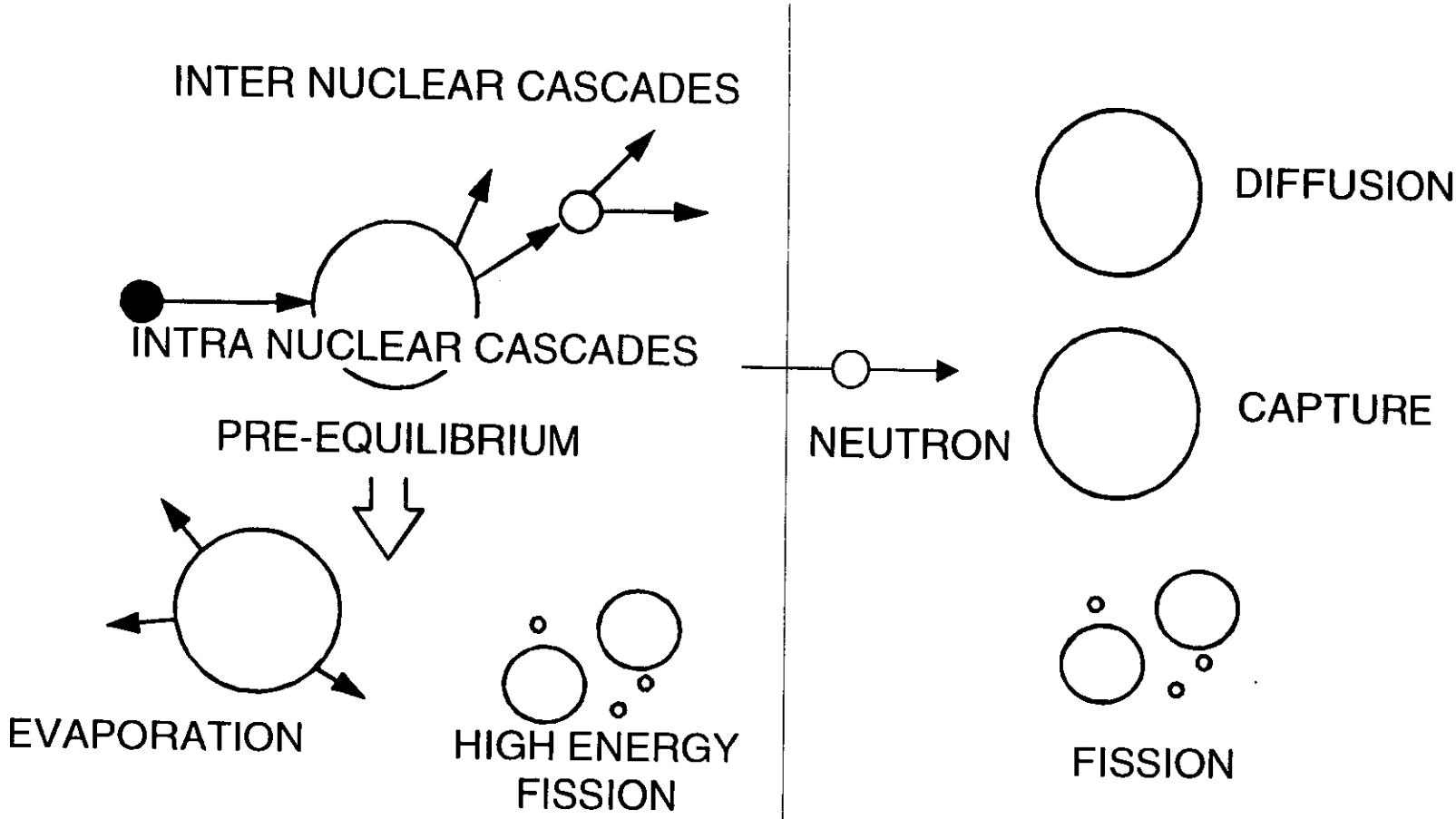
  A few examples
- The physics of transmutation and role of ADS.
- Uncertainty analysis and data needs.

ADS : a schematic view



Spallation Reaction Scheme

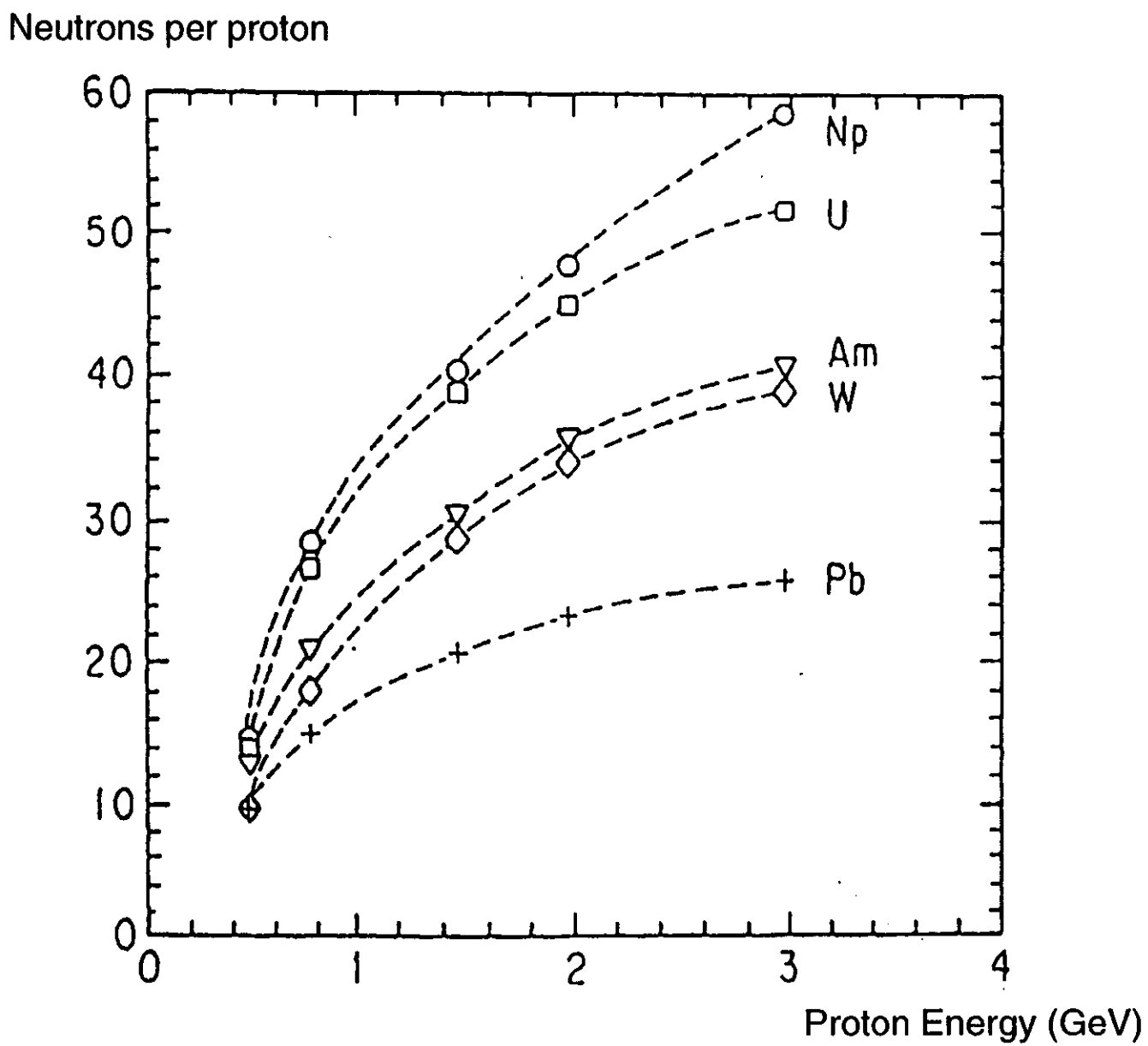




CUT-OFF ENERGY
15 ~ 20 MeV

High energy Particle Transport Codes
(HETC, etc ...)

Neutron transport codes
(deterministic S_N or Monte-Carlo)


Neutrons per proton
60
50
40
30
20
10
0
0
1
2
3
4
Np
U
Am
W
Pb
Proton Energy (GeV)

$p + Pb_{nat}$ (thin target)

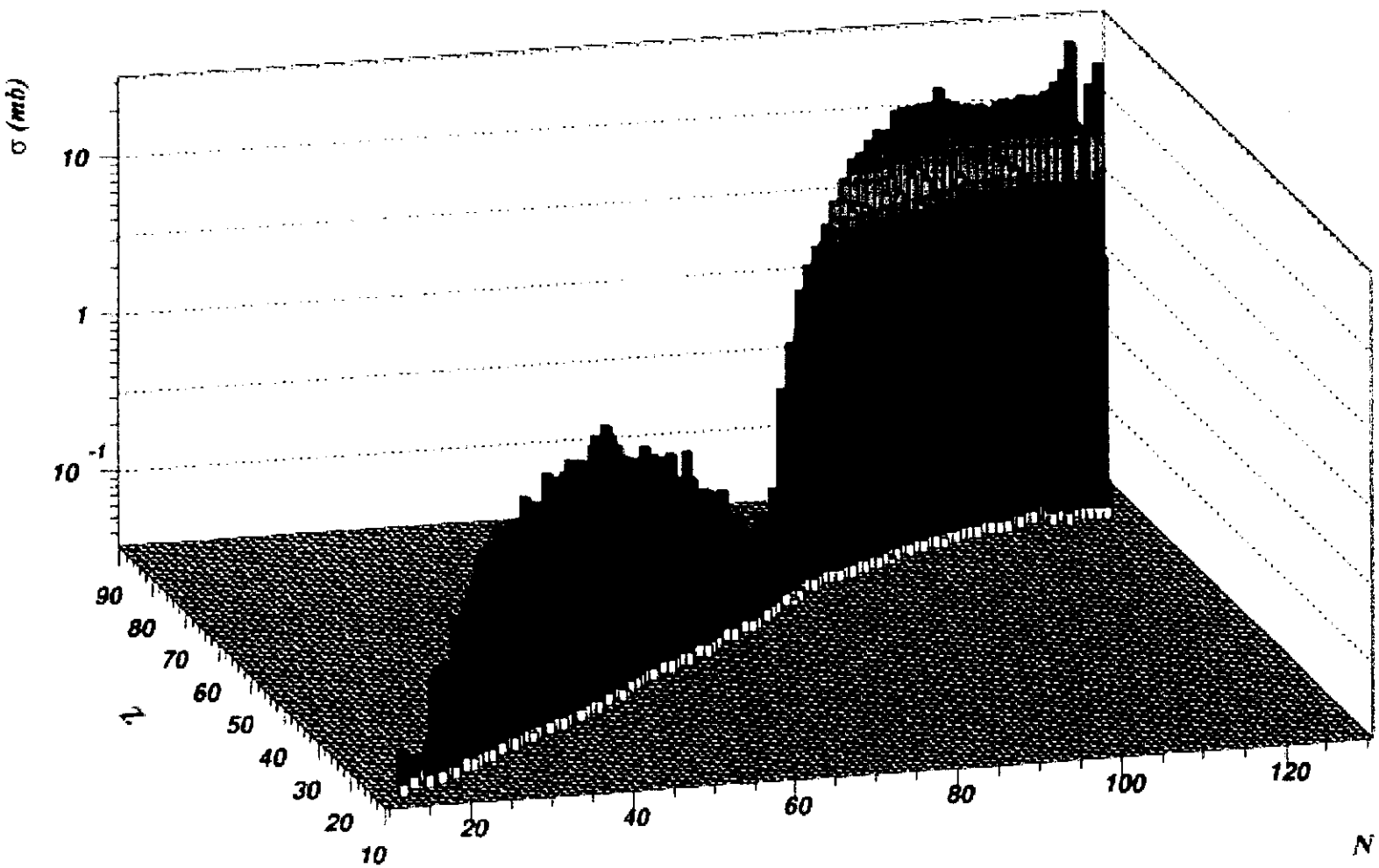
Number of particles per incident proton				
E_p (MeV)	500	1000	2000	3000
proton	2.1	4.0	7.4	10.2
neutron	*13.0*	*18.8*	*26.0*	*31.1*
deuton	0.1	0.5	1.3	2.1
triton	0.04	0.1	0.4	0.6
hélium 3	0.001	0.01	0.07	0.1
alpha	0.09	0.3	0.6	0.8
$\pi^{+,0,-}$	0.1	0.5	1.1	1.7

$p + U_{nat}$ (thin target)

Number of particles per incident proton				
E_p (MeV)	500	1000	2000	3000
proton	1.4	2.9	6.3	9.4
neutron	*18.0*	*24.9*	*32.7*	*38.0*
deuton	0.06	0.3	1.1	2.0
triton	0.03	0.1	0.4	0.6
hélium 3	0.0004	0.007	0.05	0.1
alpha	0.06	0.2	0.6	0.8
$\pi^{+,0,-}$	0.1	0.5	1.1	1.7

Spallation : residual nuclei

800 MeV p+^{208}Pb



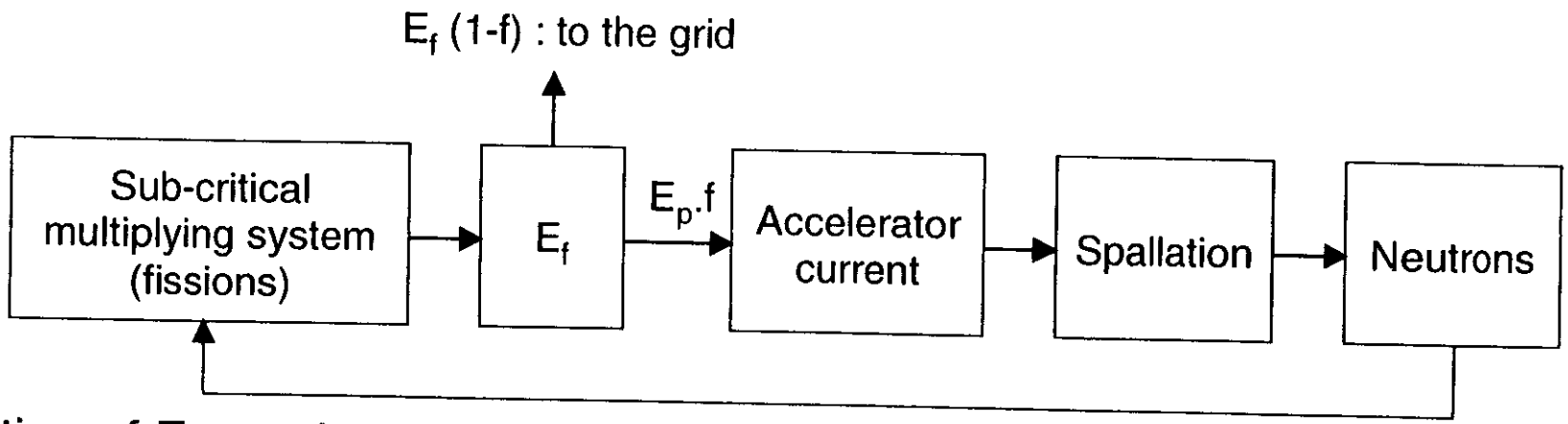
From an heuristic point of view, in a sub-critical system ($K_{eff} < 1$), the condition to have a stationary system can be written as follows :

$$S_{ext} + \nu = \frac{\nu}{K_{eff}}$$

(ν : prompt neutrons/fission)

For an ADS : $S_{ext} = \Gamma.f$

where Γ is the number of neutrons which come back to the sub-critical core if <u>all</u> the fission energy E_f is transformed into proton current :



f : fraction of E_f used to feed the accelerator.

