FIRST RESULTS AND FUTURE TRENDS FOR THE TRANSMUTATION OF LONG-LIVED RADIOACTIVE WASTES

by

C. PRUNIER, M. SALVATORES, Y. GUERIN et A. ZAETTA
CEA/CE-Cadarache
F-13108 Saint-Paul-lez-Durance
Cedex, France

ABSTRACT

In the frame of the CEA SPIN program, a project has been set-up at the Direction of Nuclear Reactors of CEA, to study the transmutation of long-lived radioactive produets (both minor actinides and fission produets) resulting from the operation of current nuclear power plants.

The program is focused on:

- transmutation of minor actinides (Np, Am) in fission reactors of known technology (both of the PWR or the fast reactor type), using the so-called "homogeneous" (mixed with Uranium or Uranium-Plutonium), and "heterogeneous" (with inert matrix) recycling modes for both type of reactors. A special version of the "heterogeneous" mode is explored for some speeific fission produets (e.g. Tc-99, 1-129).

- Transmutation studies in dedicated devices (both fission reactors with actinide/plutonium fuel or with high thermal flux, and particle accelerator-based systems).

- **Fuel studies related to both homogeneous and heterogeneous recycling modes in fission reactors. For the homogeneous recycling mode, some experimental irradiations results are available from past PHENIX programs. For the heterogeneous mode, very limited experimental results are available, and new theoretical and experimental work is underway on the use of appropriate inert matrices.**

- **Basic data studies** to assess the quality of existing nuclear data for fission reactor transmutation studies, future data needs of relevance, and model/data developments needed for accelerator-based systems.

- Strategy studies, to assess the consequences of the different transmutation options on fuel cycle, according to different scenarios of nuclear power development. These studies are performed in tight relation with the parallel studies that CEA is performing on the partition aspects.

The full paper will present:

- the program guidelines and objectives ;

- results obtained up to now in the different fields mentioned above, with special emphasis on the radiotoxicity source reduction;

- the trends for future developments, both in terms of theoretical and experimental studies.

TRANSMUTATION STUDIES AT CEA
(in the frame of the SPIN program)

O Transmutation in fission reactors of known technology (fast reactors, PWRs).

O Transmutation in dedicated devices.

O Basic studies (nuclear data, high energy particle/nucleus interaction models, etc.).

O Material studies.

O Fuel cycle and strategy studies.

OBJECTIVE

Reduction of the long term radiotoxicity source by transmutation of radioactive nucleides with half-lives higher than a few decades into stable nucleides, either directly, or after decay of nucleides of much shorter half-lives.

The nucleides of interest :

- O actinides of the 4 α-decay families (radiotoxicity source in the repository) ;
- O long-lived fission products (return to the bio-sphere afire solution and migration in the geologic environment).

THE ACTINIDE PRODUCTION (Example for the present French situation) :

- First, Pu (~200 g/MWe.y).
  If losses during reprocessing are= 0.3 %.
  Pu → wastes ≈ 30 Kg/y.

- **Np-237 : 10 g/MWe.y.**
  For the long term, it is useful to add the quantities resulting from Pu-241, Am-241 and Cm-245.
  If Pu losses ≈ 0.3 %, potential Np-237 → ~800 Kg/y.

- ***Americium :***
  Am-241 : For a reprocessing 3 years after exit from reactor, production of 5 g/MWe. y (→ 250 Kg/y).
  Am-242m → 0.014 g/MWe.y (small).
  Am-243 → 3 g/MWe.y.
  It represents the major source of Pu-239 in the wastes at t = $10^4 \div 10^5$ years (7+8 times higher than Pu-239 resulting from reprocessing losses).

- ***Curium :***
  Cm-242 (T = 6 months) → Pu-238.
  Cm-243 (T = 28.5 y) → Pu-239 in ' negligible amount with respect to Am-243.
  Cm-244 (T = 18 y) → Pu-240 in an amount 5 times higher than reprocessing losses.
  Cm-245 (T = 8.5×10^3 y) → Np-237, but in small amount.

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 :

$$R(t) = a \sum_i \frac{Q_i(t)}{Q_o(o)} \frac{D_i}{M T_i} \quad (sV/g)$$

- where is a constant $(= 1.322 \times 10^6)$;
- 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.

