AN OVERVIEW OF THE SPIN PROGRAMME

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I. BACKGROUND

The SPIN programme fits into the important and increasing popular concern regarding high activity and long-lived nuclear waste management.

Subsequent to Mr. Christian Bataille's report, the National Assembly and the Senate discussed and adopted a law aimed at ensuring radioactive waste management in compliance with the protection of nature, the environment and health, taking the rights of future generations into consideration.

The French Law of December 30, 1991 explicitly requests that three subjects be studied over a period of 15 years:

- The search for alternatives enabling the separation and transmutation of long-lived radioactive elements present in these wastes.
- The study of reversible or irreversible storage possibilities in deep geological formations, and particularly thanks to the implementation of underground laboratories.
- The study of conditioning and long duration interim surface storage processes for these wastes.

Upon completion of these studies in 2006, Parliament will possibly have to come to a decision on the construction of an underground disposal site and the continuation of other lines of research.

I.1. Radionuclides to be taken into consideration:

The starting point for the study of the first subject is the inventory in radionuclides of the fuel from a PWR 900 irradiated to 33 $GWdt^{-1}$ (UOX enriched at 3.25%) and cooled for three years, the assessment of its radiotoxicity and the search for ways by which to reduce it.

The irradiated fuel discharged per year from a PWR 900 MWe contains 23 t of Uranium (with 0.9% of 235), 240 kg of Pu, 800 kg of fission products, 18 kg of minor actinides.

Its potential radiotoxicity can be defined, outside any notion of confinement barrier, by the calculation of a "source term" obtained by weighting the activity of each radionuclide by its coefficient of specific toxicity (upon ingestion or inhalation), and then by summing up the values obtained.

Table 1 gives the value of the **potential radiotoxicity** of an **irradiated fuel** and the contribution of each long-lived radioisotope, on the basis of its parent product present today. These values are expressed in sieverts per TWh_e generated.

After the decay of highly radioactive fission products (Cs 137 - Sr 90) estimated at 300 years, the major contribution comes from plutonium, up to 10^6 years. Americium has a preponderant place among the minor actinides between 10^2 and 10^5 years. Neptunium becomes important after about 10^5 years. Curium makes a large contribution before 10^4 years. The contribution of fission products is negligible.

When the notion of confinement barrier is adopted, for deep disposal for example, the return of radionuclides to the biosphere must be assessed according to the degradation of the waste packages with time and to the transport of radionuclides through the geological media. This release of activity, weighted by the values of specific toxicity coefficients, leads to the notion of residual toxicity. The models are complex and site-dependent. A study performed in 1990 (PAGIS) for glass packages (containing no uranium, plutonium or iodine) in a granitic site (AURIAT) and for a standard scenario, showed that no dose was released before 10^4 years, that it then remained lower than the limit recommended by the ICRP and that the elements to be taken into consideration for storage safety were the most mobile ones (technetium, cesium) and the most radiotoxic (actinides). (See Figure 1). Incident scenarios are also to be considered.

I.2. Strategy Developed:

A range of concern is developed below:

Concerning **potential radiotoxicity**, the high priority is to use or destroy plutonium, and then the minor actinides. The theoretical gains thus obtained may reach a factor of 500 for a standard fuel from a PWR 900 between the open cycle in which the irradiated fuel is sent to waste and that in which the plutonium and the minor actinides would be separated and destroyed with respective efficiencies of 99.9% for Pu and 99% for minor actinides.

But there is a wide gap between theoretical gains and real gains, separation and transmutation having limited yields and the latter being accompanied by the creation of radiotoxic elements (generally by neutron capture). Multi-recycling and global optimisation of the system is necessary to reach the best results concerning the use of resources and the limitation of the production of radiotoxic isotopes during multi-recycling.

Concerning **residual radiotoxicity**, the true to life model for deep disposal will be available only once the storage site is known and only tendancies can be seen which give, for example, iodine and cesium as the most soluble elements in reducing environments.

I.3. Advantage of Reprocessing

The reprocessing of irradiated fuel, which recovers energy-bearing materials, already allows 99.5% of the plutonium to be separated (guaranteed value), soon to be 99.9% thanks to the efforts made in the PURETEX programme to improve the management of reprocessing wastes so as to reduce their volume and activity.

Reprocessing also allows to envisage strategies for separating other radionuclides in line with current techniques, either by modifying the process (Np, Tc, I, Zr) or by adding separation operations to the high activity stream (Am, Cm, Cs).