To evaluate Γ :

If $E_p \simeq 1 \div 1.5$ GeV and for targets such that :

Z = neutrons/proton $\simeq 20 \div 30$ (case of a Pb thick target)

$$\Gamma = Z\, \eta_e\, \eta_p\, \frac{E_f}{E_p} \Rightarrow 1 \div 1.2 \text{ neutrons/fission}$$

($\eta e \simeq 0.4, \eta p \simeq 0.5$)

With this value for Γ, one can gain some insight on f :

$$\Gamma f + \nu = \frac{\nu}{K_{eff}} \Rightarrow f = \frac{\nu}{\Gamma}\frac{1-K_{eff}}{K_{eff}}$$

If $\nu \simeq 2.8 \div 3$ $\Gamma = 1 \div 1.2$ $K_{eff} = 0.95$

$$\boxed{f \simeq 10 \div 15\ \%}$$

Some consequences :

- If all the energy produced by the sub-critical reactor is used to feed the accelerator, one can be very sub-critical (e.g. $K_{eff} \simeq 0.7$).
- Since the external source $S_{ext} = \Gamma f$ and Γ has a value which is fixed by E_p and the type of target, S_{ext} will be increasingly relevant with respect to ν ($\simeq 2.5 \div 3.0$) as the system will be sub-critical ($f \Rightarrow 1$, $S_{ext}^{max} \Rightarrow 1 \div 1.2$).
- If $f \simeq 10 \div 20\ \%$, the contribution of the external source is of ≈ 0.2 neutrons/fission.

As for the current i_p the accelerator :

$$i_p = \frac{\Gamma.f}{Z} W \frac{1}{E_f} \Rightarrow \qquad i_p \simeq 20 \div 50 \text{ mA/GWth}$$

$$(f = 0.1 \div 0.2,\ Z = 20 \div 30 \text{ n/p})$$

$\Rightarrow$ i_p will be increasingly small, when the system will be closer to criticality and when the power of the sub-critical reactor will be low.

Example : $K_{eff} = 0.95$, $W = 300$ MWth $\Rightarrow i_p \simeq 5$ mA

A few consideration on safety and ADS role

Three major criteria have to be respected (for standard fission reactors) :

- Reactivity Control
- Decay heat removal capability
- Radioactivity confinement

Reactivity control

With ~ 10 $ of subcriticality margins, one should be able to avoid (?) a global reactivity accident. However, as for a critical system, if the core is not in its most reactive configuration, re-criticality after a significant core melting cannot be a-priori excluded.

Decay heat

Some problems are faced in a critical or sub-critical system (it depends only on the power density of the installation).

Radioactivity confinement

There is a need to provide a containment, i.e. the "third" barrier, which includes the primary circuits.

THE NEUTRONICS OF A SUBCRITICAL SOURCE-DRIVEN SYSTEM

Two major approaches for the actual neutronics calculations :

- Montecarlo Simulation (e.g. from high energy reactions down to neutron slowing down).
- Deterministic Methods (still widely used as reference for critical fission reactors).

For illustration purposes, we will use the Boltzman transport equation, without specification of the method of solution.

The neutron flux distribution in the subcritical core is the solution of the inhomogeneous Boltzman equation representing the neutron balance equation :

$$A\phi = M\phi + S$$

where A and M are, respectively, the net neutron loss and fission production operators and S is the external neutron source.

In the case of an ADS S is given by :

$$S = \Gamma f W g(r,E)$$ (g being a normalized energy and space distribution)

Moreover, in a non-conservative system, one can define a meaningful adjoint function ϕ^*, such that :

$$< \phi^*, (A - M)\, \phi > = < (A - M)^*\, \phi^*, \phi >$$

If one characterizes the subcriticality by a $K_{eff} < 1$, one has for the adjoint flux :

$$A^*\, \phi^* = \frac{1}{K_{eff}}\, M^*\, \phi^*$$

Previously, we have given the steady state condition in the following form :

$$\Gamma + \nu = \frac{\nu}{K_{eff}}$$

In reality, it can be shown that the above equation should be modified, to take into account the relative « importance » of the source neutrons with respect to fission neutrons :

$$\Gamma\, \varphi^* + \bar{\nu} = \frac{\bar{\nu}}{K_{eff}}$$

In fact combining the previous equations for ϕ et ϕ^*, one gets :

$$\frac{1}{K_{eff}} = 1 + \frac{\langle \phi^*, S \rangle}{\langle \phi^*\, M\phi \rangle} = 1 + \frac{\Gamma f}{\bar{\nu}} \frac{\phi_S^*}{\phi_F^*} = 1 + \frac{\Gamma f\, \varphi^*}{\bar{\nu}}$$

where

$$\varphi^* = \phi_S^* / \phi_F^*$$

is the ratio of the average importance of external source neutrons to the average importance of fission neutrons, and $\bar{\nu}$ is the averaged prompt neutron number.

φ^* can have values $\lesssim 1$, depending on the source geometry and neutron energy distribution.

As an example of its impact on the system economy, we consider the proton beam current i_p value :

$$i_p = \frac{f\Gamma}{Z}\frac{W}{E_f} = \frac{\bar{\nu}\left(\frac{1}{K_{eff}} - 1\right)}{\varphi^* Z}\frac{W}{E_f}$$

If, e.g., $\varphi^* = 1.2$, one need approximately 20 % less current to have the same performances (subcriticality, fraction f of energy needed etc).

Some examples of ADS neutronics

Whatever the fuel, the coolant etc, the specific features of the neutronics of the subcritical core of an ADS will be :

- The neutron spatial distribution, given by the inhomogeneous Boltzman equation. Example : if $K_{eff} << 1$, exponential behaviour.
- External neutron source energy distribution :
  - neutron spectrum variation in space (at least close to the target/core interface),
  - φ^* value which can have values different from unity,
  - high energy neutron propagation (shielding and material damage).
- Kinetic behaviour, as driven by the presence of much larger « delayed neutron effective fraction ».
- Also : because of the ambiguity of the notion of « reactivity » in a subcritical core, the most meaningful parameter to follow the time behaviour is the power level variation.

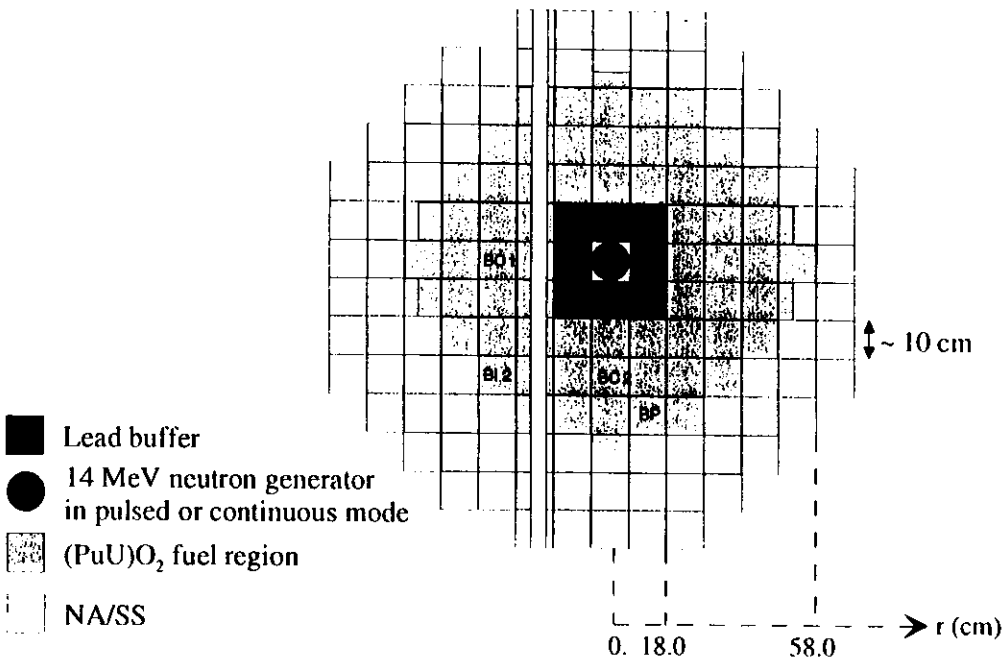
THE NEUTRON SPATIAL DISTRIBUTIONS

Examples will be taken from :

- An actual subcritical source driven experiment : the MUSE experiment at the CEA-Cadarache facility MASURCA (High accuracy measurement of the U-235 fission rate spatial distributions).

- A proposed demonstration experiment (the HADRON concept : ~ 100 MWt core, driven by a proton accelerator, providing ~ 2 ÷ 3 mA of 1 GeV protons on a liquid lead/bismuth target). Calculation of the power distributions.

The MUSE experiments at CEA-Cadarache



Midplane section of a typical MUSE core in the MASURCA reactor at CADARACHE for subcritical measurements in an external source driven configuration.

Subcritical range : Keff $\simeq$ 0.97 ÷ 0.99

Measured parameters :

- subcriticality
- flux distribution
- external neutron source importance
- neutron spectrum

Fuel type : $(PuU)O_2$ or $ThO_2 + UO_2$

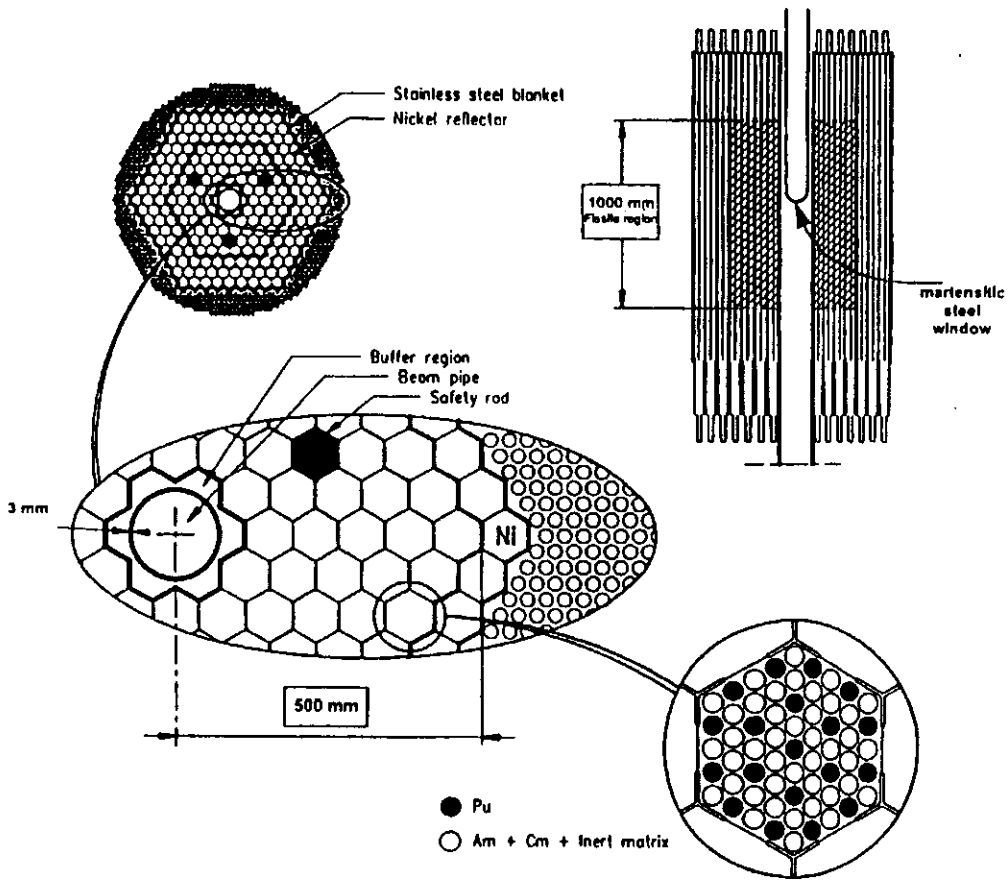
Simulated coolant : Na - In future : Pb

Source position : core center or axially displaced

Buffer type : Pb, Na, SS (for parametric studies)