CONTRIBUTION OF EACH ISOTOPE TO RADIOTOXICITY (%)
IRRADIATED PWR FUEL (33 000 MWd/t)
3 years cooling 100 % Pu, Np, Am, Cm (open cycle)

ACTINIDES	MASS (g)	TIME (y) 10^2	10^3	10^4	10^5	10^6	10^7
Np 237	10040.				1.	17.	12.
Am 241	5187.	9.	8.			8.	6.
Am 242m	14.						
Am 243	2954.		1.	3.	2.5		
Am		9.	9.	3.	2.5	8.	6.
Cm 243	10.						
Cm 244	768.						
Cm 245	38.						
Cm		0.	0.	0.	0.	0.	0.
Pu 238	4343.	17.			4.	6.	
Pu 239	137771.	5.	17.	58.	78.	3.	39.
Pu 240	52840.	7.	22.	38.			3.
Pu 241	33297.	61.	51.		7.	53.	38.
Pu 242	130029.				7.	13.	
Pu		90.	90.	96.	96.	75.	80.
Radiotoxicity (SV)		$7.37\ 10^9$	$2.07\ 10^9$	$4.75\ 10^8$	$2.6\ 10^7$	$2.57\ 10^6$	$1.90\ 10^5$

CONTRIBUTION OF EACH ISOTOPE TO RADIOTOXICITY (%)
IRRADIATED PWR FUEL (33.000 MWd/t)
3 years cooling Losses 0.3% Pu 100% Np, Am, Cm to the wastes

ACTINIDES	MASS (g)	TIME (y) 10^2	10^3	10^4	10^5	10^6	10^7
Np 237	10040.			1.5	29.	65.	63.
Am 241	5187.	88.	81.		15.	33.	32.
Am 242m	14.						
Am 243	2954.	3.	12.	75.	50.		4.
Am		91.	93.	75.	65.	33.	36.
Cm 243	10.						
Cm 244	768.	5.	3.	13.5			
Cm 245	38.			1.5			
Cm		5.	3.	15.	0.	0.	0.
Pu 238	13.						
Pu 239	413.			4.	5.		
Pu 240	158.			3.			
Pu 241	100.	2.	2.				
Pu 242	39.						
Pu		2.	2.	7.	5.	0.	0.
Radiotoxicity (SV)		$7.5\ 10^8$	$1.9\ 10^8$	$1.9\ 10^7$	$1.3\ 10^6$	$6.5\ 10^5$	$3.9\ 10^4$
Fission Products							
Tc 99	17405.	$3.7\ 10^3$	$3.7\ 10^3$	$3.6\ 10^3$	$2.7\ 10^3$	$1.4\ 10^2$	0.
I 129	4026.	$1.9\ 10^3$	$1.9\ 10^3$	$1.9\ 10^3$	$1.9\ 10^3$	$1.9\ 10^3$	$1.2\ 10^3$
Cs 135	9768.	$7.9\ 10^2$	$7.9\ 10^2$	$7.9\ 10^2$	$7.8\ 10^2$	$5.9\ 10^2$	$3.9\ 10^1$

ASPECTS TO BE ACCOUNTED FOR IN FISSION REACTOR TRANSMUTATION

O Since fission is the essential **mechanism,** increase of **$\sigma f/\sigma c$** should be looked for (cumulated fissions).