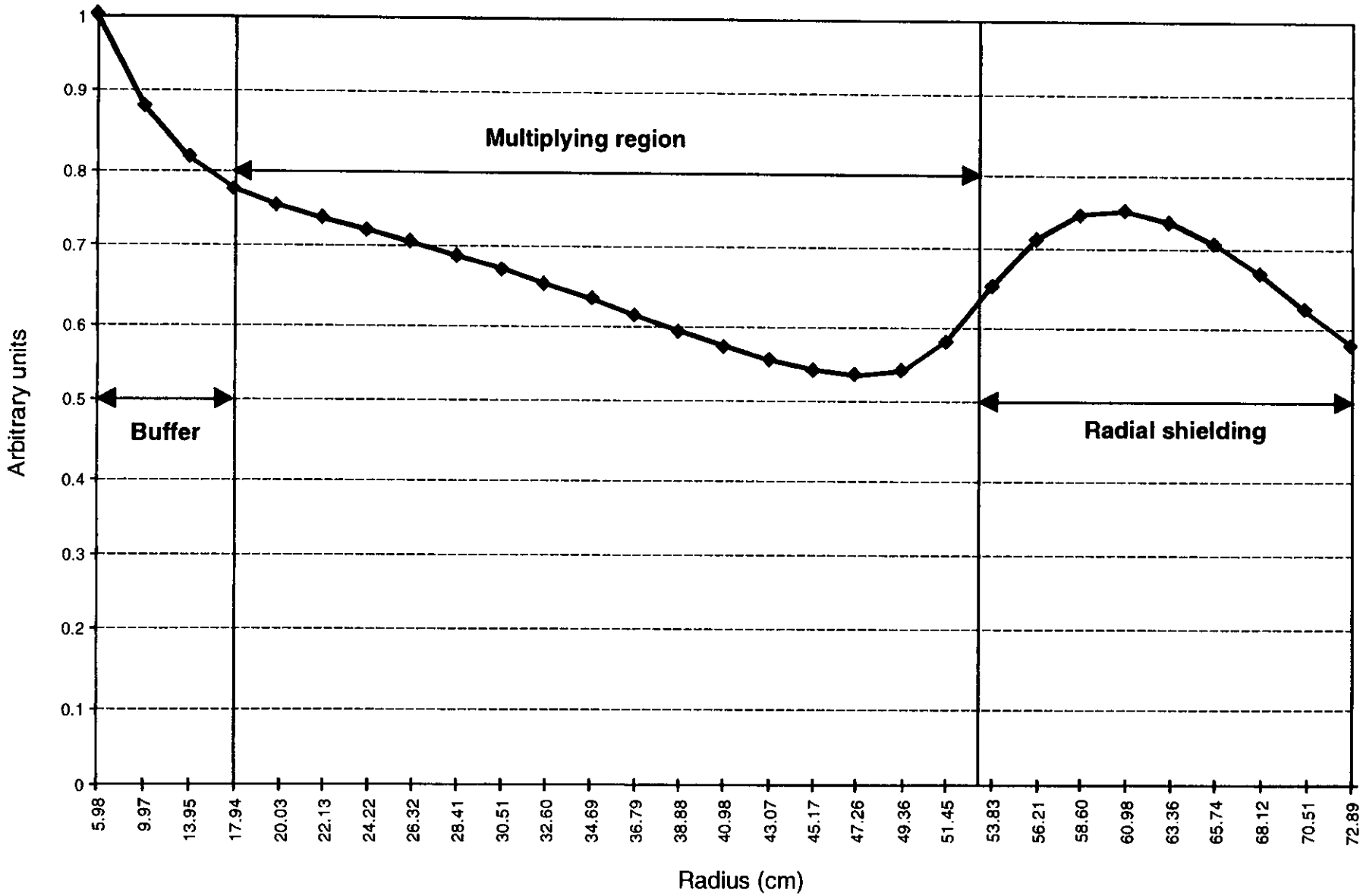
An example : The HADRON concept with Advanced Oxide Fuel

Fuel :	$[Pu+(Am+Cm)]O_2+MgO$
Fuel mass :	1480 kg (MOX)
Coolant :	Na / [Pb][(a)]
Target material :	Pb-Bi (in a steel container)
Keff :	0.95
Reactivity loss :	- 2 pcm/day
Power :	50 MWTh
Average flux :	$0.7\ 10^{15}$ n/cm^2.s
Beam current i :	1.5 mA (Ep = 1 GeV)
	(~ 25 µA/cm^2)
Target power :	1.2 MW
Matrix :	MgO (50% volume)
Enrichment :	24% Pu [21% Pu][(a)]
Core volume :	770 l

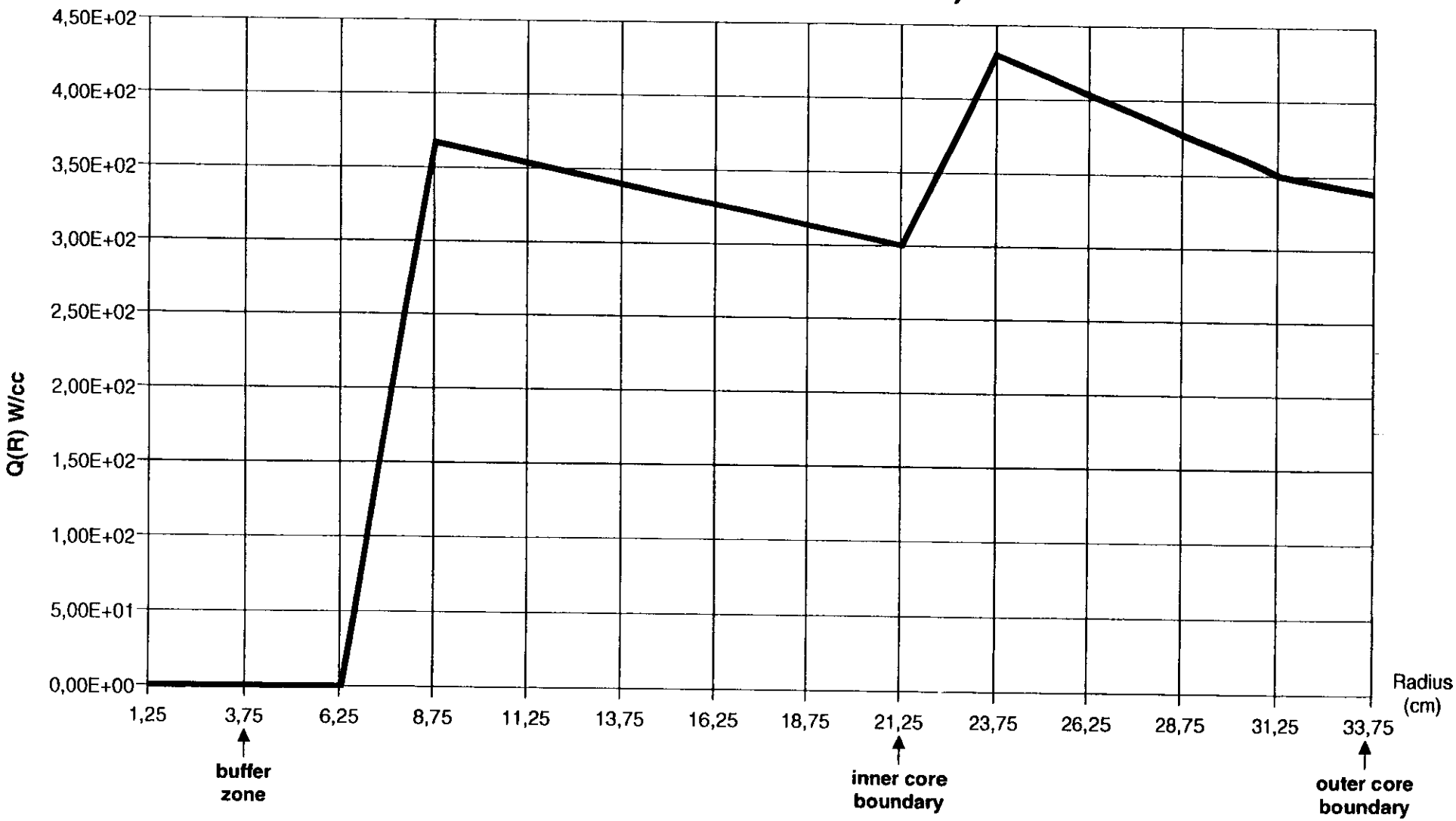


(a) Lead coolant variant

U-235 fission rate in the MUSE-3 configuration



Radial power distribution expected in a HADRON configuration
(2 radial enrichment zones)

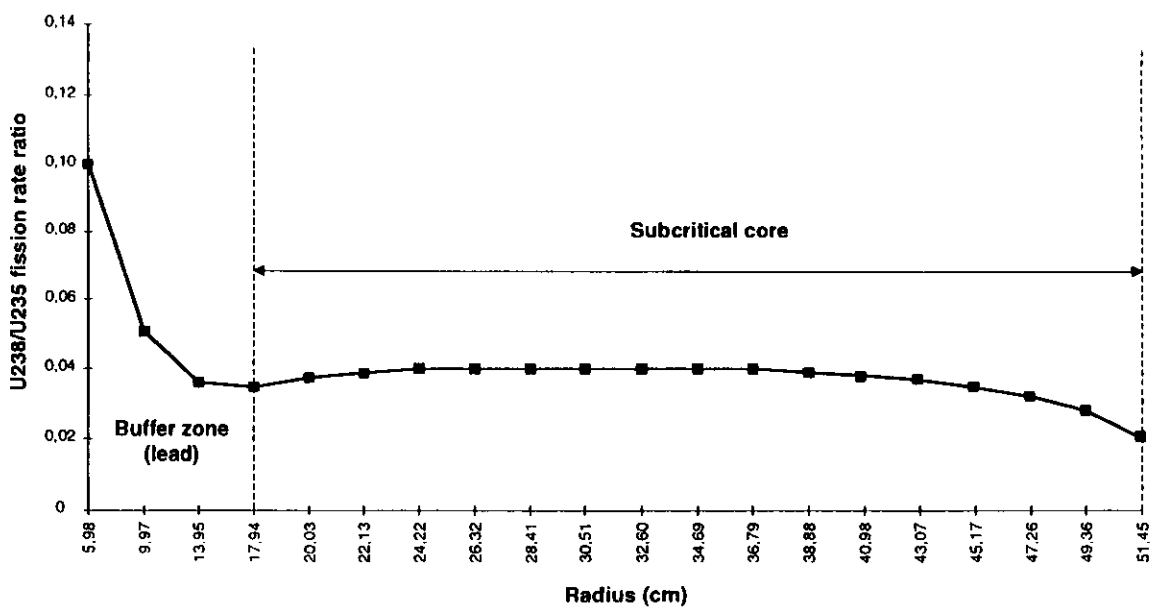


The neutron spectrum variation : from the core/target interface to the outer core boundary

- The observable : the ratio of the threshold U-238 fission rate to the U-235 fission rate (spectral index, i.e. the ratio of the integral of high energy neutrons to the).

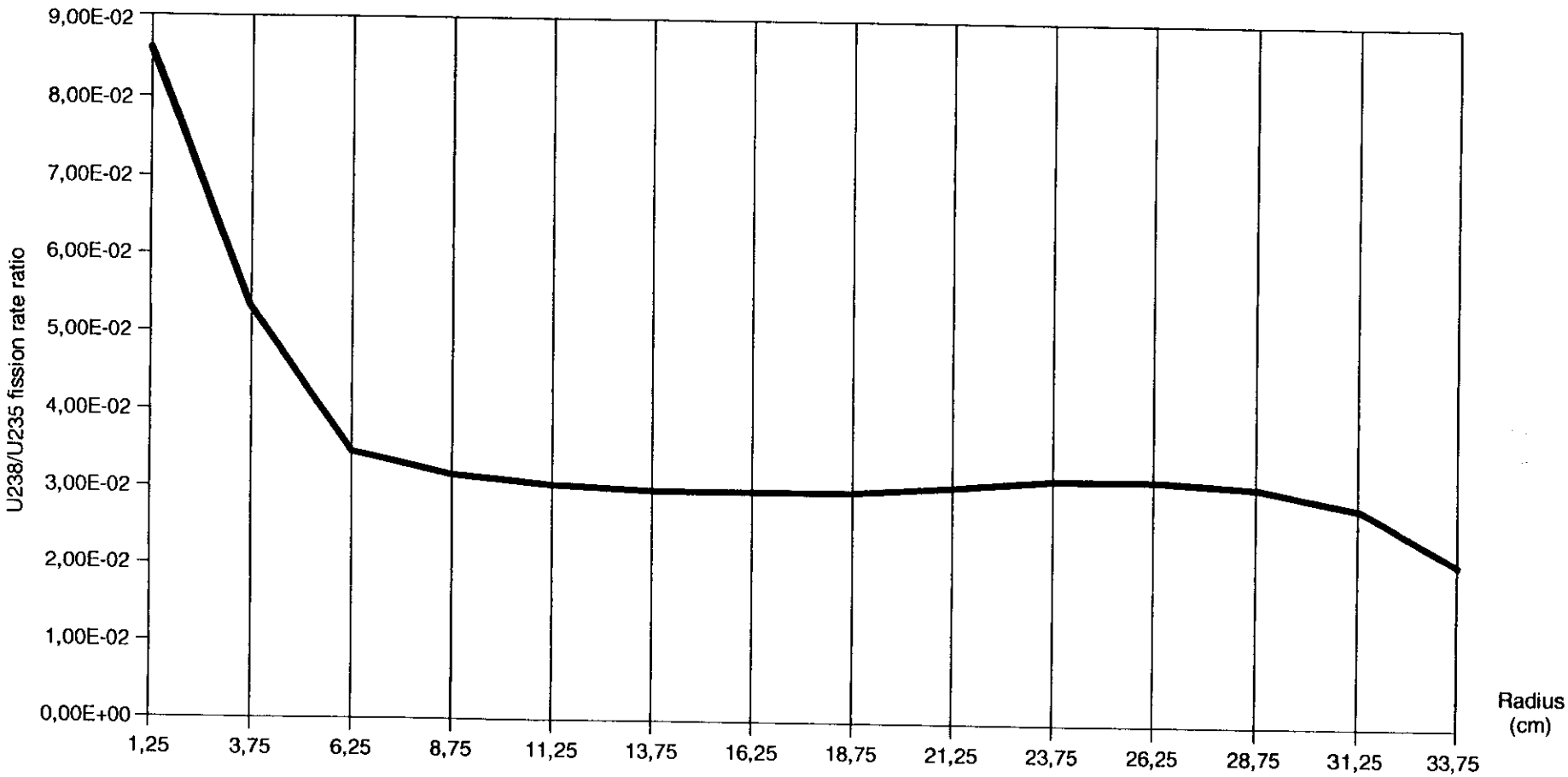
- In the MUSE experiment : (14 MeV neutron source)

MUSE3 Experiment : FU238/FU235 fission spectrum index in the multiplying region (radial distribution)



- In the HADRON configuration :

HADRON configuration : Radial distribution of the F U-238/F U-235 fission spectrum index



φ* : How important are the source neutrons when compared to fission neutrons (« external » versus « internal » neutrons)