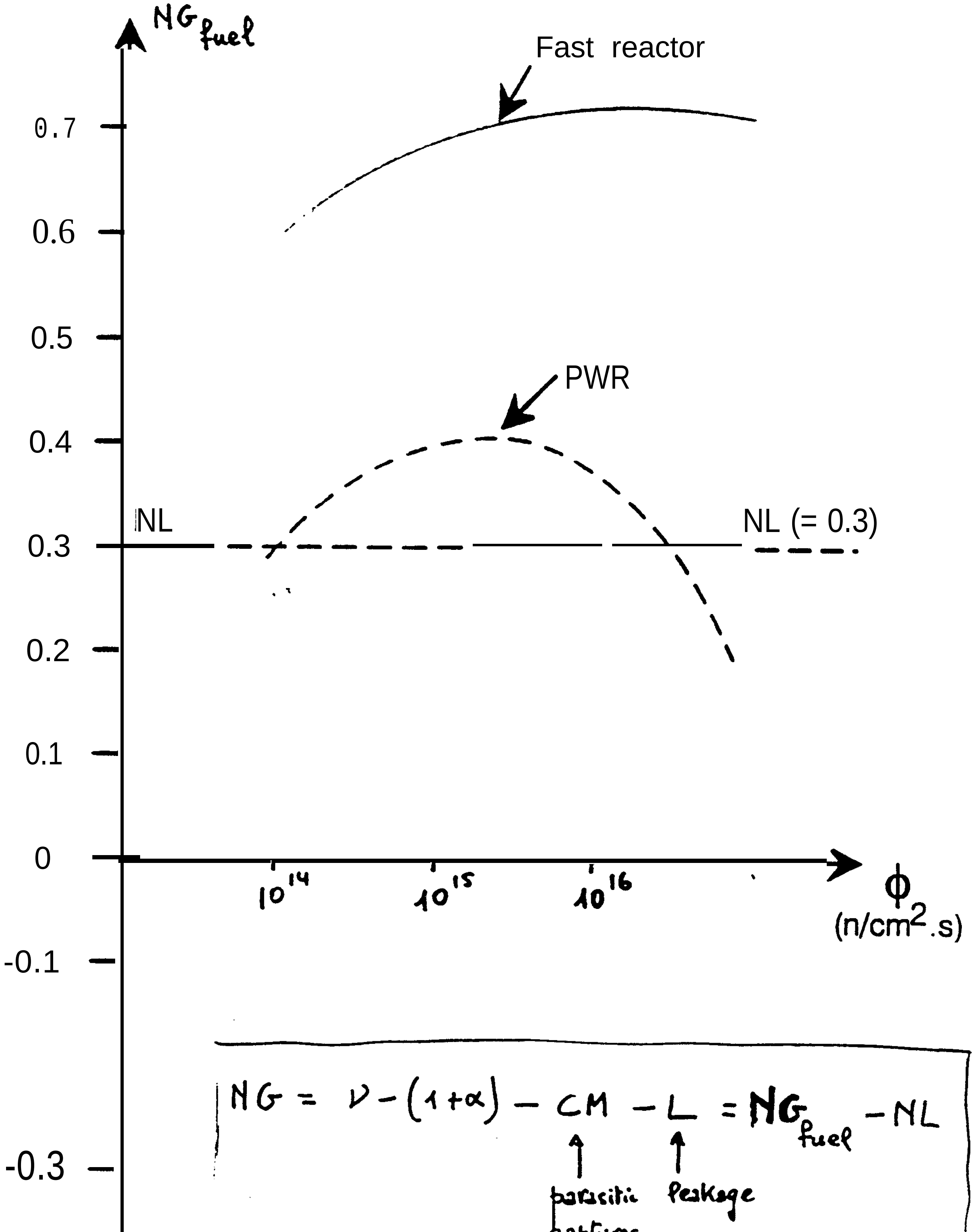
O **The production** of higher isotopes **by** **(n,γ), $(n,2n)$,** etc,.“” should be limited (consequences on the **fuel** cycle).

O Neutron balance effectiveness.

O Consequences on ***the*** neurotics core parameters.

0 Fuel fabrication issues.

Neutron gain NG fuel as a function of ϕ



$$NG = \nu - (1+\alpha) - CM - L = NG_{fuel} - NL$$

CM: parasitic captures

L: leakage

FISS1ON REACTOR STUDIES

◎ Minor Actinides transmutation :

homogeneous
recycling modes
heterogeneous

in fast reactors and PWRs.

O Influence of
- fuel type (MOX, UOX, metal, . ..).
- reactor size / spectrum,
- moderator / fuel ratio.

0 Consequences on the core neutronics parameters.

o Consequences on the fuel cycle (neutron sources, activity, decay heat, . ..).

◎ Fission Products transmutation :