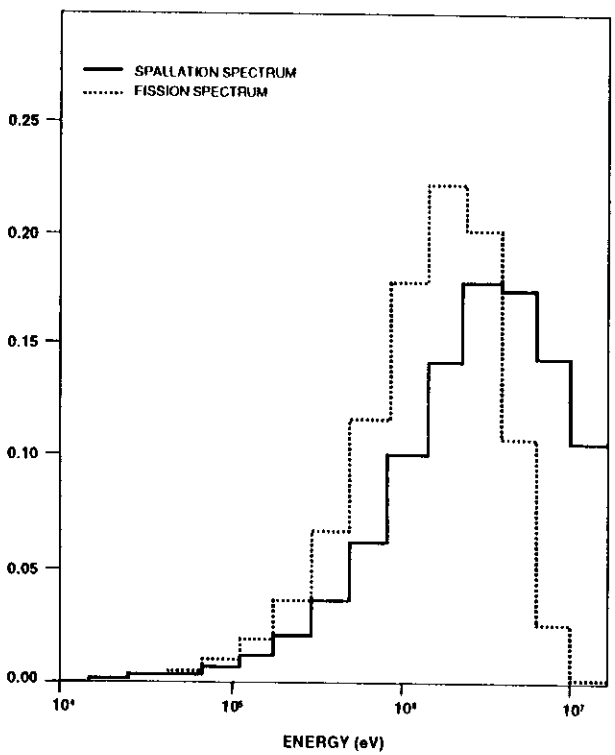
- φ* is an observable and can be measured with high accuracy
  Measurements performed at MASURCA (the MUSE experiment)
  Typical results :

Configuration	Calculated φ*	Measured φ*
Cf-252 source at core center	1.25	1.19 ± 4 %
Cf-252 source at core/blanket axial interface	0.91	0.90 ± 4 %

- φ* value calculated for the HADRON concept : φ* = 0.8

« Decoupling » of the spallation target physics (the external neutron source production) and the subcritical core neutronics (the source neutron propagation in the core) ⇒ How representative are the MUSE experiments

- The spallation neutron spectrum is « harder » than the pure fission neutron spectrum :

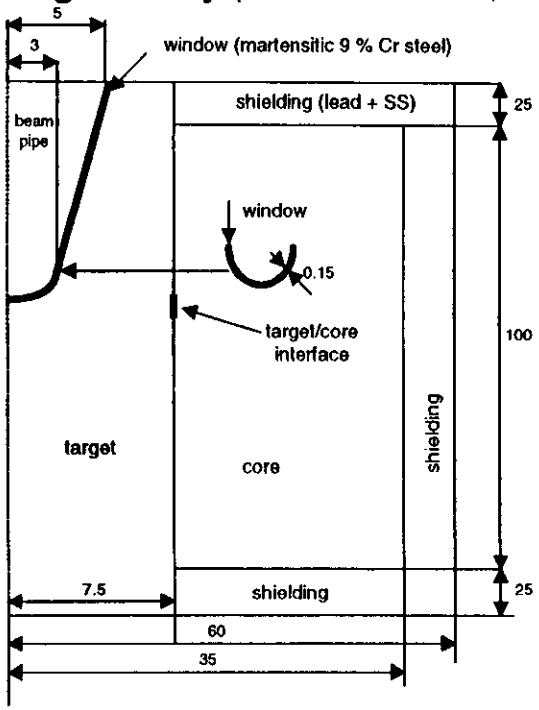


- But the source (external) neutrons, when entering the multiplying subcritical core, quickly (i.e. after ~ 1 mean free path), « forget » their origin and are assimilated to the « internal » (subcritical core) neutrons.

Neutron damage, gas production and the like

Calculations performed for the HADRON configuration, taken as example :

Schematic geometry (dimensions in cm, not to scale)



Displacement per atom in the window and at the target/core interface for high (E > 20 MeV) and low (E ≤ 20 MeV) energy particles :

Component	dpa/year :		
	E > 20 MeV	E ≤ 20 MeV	All energies
Window	~ 40	~ 60	~ 100
Target/core interface	~ 1.5	~ 50	~ 52

Hydrogen isotope production (H-D-T) and He production given in atomic ppm (appm) per dpa (appm/dpa) :

Component	H-D-T	He
Window	~ 900	~ 50
Target/core interface	~ 650	~ 15

Irradiation damages have two components :

- Displacements per atom (dpa) : in a material having received 100 dpa, each atom has been displaced, in average, 100 times from its equilibrium location in the lattice.
- Production of spallation residuals - He and hydrogen isotopes are of interest, since they can enhance the material swelling.

Neutron kinetics of sub-critical systems

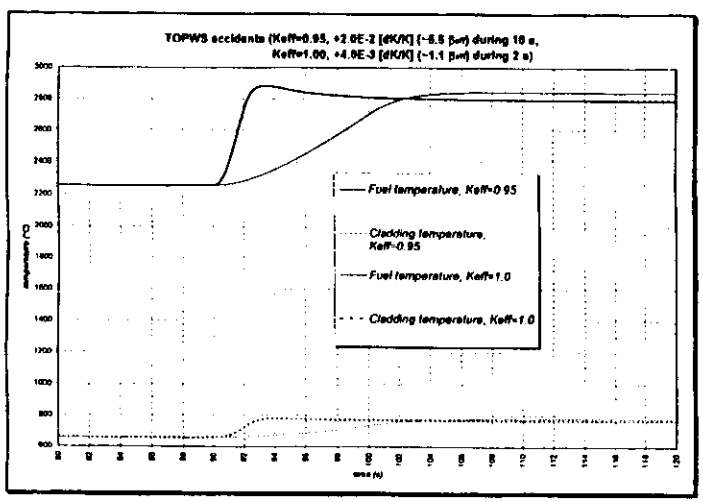
ANALYSIS OF ACCIDENTAL TRANSIENTS

Comparison Sub-critical (k_{eff} = 0.95) - Critical Systems "Phenix type"

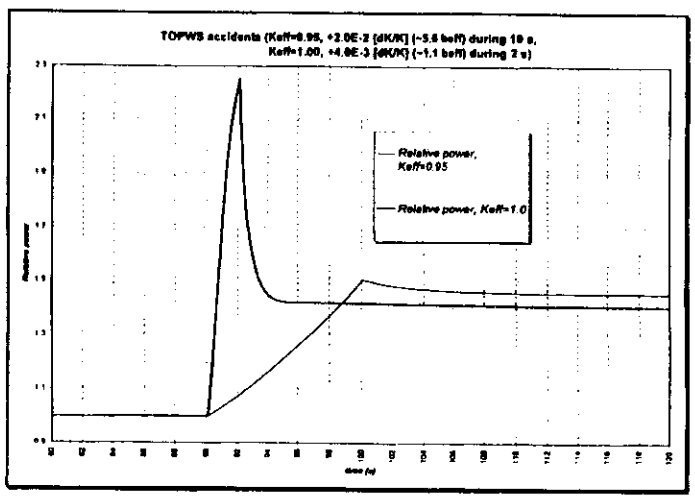
1 - Transient of Power without scram or source shutdown

Simulation with a reactivity ramp insertion at a rate of 0.55 $/s until fuel melting.

TOPWS accident



Temperature evolution during TOPWS accident in PHENIX type reactor with different k_{eff}



Power evolution during TOPWS accident in PHENIX type reactor with different k_{eff}

Result : No risk of prompt criticality type in the case of sub-critical reactor : The power and temperature changes are much slower than for a critical reactor (the power increase is limited as $P/P_0 = \rho_0/(\rho_0+\Delta\rho)$

	Fuel melting	$\Delta\rho$	P/P_0
Critical reactor	2 sec.	1.1 $	2.2
Sub-critical reactor	12 sec.	6.6 $	1.5

In a subcritical system, the coupling between reactivity and power is less strong than in a critical one.

In fact :

1 - The neutron population N (proportional to power) is given in steady state with source S by :

$$N \simeq -\frac{\Lambda}{\rho - \beta} S$$

where Λ is prompt neutron lifetime
β total delayed neutron fraction
ρ total initial reactivity

2 - If a $\Delta\rho$ reactivity is inserted $N \rightarrow N'$:

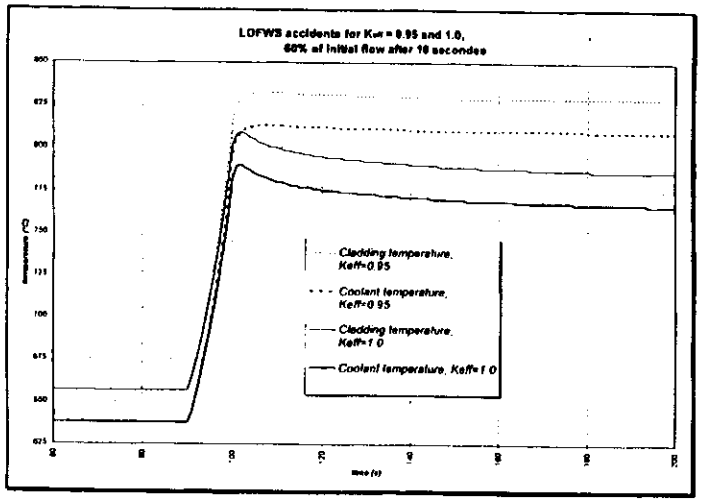
$$\frac{N'}{N} = 1 - \frac{\Delta\rho}{\rho + \Delta\rho - \beta}$$

If $\rho \rightarrow 0$ (critical system), reactivity insertion means power going "quickly" to zero. That is not the case for a subcritical system ($\rho \neq 0$ and large with respect to $\Delta\rho$).

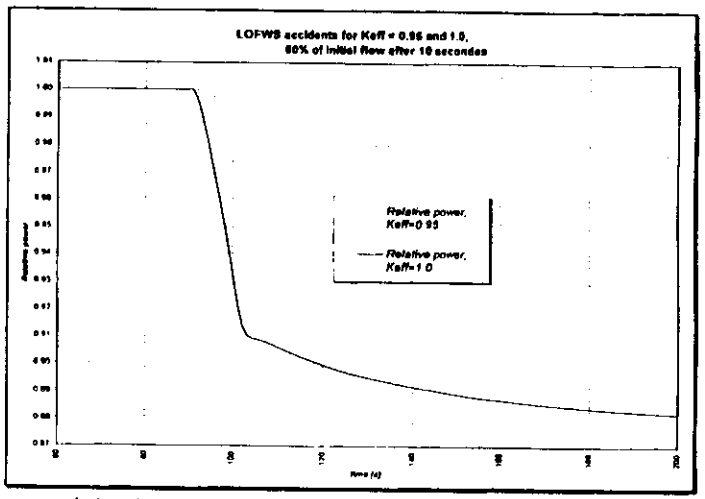
2 - Loss of Flow without scram or source shutdown

Simulation with primary flow decrease from 100 % to 60 % during 10 seconds until clad melting.

LOFWS accident



Temperature evolution during LOFWS accident in PHENIX type reactor with different k_{eff}



Power evolution during LOFWS accident in PHENIX type reactor with different k_{eff}

Result : The reactivity feedback effect is much less than for a critical reactor, the power decrease is smaller, hence a higher clad temperature.

3 - Conclusion

- Small impact of reactivity on power for the sub-critical reactor.
- Power change is slower than for a critical reactor.

Hence :

- Very important margin before fuel melting in the case of TOP.
- Less favourable behaviour regarding LOF (but possibility to stop easily the spallation source).

To validate the principles of an ADS :

A - Feasibility of high intensity accelerators. LINACs, cyclotrons ? Interface accelerator/target (window) : heat load, damages.

B - Spallation physics and high energy simulation validations :

- Z number of neutrons/proton in different target geometries.
- Energy and angle spectrum of emitted neutrons (how many neutrons above 20 MeV ? How many neutrons are back scattered ?).
- Distribution in mass and charge of the residual nuclei after spallation (how much induced activity/toxicity ?).

C - Target technology (heat removal, damages induced by neutrons with $E_n \geq 20$ MeV and enhanced by α, p etc production). An example : the MEGAPIE experiment.

D - Physics of sub-critical multiplying systems :

Since after $1 \div 2$ interactions, neutrons loose memory of the difference of energy distribution between fission and spallation, one can envisage to study the sub-critical system behaviour using a well known external source (e.g. : a Cf-252 source).

$\Rightarrow$ One can decouple the spallation neutron physics and the sub-critical system physics (the MUSE experiments).

The MEGAPIE Initiative

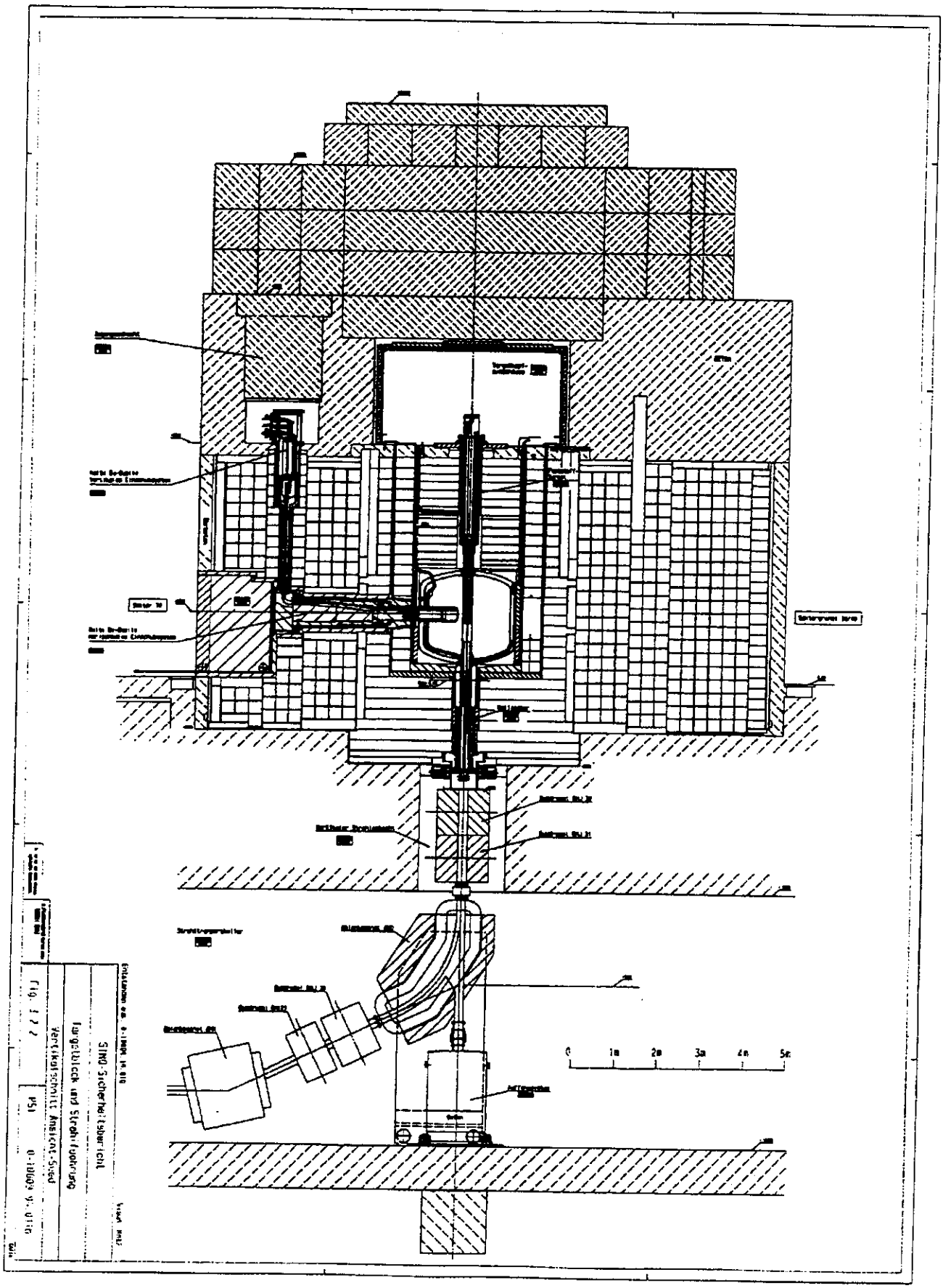
CEA-Cadarache, FZ Karlsruhe and PSI started an initiative to carry out a joint