0 use of moderated S/A at the periphery of a fast reactor,

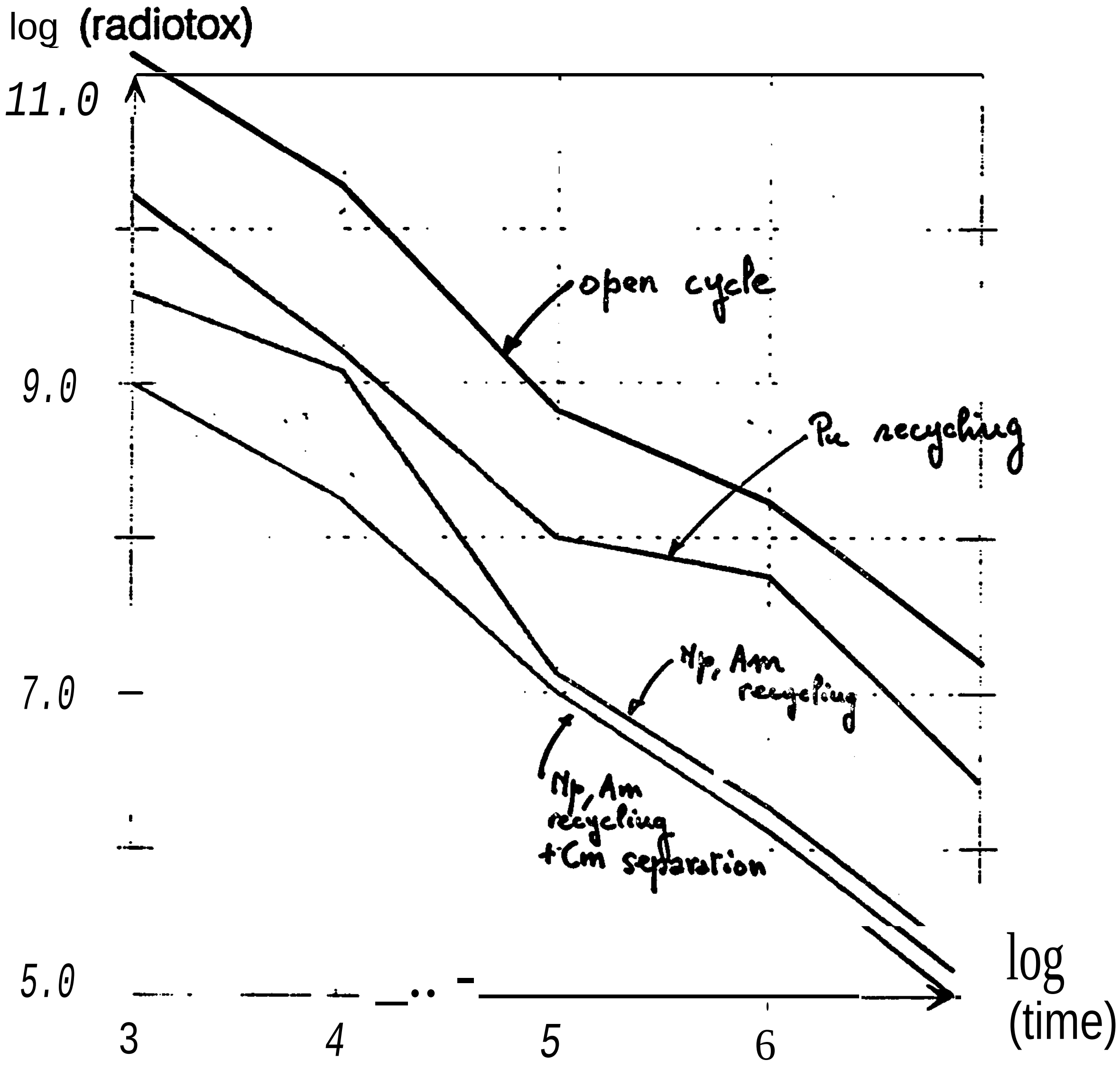
O fission product targets.

MINOR ACTINIDE BURNING IN FAST REACTORS

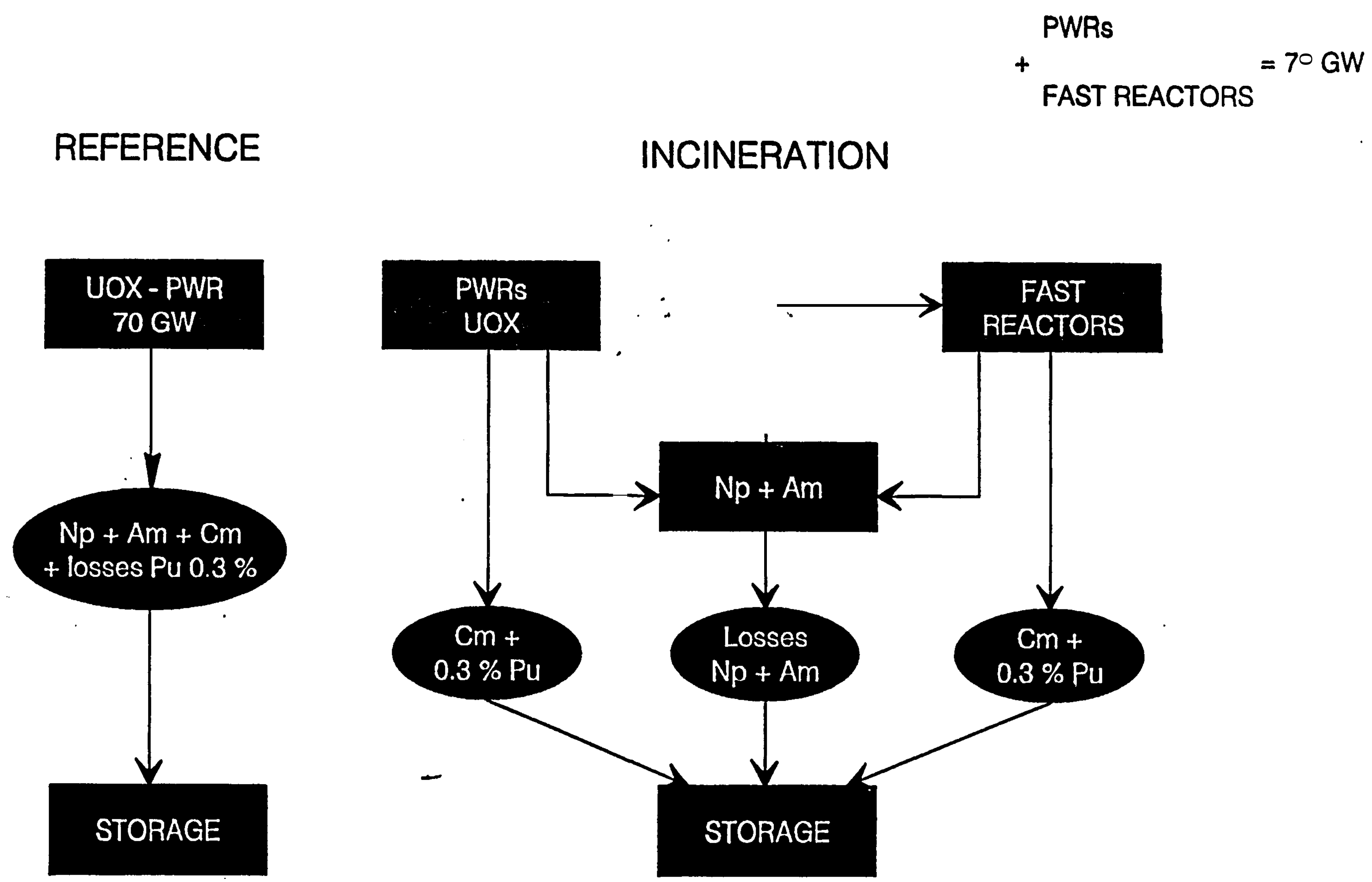
	HOMOGENEOUS RECYCLING	HETEROGENEOUS RECYCLING
Consequences on the core parameters	Na void coeff. ↑ (BOC) Na void coeff ~ const (EOC) Doppler coeff. ↓ Δρ/cycle ↓	Control of power distributions at the core outer boundary
Pu-238 content (Pu-238/Pu)	~ 5 %	~ 80 %
Envisageable MA content	~ 2.5 % (EFR-type) ~ 5 % (PRISM-type)	~ 50 % (Np) (less for Am)
Spectrum, fuel type, reactor size effects	Small	-
Waste radiotoxicity reduction	Factor ~ 10 to ~ 30	Factor ~ < 10 to 30
Ratio FR/Park	~ 20 %	~ 30 %
Recycling mode	Multiple recycling	Multiple recycling
Consequences on the fuel cycle (preliminary)	Decay heat : + 20÷50% n source : + factor 2	Not available
Experimental validation	SUPERFACT experiment	Foreseen
Uncertainties	To be evaluated (small on the core parameters)	

Homogeneous actiniderecycling in a EFR-type reactor

	Radiotoxicity reduction factor at time t (years)					
	t=lo[2]	10[3]	10[4] -	10[5]	10[6]	10[7]
EFR (Np + Am) (losses = 1 %)	3.C	4.5	1.4	7	20	14
EFR (Np + Am + Cm) (losses 1 %)	14	16	7	8	33	25



Open cycle (Pu, MA -> wastes)
— Closed cycle (MA -> wastes) Pu losses: 0.3 %
— Np, Am recycling (1 % losses -> wastes) Cm -> wastes
—— Np, Am, Cm recycling (1 % losses to wastes)


PWRs
+
FAST REACTORS
= 7° GW
REFERENCE
INCINERATION
UOX - PWR
70 GW
Np + Am + Cm
+ losses Pu 0.3 %
STORAGE
PWRs
UOX
FAST
REACTORS
Np + Am
Cm +
0.3 % Pu
Losses
Np + Am
Cm +
0.3 % Pu
STORAGE