MEGAwatt PIlot Experiment

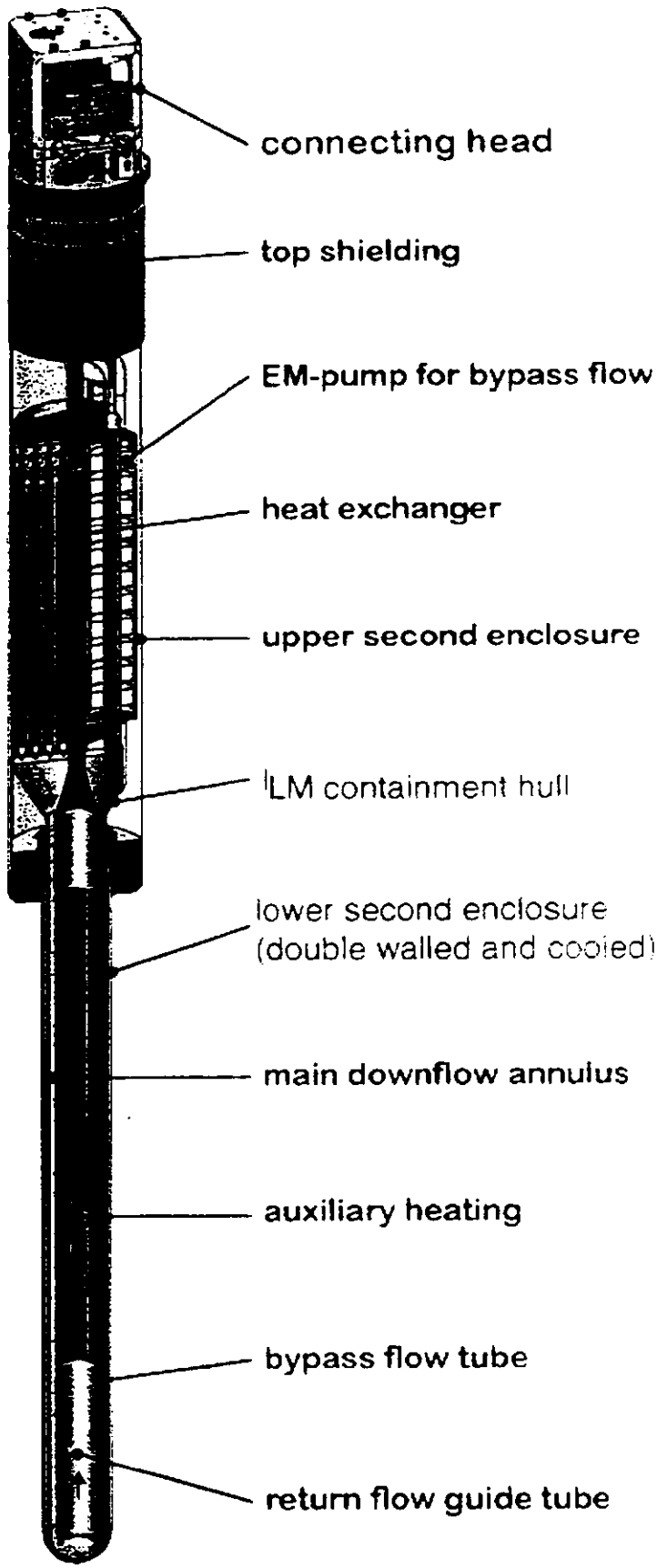
taking advantage of PSI's high intensity proton beam to demonstrate the feasibility of a liquid PbBi target at power levels relevant to ADS and to gain experience in designing, operating and disposing of such a target.

Goals, Objectives and Activities

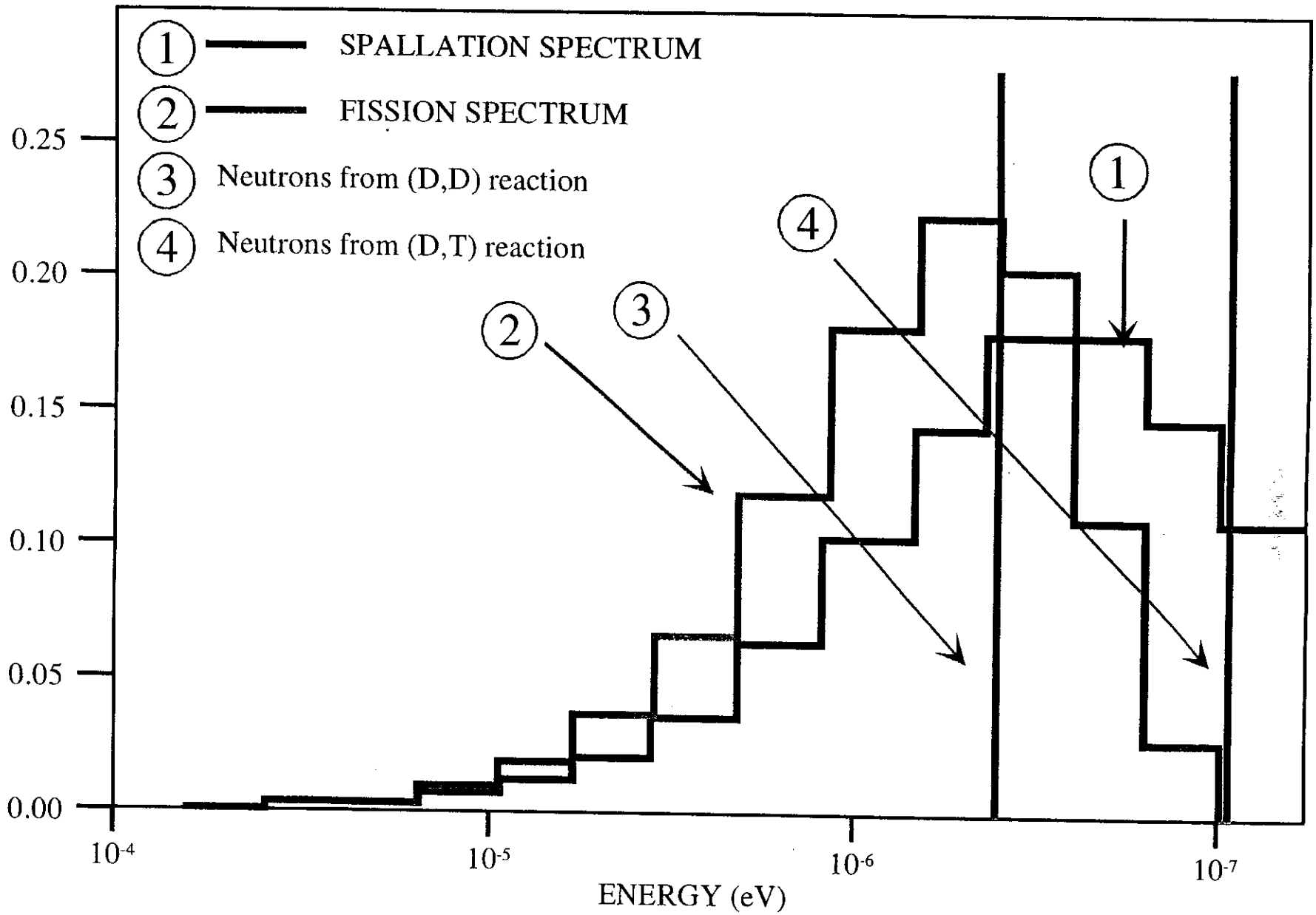
- Continue, intensify and co-ordinate ongoing and planned R&D efforts at participating institutions
- Have a well researched and instrumented PbBi-target ready to go in the SINQ target position in the beginning of the year 2004
- Operate the target at full SINQ beam power for 3-6 months in order to accumulate relevant integrated leads
- Monitor spallation products during operation and carry out post operation investigations to assess the service potential and desirable design changes for a prototype target

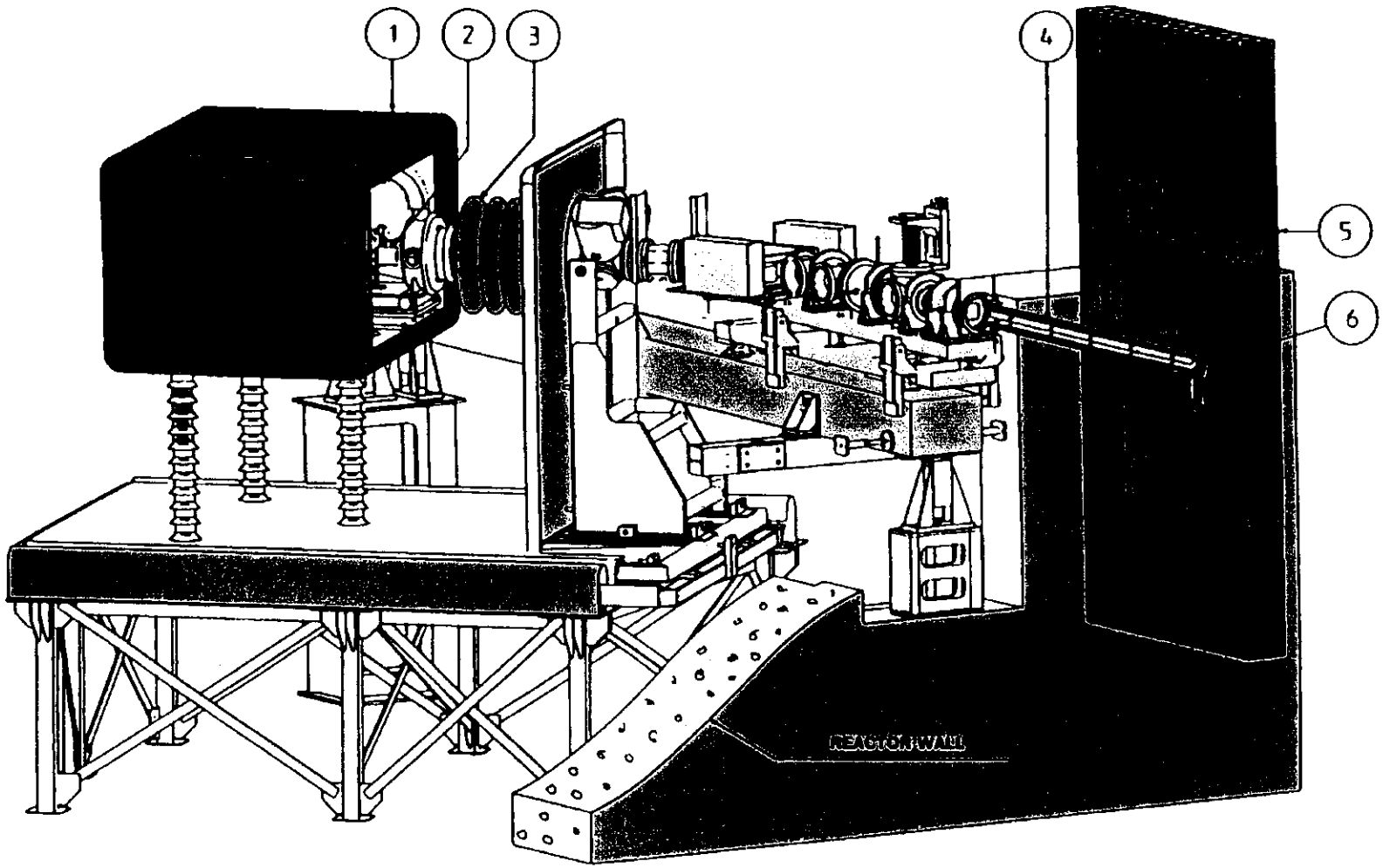

SINQ-Sicherheitsbericht
Übergabeblock und Strahlführung
Vertikalschnitt Ansicht-Süd
0 1m 2m 3m 4m 5m

MEGAPIE Liquid Metal Target for SINQ conceptual draft



Conceptual draft of the MEGAPIE Target for SINQ


1 SPALLATION SPECTRUM
2 FISSION SPECTRUM
3 Neutrons from (D,D) reaction
4 Neutrons from (D,T) reaction
0.25
0.20
0.15
0.10
0.05
0.00
10-4
10-5
10-6
10-7
ENERGY (eV)