MINOR ACTINIDE BURNING *IN PWRs*

	HOMOGENEOUS RECYCLING	HETEROGENEOUS RECYCLING
Consequences on the core parameters	Reactivity coefficients (case of MOX fuel)	Control of power and its evolution during the cycle
Pu-238 content (Pu-238/Pu)	~ 20 %	~ 80 %
Maximum MA content	1% (UOX, less with MOX)	< 50 % (Am to be found)
Extra enrichment	1 % (case of UOX)	1 % (case of Np)
Spectrum, fuel type, reactor size effects	To be defined for MOX fuel	-
Waste radiotoxicity reduction	Factor ~ 10 to ~ 50	Factor ~ 10 to 25
Ratio PWR*/Park	~ 20 %	~ 30 %
Recycling mode	Multiple recycling (One-through for Am ?)	Multiple recycling (One-through for Am ?)
Consequences on the fuel cycle (preliminary)	n source $\uparrow$ (Factor 30) Activity $\uparrow$ (Factor 15)	Not available
Experimental validation	Foreseen	Pu-238 production at CELESTIN reactor More foreseen
Uncertainties	To be evaluated	

FISSION PRODUCT BURNING (Tc-99, I-129)

Fast reactors : high flux, small σ_C

Thermal reactors : low flux, high σ_C (but self-shielding).

→ Possibility to use S/A with moderator materials, at the periphery of a fast reactor core (experience at PHENIX for CO-60 production) : high thermal flux, high σ_C.

Preliminary results :

Irradiation in :		Half-lives (y) Tc-99	Half-lives (y) I-129
PHENIX radial blanket	1st row (moderated S/A)	5	3
	2nd row (moderated S/A)	10	7
	3rd row (moderated S/A)	20	13
	1st row (standard S/A)	47	44
PWR		13	47

0 No adverse effects due to other isotopes (Tc, I).
O More difficult for Cs.
O Experimental validation foreseen in PHENIX for Tc-99.

⇨ Experimental validation on CO-60 production in PHENIX : C/E ~1 ÷ 1.3.

PRESENT TRENDS

- O The physical feasibility of MA burning in fission reactors is well understood.

- O In fast reactors this operation is more favorable.

- O The homogeneous recycling mode has had a preliminary validation in PHENIX (the SUPERFACT experiment).

  For the heterogeneous recycling mode, past experience for Pu-238 production in thermal spectrum was obtained with NpO_2-MgO targets.

  An irradiation experiment in PHENIX blanket (moderated S/A) is foreseen with the same type of target.

- O The Np-237 problem seems to be most conveniently treated, even in the short term, by homogeneous recycling with Pu in fast reactors.

  The Am heterogeneous recycling potential should be investigated more carefully.

  Cm should be separated, but probably not recycled in reactors. The Pu formed could be successively re-used.

A DEMONSTRATION IN SUPER PHENIX IS FORESEEN:

⇒ Next core (~1995 ÷ 1996) :

~1 Kg Np-237 → "homogeneous" pins (~2 % Np) in a standard S/A of the core (~1/2 S/A).

⇒ Following core (intended to enhance Pu burning) :

~10 Kg Np-237.

→ ~4 S/A with "homogeneous (Pu U Np) O_2
(possibility for some heterogeneous recycling demonstration)
~1 ÷ 2 pins with ~ 2 % Am (homogeneous)

ADVANCED SYSTEMS

O Preliminary study of a fast reactor with Pu/Np/Am fuels :

Na void coefficient ↗
Doppler : ~0
β_{eff} : ~3 times smaller than in a standard fast reactor
High reactivity loss/cycle.

Theoretical burning potential high :

~1000- 1500 kg MA per year, to be compared to ~100 kg/y in a fast reactor with homogeneous recycling (2.5 % Np, Am) (3600 MWth).

O "Hybride" system (p accelerator / sub-critical region) :

- Potential for Np, Tc-99 and other fission products. Not evident for Am (and Pu).
- Basic studies necessary on cascade code *performances (both* experimental and theoretical intercomparisons).
- Target performance issues.

ADVANCED SYSTEMS

O Optimisation of a **"Pu/MA fuel"** core.

O High flux **reactors** potential.

O Analysis of accelerator-basal systems.

Is high **current** necessary ?
Is high **thermal** flux always necessary ?

O New options for fission products ?