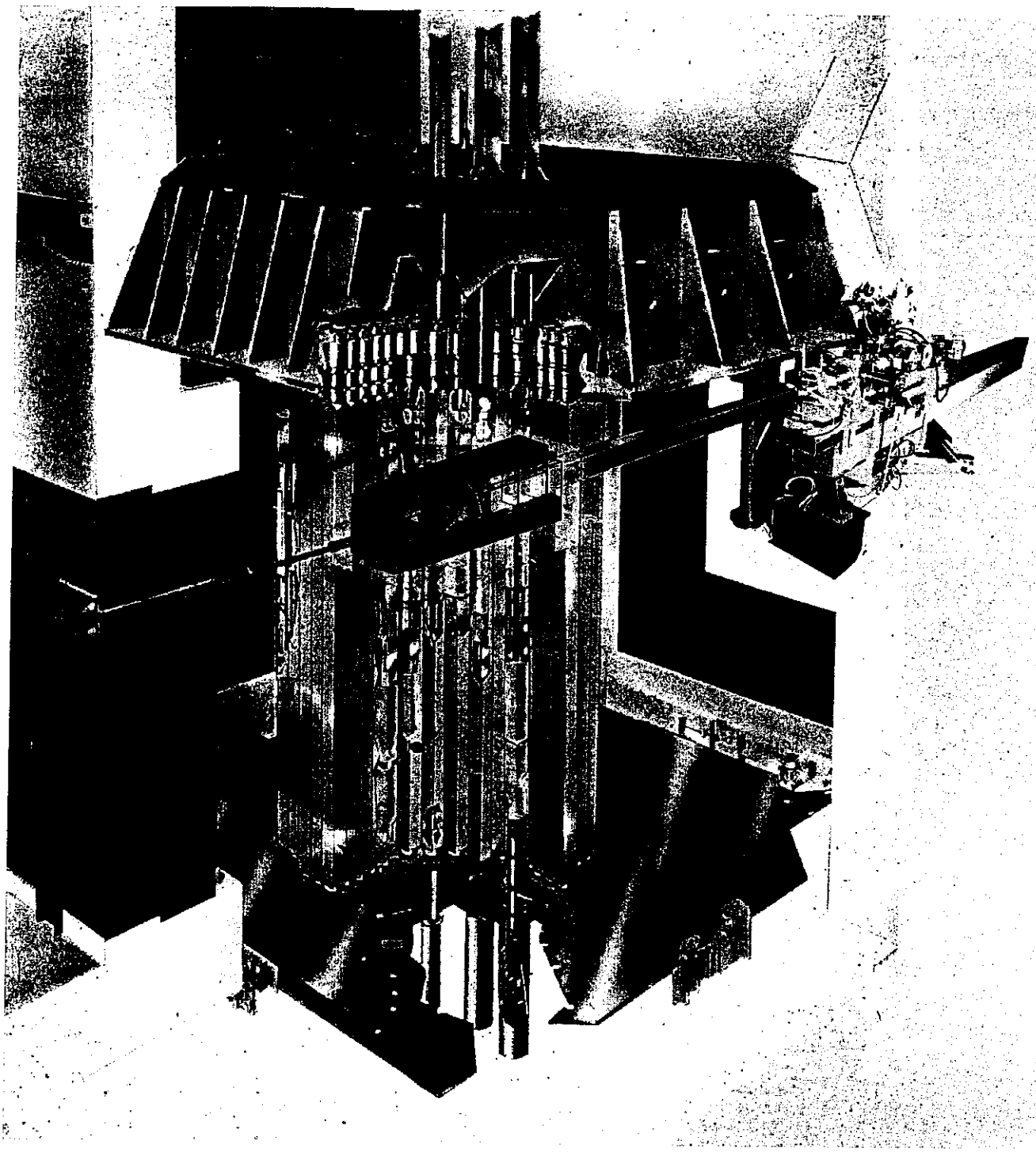


THE MASURCA facility with GENEPI neutron source

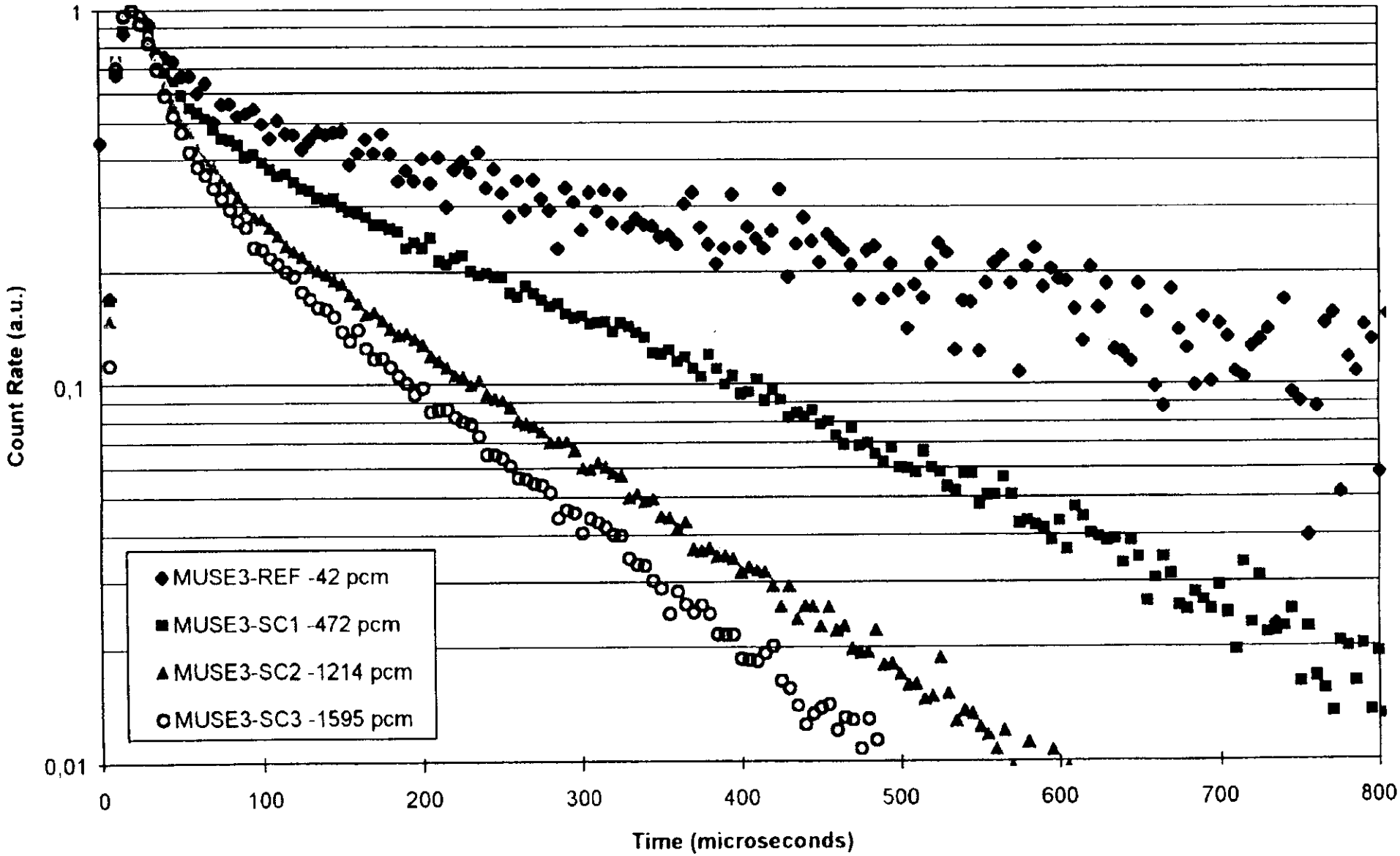
① : High Voltage : 250 kV - ② : Ions source
③ : Accelerator guide - ④ : Deutons beam guide with 6 internal quadrupoles
⑤ : MASURCA subassembly - ⑥ : Tritium or Deuterium target for neutrons production

MASURCA

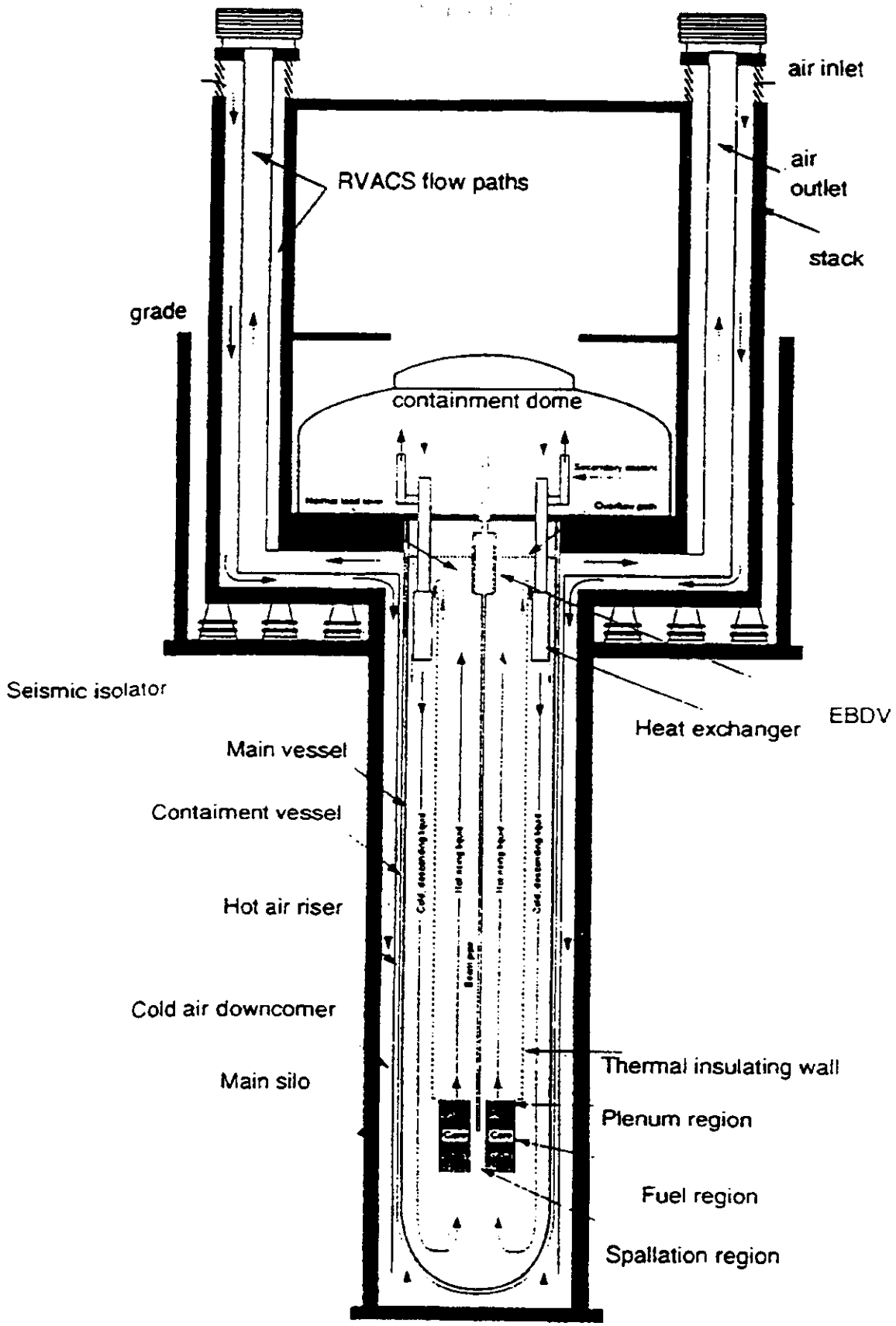
Programme Muse



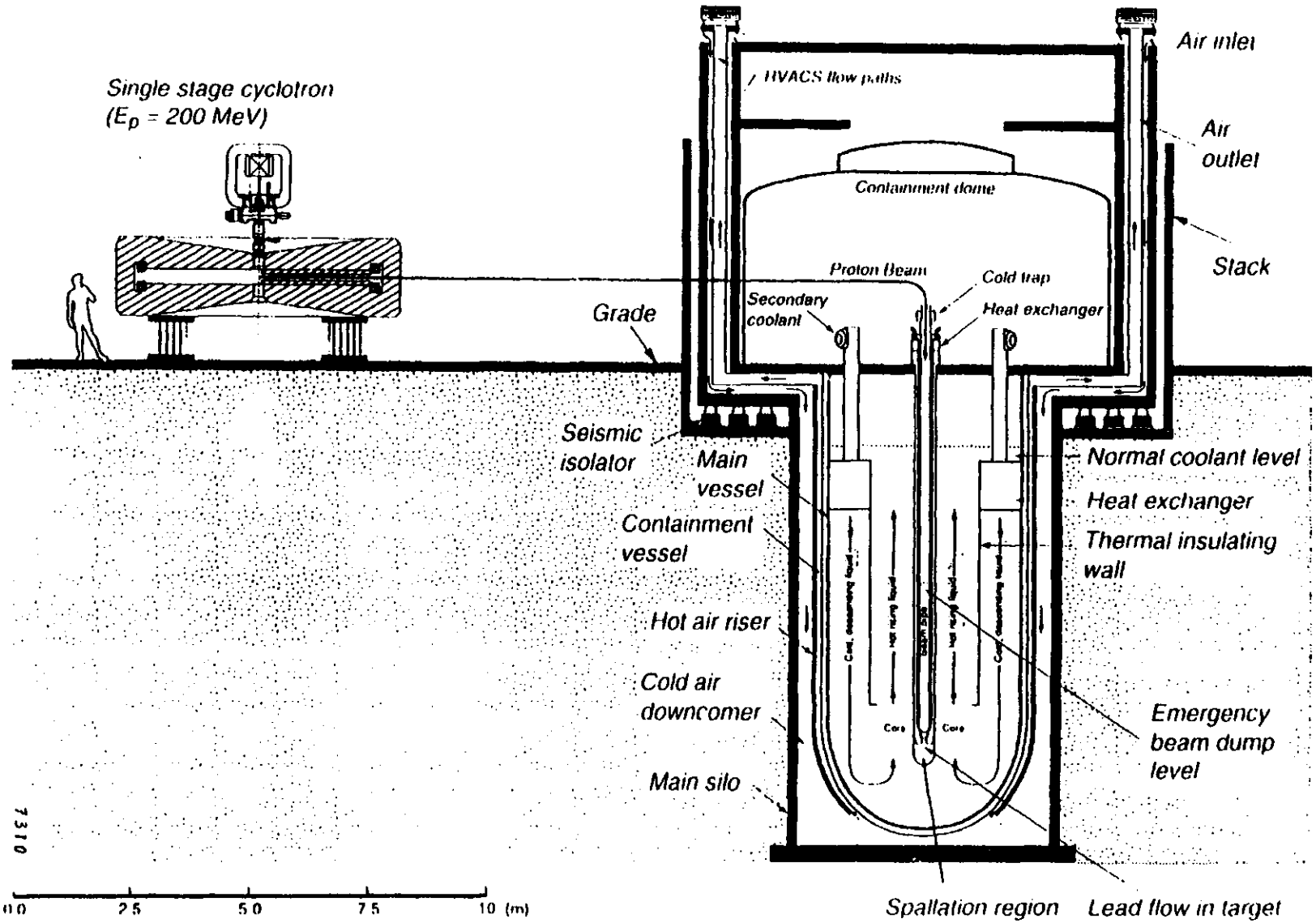
Dynamic Measurements MUSE2-REF, MUSE3-SC1, MUSE3-SC2, MUSE3-SC3