Neutron consuption for isotope J, NC_J:

$$NC_J = \sum_{J1} P_{J \to J1} \left(R_{J1} + \sum_{J2} P_{J1 \to J2} \left(R_{J1} + \sum_{J3} P_{J2 \to J3} \left(\cdots \right.\right.\right.$$

R_{J1} = n° of neutrons to produce J1

(=0 for decay ; =1 for capt ; $-\nu+1$ for fiss. etc)

Type of spectrum Φ level ($n \cdot cm^{-2} \cdot s^{-1}$)	FR 10^{15}	FR 10^{17}	PWR 10^{14}	PWR 10^{16}	D_2O 10^{16}
Pu-238	-1.4	-1.5	0.6	0.04	=(0),142
Pu-239	-1.46	-1.5	-0,67	-0.8	-1.06
Pu-240	-0.96	-1.2	0.44	0.09	0.17
Pu-241	-1.24	-1.6	-0.56	-0.9	-0.8
Pu-242	-0.44	- 0.75	1.76	1.1	1.2
Np-237	-0s9	-0.72	1.12	0. S	-0.4
Am-241	-0.62	-0.78	1.1	0.08	-0.6
Am-242	-1.4	-1.5	0.4S	40.9	-1.6
Am-243	-0.6	-1.1	0. 8	0.16	0.19
Cm-243	-2.1	-2.3	-4"9	-2.0	-1.6
Cm-244	-1.4	-4*9	- 0.15	-0.5	-0.5

FUEL STUDIES

- Theoretical and experimental studies on inert matrices in particular for Am and Fission Products.

- SUPERFACT-2 irradiation in PHENIX (higher bum-up).

- Experimental 'validation of heterogeneous mode in PHENIX (Am, FP).

- Experimental irradiation program in the OSIRIS reactor (ACTINEAU program for PWR homogeneous and heterogeneous recycling options).