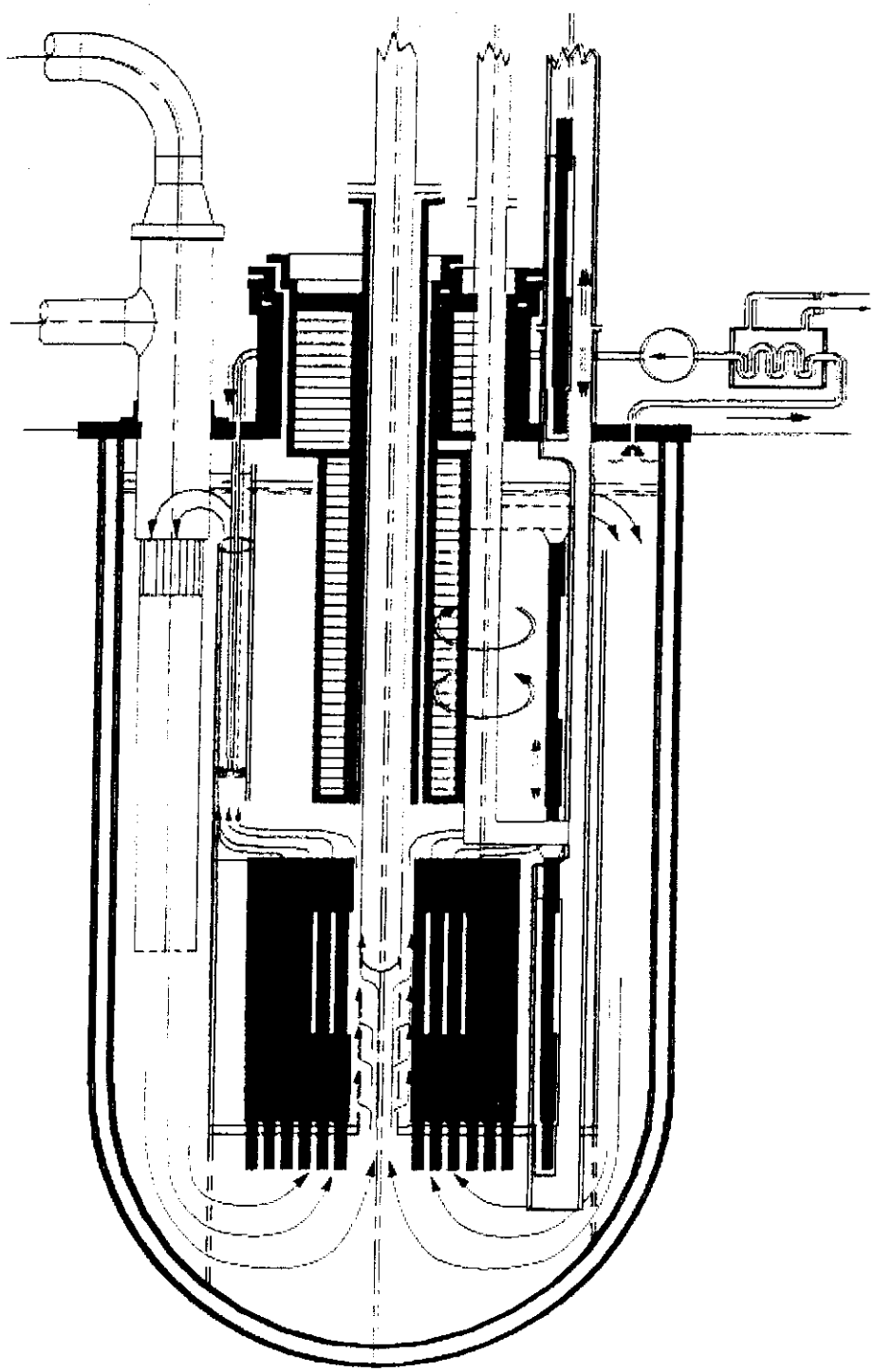


The « RUBBIATRON »



A « DEMO » for the RUBBIATRON





Accelerator Driven System
Scheme of the Reactor Assembly of a 80 MW Demonstration Facility

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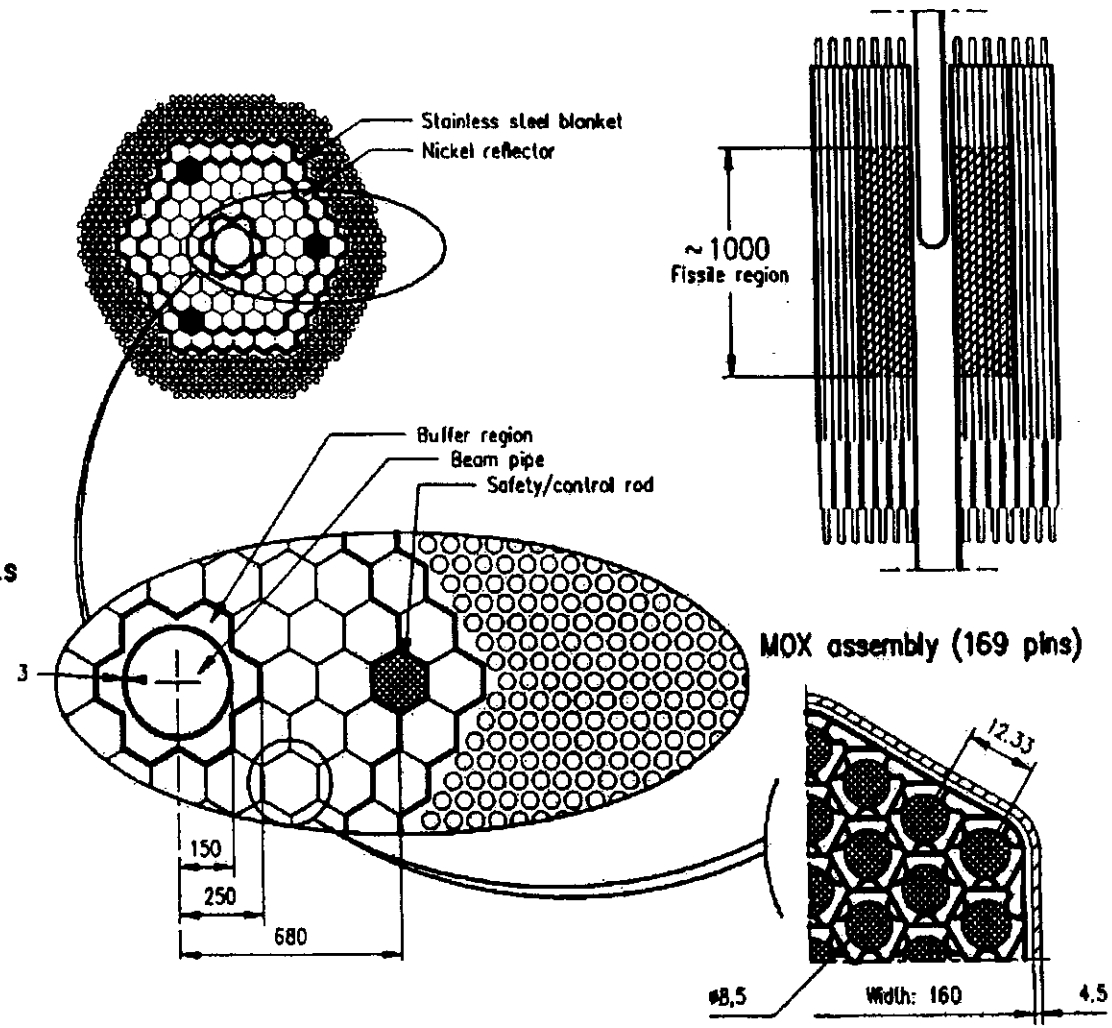
GAS–COOLED DEMONSTRATION MOX ADS

Fuel:	$(U,Pu)O_2$
Fuel mass:	3040 kg (U+Pu)
Coolant:	He
Target material:	Pb–Bi
Keff:	0,95
Reactivity loss:	–8 pcm/day
Power:	100 MWTh
Average flux (inner region):	1,1 10^{15} n/cm^2.s
Beam current i:	1,9 mA
Target power:	1,5 MW
Enrichment:	32% Pu/U+Pu
Core volume:	1270 l



The physics of transmutation and role of ADS

ADS can find a role in :

a - Energy production (widely discussed by C. Rubbia in the frame of the EA proposal).

b - Symbiosis with standard reactors (PWRs with UOX and MOX fuel, Fast Reactors with MOX fuel), in order to manage minor actinides (Np, Am, Cm) and selected Long Lived Fission Products (LLFP) in a separate « stratum » of the fuel cycle.

c - Symbiosis with standard reactors (PWRs with UOX fuel), in order to manage Pu, Minor actinides and LLFP.

In the « transmutation » scenarios (b and c), when compared to the corresponding critical reactor, an ADS has the following features :

- same transmutation rates for actinides (since proportional to the power in the core),
- extra-neutron availability to transmute LLFP (a neutron consuming process),
- subcriticality can allow to have a core with a very small fraction of delayed neutrons (case of the dedicated cores of scenario b).

THE PHYSICS OF TRANSMUTATION

"Transmutation" means excess neutron availability in the reactor core neutron balance.

The eventual "excess" G (expressed in neutrons/fission) is given by :

$$G = -\sum \varepsilon_J D_J - (CM + L) \qquad \text{(critical system)}$$

$$G = S_{ext} - \sum \varepsilon_J D_J - (CM - L) \qquad \text{(external source driven system)}$$

where : CM + L are neutrons (per fission) lost in parasitic captures or leakage
D_J is the neutron consumption (per fission) to transmute isotope J (down to fission products)
ε_J is the fraction of isotope J in the fuel of the system

Since CM + L is generally equal to ≈ 0.3 n/fission (for most systems) one can assess what system can allow "transmutation".

A positive G value can be obtained with fast reactors and practically any fuel. A positive G can be obtained with thermal neutron spectra only in very few cases.

THE "DOUBLE STRATA" SCENARIO AND THE ROLE OF ADS

A relevant question is if it is possible to reduce the number of reactors which should support the transmutation process.

One obvious way is to concentrate all the MA (and possibly some LLFP) in a few installations. If the fuel in the core is made only with MAs, the subcriticality characteristics offered by ADS can be relevant for safety reasons (improvement of β_{eff} and reactivity coefficients).

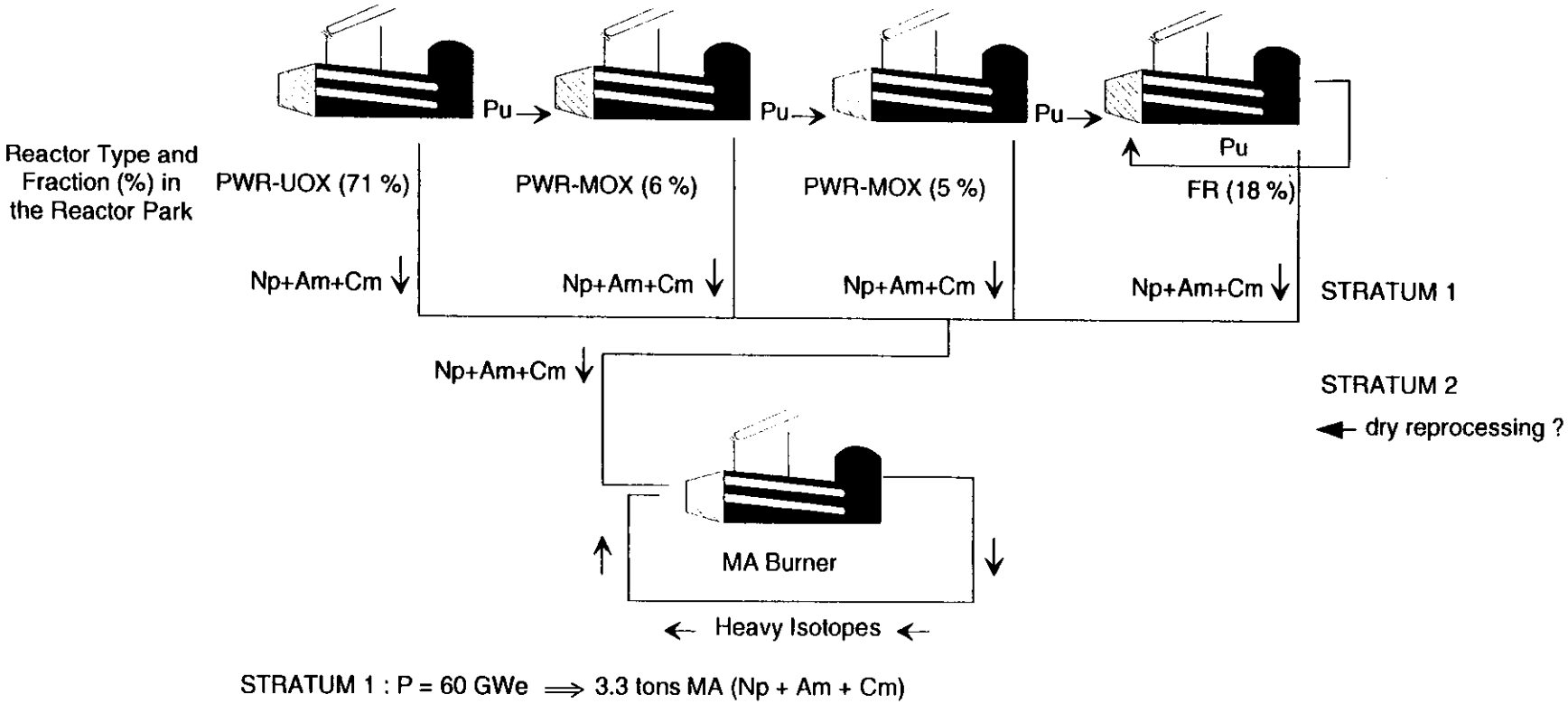
Moreover these ADS should have a fast spectrum :

G-values of MA fuel as unloaded from LWR's(*)

Criticality factor	LWR-Spectrum $\phi = 10^{14}$ n/cm^{2}s	LWR-Spectrum $\phi = 10^{15}$ n/cm^{2}s	FR-Spectrum $\phi = 10^{15}$ n/cm^{2}s
Keff = 0.95	-0.97	-0.67	+0.67

(*) negative value of G shows a lack of neutrons.

Scenario "Double strata", with Np, Am and Cm Transmutation



This reactor park allows to reduce waste potential radiotoxicity by a factor of 100 with respect to a reactor park operated in open cycle and producing the same amount of energy.

The limit is due to the level of losses during reprocessing (here assumed to be 1% for all MA, and 0.1% for U and Pu). If dry reprocessing is used, loss fractions can be lower.

Using dedicated MA-burner ADS, the following "equilibrium" NP park can be envisaged :

- ~ 70% of traditional LWR (UOX),
- ~ 6% of LWR (MOX) using a fuel of t he LWR-UOX discharge (the first recycle),
- ~ 5% of LWR (MOX) (the second recycle).

Besides, to prevent Pu-accumulation, one can use :

- ~ 18% of Sodium Fast Reactors (FR) in "burner mode" (CAPRA-type).

The remaining fuel (mostly Minor Actinides-MA) could be transmuted in subcritical ADS's.

- ~ 6% of ADS park is needed at equilibrium.

This park is also able to produce about 0.17 neutron/fission to incinerate the most dangerous long lived fission products (Tc, I, Cs) completely, as we will see in the next paragraph.

Finally, it is essential to remind that, whatever the performance of this system in terms of radioactive waste transmutation, a geological repository is always needed even if its realisation can be made easier in the context of the scenario indicated above.

THE LONG-LIVED FISSION PRODUCT (LLFP) TRANSMUTATION

The LLFP transmutation is related to a large neutron surplus availability (in units of neutrons/fission).

The neutron consumption/fission (D parameter), necessary to transmute Tc + I + Cs (elements) or the same D when only the long-lived isotopes (Tc-99, I-129, Cs-135) are supposed to be transmuted (i.e. after isotopic separations) is the following :

$$D_{(Tc + Cs + I)} = 0.15 \text{ n/fission}$$

$$D_{(Tc\text{-}99 + Cs\text{-}135 + I\text{-}129)} = 0.08 \text{ n/fission}$$

This number of neutrons per fission can be available if fast neutron spectra are used, both in critical or subcritical systems.

In practice, a possible technique could be to use the high neutron flux ($> 10^{15}$ n/cm^2.s) leaking out from a fast reactor core (critical or subcritical) in order to use it to transmute e.g. Tc-99 targets, in a moderated (e.g. by CaH_2 or $B_4^{11}C$) subassembly at the periphery of the core.

An example of application

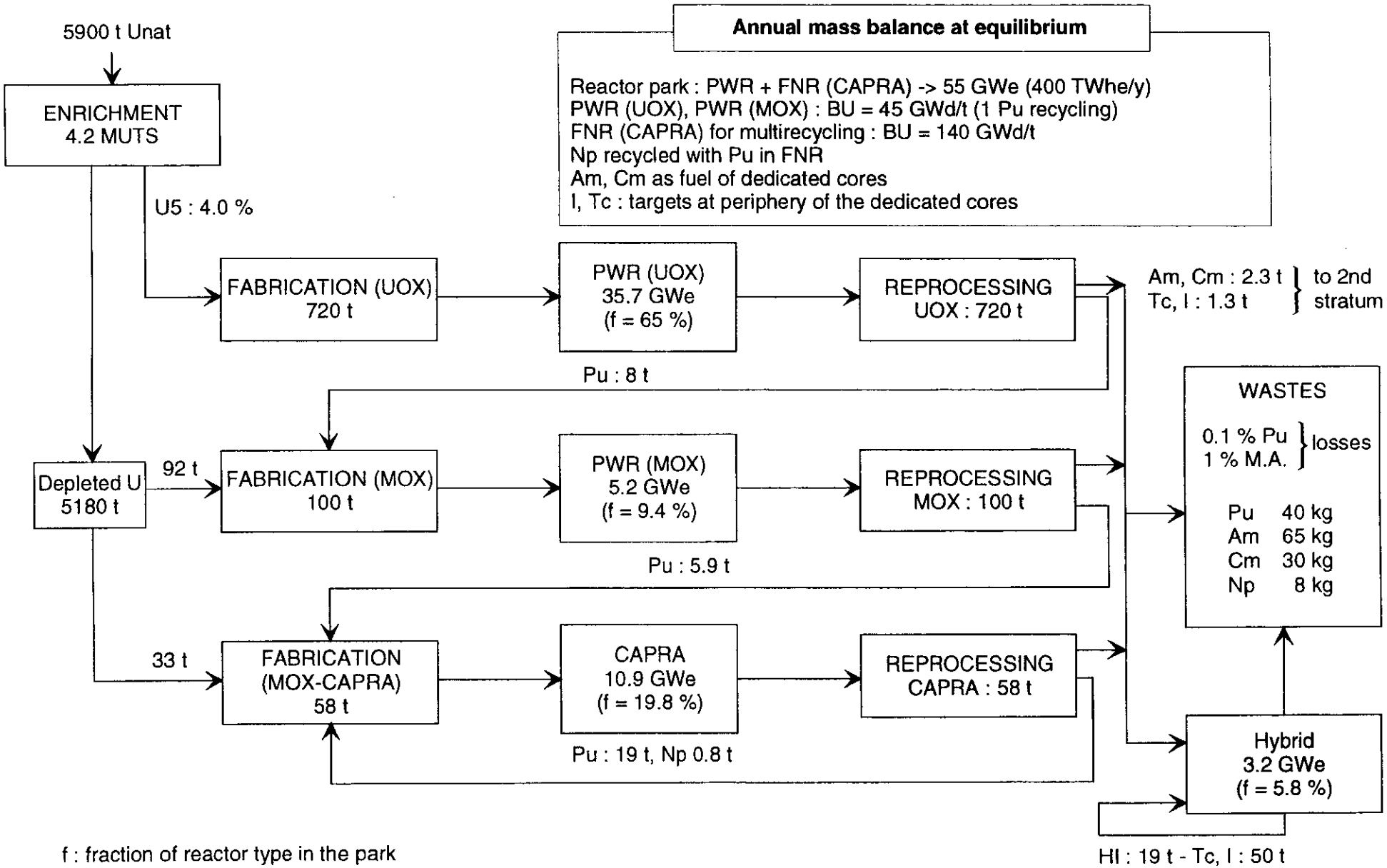
Dedicated burner cores can be of two types :

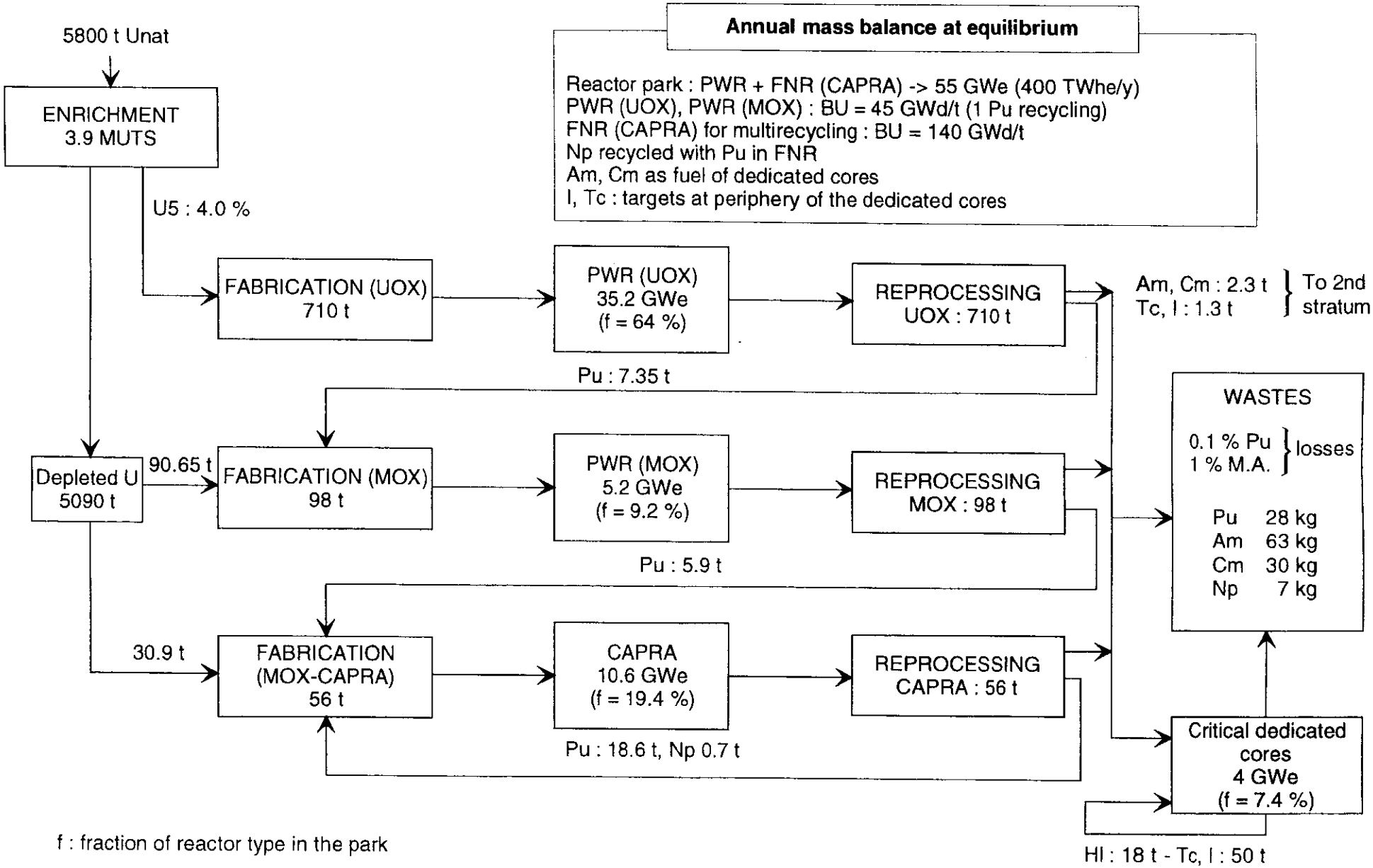
critical or subcritical (ADS).

	Critical	**Subcritical (ADS)**
Power	130 MWt	130 MWt
Neutron spectrum	fast	fast
Coolant	Pb/Bi	Pb/Bi
Fuel	Pu + MA	Pu + MA
MA burning rate	- 78 kg/TWhe	- 99 kg/TWhe
β_{eff}	~ 170 pcm	-
Target/beam	-	Pb/Bi target - Beam : E_p = 1 GeV - i_p = 1 mA (1.6 mA at EOC)

- If LLFP (I-129, Tc-99) introduced in « moderated flux » S/A at the periphery of the core (LSD concept), the following transmutation rates are obtained :

Tc-99	- 30 kg/TWhe	(both cases, critical or ADS)
I-129	- 18 kg/TWhe	


Annual mass balance at equilibrium
Reactor park : PWR + FNR (CAPRA) -> 55 GWe (400 TWhe/y)
PWR (UOX), PWR (MOX) : BU = 45 GWd/t (1 Pu recycling)
FNR (CAPRA) for multirecycling : BU = 140 GWd/t
Np recycled with Pu in FNR
Am, Cm as fuel of dedicated cores
I, Tc : targets at periphery of the dedicated cores
5900 t Unat
ENRICHMENT
4.2 MUTS
U5 : 4.0 %
Depleted U
5180 t
92 t
33 t
FABRICATION (UOX)
720 t
FABRICATION (MOX)
100 t
FABRICATION
(MOX-CAPRA)
58 t
PWR (UOX)
35.7 GWe
(f = 65 %)
PWR (MOX)
5.2 GWe
(f = 9.4 %)
CAPRA
10.9 GWe
(f = 19.8 %)
REPROCESSING
UOX : 720 t
REPROCESSING
MOX : 100 t
REPROCESSING
CAPRA : 58 t
Pu : 8 t
Pu : 5.9 t
Pu : 19 t, Np 0.8 t
Am, Cm : 2.3 t
Tc, I : 1.3 t
to 2nd stratum
WASTES
0.1 % Pu
1 % M.A.
losses
Pu 40 kg
Am 65 kg
Cm 30 kg
Np 8 kg
Hybrid
3.2 GWe
(f = 5.8 %)
HI : 19 t - Tc, I : 50 t
f : fraction of reactor type in the park



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Uncertainty analysis and data needs

- The validation of the neutronics of a subcritical core in the MUSE experiments has been satisfactory up to now. Same indications from the Rubbia experiments at CERN.

- The present uncertainties allow today meaningful assessments on ADS performances.

- However, in more advanced phases of design studies, higher accuracies can be needed, in order to reduce uncertainties on the nominal values of design parameters.

- How data needs can be assessed and in what areas ?

For the subcritical core, meaningful design parameters can be represented as « response » functions of the system :

$$R = < r, \phi >$$

How R varies if ϕ varies due to data uncertainties ?

$$R \rightarrow R' \text{ since } \phi \rightarrow \phi' \qquad \text{if } \begin{matrix} \sigma \rightarrow \sigma' \\ S \rightarrow S' \end{matrix}$$

Generalized Perturbation Theory (GPT) gives the formal frame.

If ϕ given by : $(A - M)\,\phi = S$

in the perturbated case : $(A' - M')\,\phi' = S \rightarrow R' = < r, \phi' >$

or

$(A-M)\,\phi'' = S' \rightarrow R'' = < r, \phi'' >$

A generalized « importance » function ψ^* is defined :

$$(A^* - M^*)\ \psi^* = r$$

which allows to derive simple expressions for δR variations :

$$\delta R = - < (\delta A - \delta M)\ \phi,\ \psi^* > \qquad \delta A = A' - A \qquad \delta M = M' - M$$
$$\delta R = < \delta S,\ \psi^* > \qquad \delta S = S' - S$$

⇒ Sensitivity and uncertainty analysis, towards data needs assessment and data adjustments (e.g. using integral experiments) :

$$(\delta R/R) / (\delta A/A) \qquad (\delta R/R) / (\delta M/M) \qquad (\delta R/R) / (\delta S/S)$$

⇒ These techniques can be applied to quantify the representativity of a demonstration experiment with respect to a prototype installation.

CONCLUSIONS

- The physics of a subcritical source driven core is well understood.
- However, integral experiments to validate data and methods are still essential.
- Some points are of crucial interest :
  - material damage (window and structures) under irradiation,
  - kinetic behaviour and safety analysis (what is the "optimal" subcriticality level ?),
  - quantification of nuclear data uncertainties and their impact.
- A demonstration experiment (~ 100 MWt) is a realistic goal, if experimental R and D is performed to justify basic design choices.