SUPERFACT

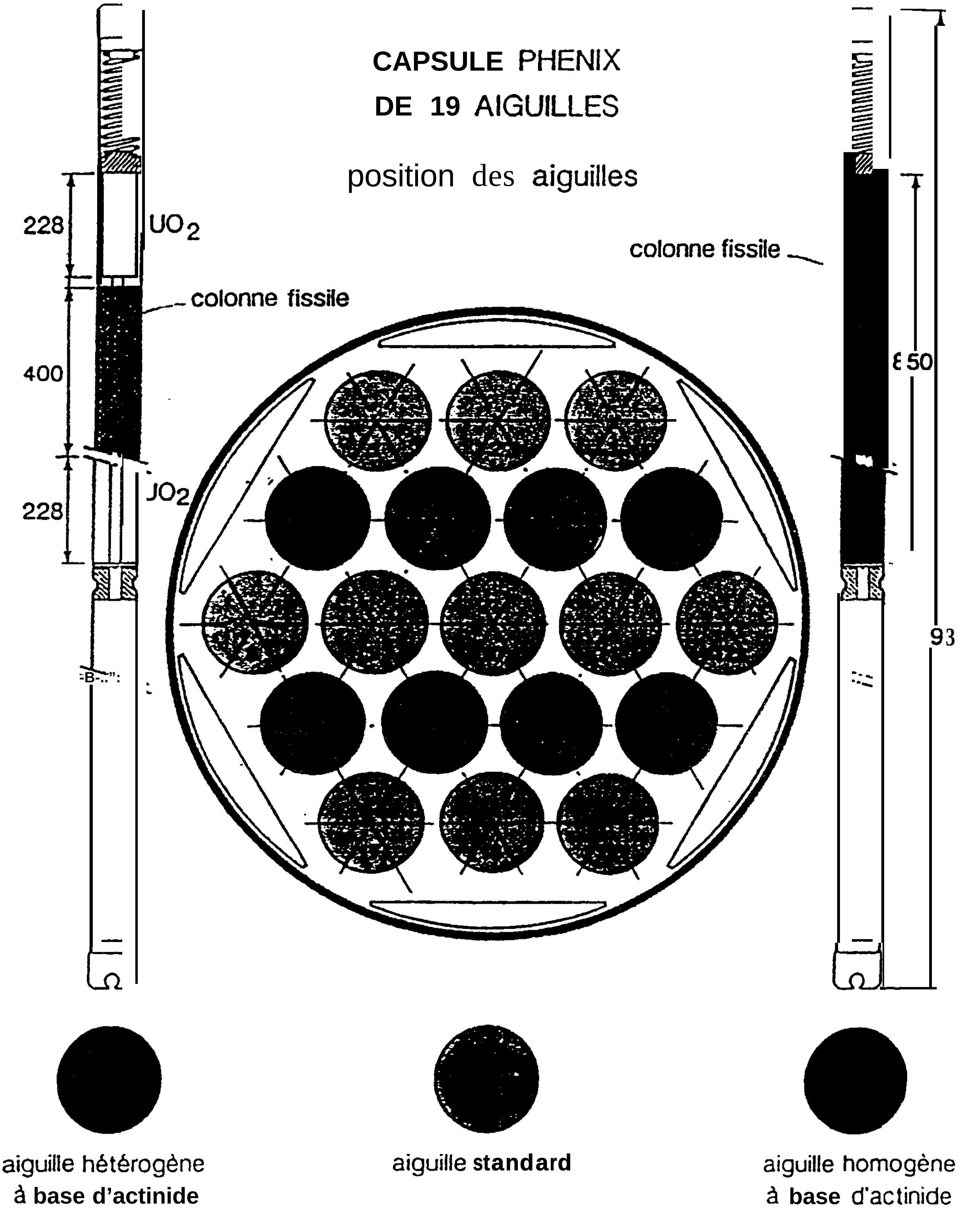


Figure 2: CONCEPTION DE LA CAPSULE D'IRRADIATION PHÉNIX

BASIC STUDIES

- O Full exploitation of irradiated fuel analysis, and separated isotope irradiations available at CEA (PROFIL, SHERWOOD, ICARE experiments, relative to fast, intermediate, thermal neutron spectra).
- 0 Sensitivity analysis and data requirement definition.
- O Cascade code validation (experimental and theoretical). Data needs assessment (in the frame of NEA-OECD).
- O Critical experiment planning.

Finally, as a contribution to data benchmarking, a re-evaluation has been made of the pure sample irradiation experiments PROFIL in PHENIX, using the recently released JEF-2 data.

In the following table the C/E results for some minor actinide reaction of interest, are indicated :

	C/E with	
	JEF-1 data	JEF-2 data
σ_c (Pu-238)	0.95	0.97
σ_c (Np-237)	0.90	0.94
$\sigma_{n,2n}$ (Np-237)	1.19	1.15
σ_c (Am-241)	1.03	1’.05
σ_c (Am-243)	0.94	0.97

STRATEGY STUDIES
(COSI CODE)

- Initial inventories according to different scenarios.
- Different hypothesis on cooling/fabrication delays.
- Losses and partitioning efficiency impact.
- Inventories in the case of introduction of actinide burning concepts (evolutionary / revolutionary) :
  - medium term effects,
  - long term effects.
- A study is underway under contract with EEC.

TRANSMUTATION →

REACTOR TYPE / REPROCESSING	PWR (UOX) 2000->2100	PWR (UOX+MOX) 2000->2100	PWR (UOX+MOX) 2000->2020 / PWR (UOX) FBR 2020->2100
NO direct disposal	R1 REFERENCE SCENARIO		
EXISTING **RP 0** U, Pu, Pf		R2 SCENARIO Recyclage Pu	R3 SCENARIO Recyclage Pu
RP 1 U, Pu, Pf, Np, Am Targets not reprocessed		RP1-1 SCENARIO Same as R2->2010 scenario plus recycling Pu, transmutation Np, Am	RP1-2 SCENARIO same as R3->2010 scenario plus recycling Pu, transmutation Np, Am
RP 2 U, Pu, Pf, Np, Am Cm, long half life FP (I129,Cs135,Tc99) Targets reprocessed		RP2 SCENARIO - Pu recycling - Incinération Np, Am, Cm and long half-live FP in dedicated reactors or particule accelerators	

SEPARATION (time axis: 00, 2010, 030)

CEA DCC / DPR / SSP	ETUDE CCE PARTITION/INCINERATION	Réf : SSP/BCE 92/	DATE : 15/09/92
	RESEARCH SCENARIOS		FIGURE 0

FUTURE STUDIES

Reactor studies :

0 Fast Reactor Optimisation for homogeneous recycling (heterogeneous concept potential).

O Heterogeneous recycling for Am (Fast Reactors, PWRs).

0 MOX fuel PWRs complementary studies.

O Detailed analysis of the consequences on the fuel cycle for the different strategies (wastes in the repository, wastes in the fuel cycle, etc.).

O Cm strategy studies

Objectives : to reduce options by the end of 1994.