II. GOALS AND ASSETS OF THE SPIN PROGRAMME

The goals of the SPIN programme are divided into two sub-programmes:

PURETEX, for the short and medium term, endeavours to reduce the volume and activity of reprocessing wastes from 1.5 m^3 to 0.5 m^3 per ton of reprocessed heavy metal (1.7 m^3/t for direct disposal involved in the open cycle).

ACTINEX, for the long term, aims at assessing the various separation and transmutation strategies, their feasibility and their insertion in nuclear power generation.

II.1. PURETEX:

II.1.1. PURETEX - Solid Wastes

There are four types of waste not acceptable for surface-storage:

- Structural wastes from assemblies, hulls and end-pieces, embedded in grout.

- Coprecipitation sludge from effluent processing stations currently embedded in bitumen.

- Technological wastes embedded in concrete.

- Vitrified high activity effluents (F.P. and minor actinides).

The table below gives the volumes of these wastes specified in the La Hague plant process book, those produced in 1993 and the goals for 1995 and 2000 (see Figure 2).

Table 2 - Volume (per ton of U in the fuel) of residues in each category:

		Packaging Modes	UP3 rated values	1993 achieved values	1995 expected values	2000 expected values
Waste not compatible with present surface-storage requirements	Fission products	Glass blocks (C)	130 l/tU	115 l/tU	115 l/tU	115 l/tU
	Hulls and end-pieces	Concrete (B)	600 l/tU	600 l/-U	<600 l/tU	150 l/tU
	Sludges	Bitumen (B)	630 l/tU	450 l/tU	0	0
	Technological wastes	CAC concrete blocks (B)	1700 l/tU	200 l/tU	200 l/tU	<200 l/tU
	TOTAL		3060 l/tU	1365 l/tU	<915 l/tU	<465 l/tU

The strategy adopted to obtain these performances consists in replacing the grouting of the hulls and end-pieces by a process still to be defined, in re-designing the whole management of liquid effluents to eliminate their processing by sludge and then bitumen-generating precipitation, in sorting the technological wastes, demoting them by decontamination whenever possible, or redefining methods for their processing.

Report on the Studies:

• The hull fusion process has been improved in the α β γ laboratory, making it possible to lower the α activity from $3.5 \cdot 10^{11}$ Bq/t (9.4 Ci/t) to $3.3 \cdot 10^{10}$ Bq/t (0.9 Ci/t) (about 10 times the surface standard).

A precise characterisation of the end-pieces may possible put demoting within reach.

Compacting of hulls and end-pieces is proposed by COGEMA, as a safe and reversible interim storage alternative.

- System studies have shown that the sodium ion is the dominant cation that prevents effluents from being processed by distillation and their activity from being sent back to vitrification, which would eliminate the precipitation of sludges and the production of bitume.

  Replacing sodium carbonate by cetomalonic acid in the treatment of solvent has been demonstrated in the α laboratory.

- The leaching process with silver II to decontaminate metallic waste highly contaminated by PuO_2 has reached its industrial implementation phase.

- The evolution of vitrification towards a series of specific types of glass has begun: a sodium glass is under study as a complement to the R7/T7 glass.

II.1.2. PURETEX waste streams:

The PURETEX programme is also involved in limiting releases of activity into the sea.

Current release authorisations are 1700 TBq $\beta\gamma$ (45 000 Ci $\beta\gamma$) exclusive of tritium and 1.7 TBq α (45 Ci α) for the La Hague reprocessing plants. The released values are much lower. Thus, releases were 0.15 TBq α (4.1 Ci α) and 110 TBq $\beta\gamma$ (3100 Ci $\beta\gamma$) respectively in 1991, 0.1 TBq α (2.9 Ci α) and 76 TBq $\beta\gamma$ (2067 Ci $\beta\gamma$) in 1992, 0.1 TBq α (2.7 Ci α) and 73 TBq $\beta\gamma$ (1980 Ci $\beta\gamma$) in 1993, i.e. about 6% and 4% of the authorised values.

COGEMA and CEA have assigned R&D ambitious reduction goals aimed at releasing only about 1% of authorised values.

II.2. ACTINEX:

To evaluate the various separation and transmutation strategies, a hierarchy must be given to the concerns (and regularly updating them) and the technical feasibility of separation and transmutation must be assessed.

The hierarchisation is more advanced in the study dealing with potential toxicity. Actinides are involved (plutonium, neptunium, americium, curium) and, starting from 10^5 years, their radiotoxicity is of the same order of magnitude that the one of wastes containing uranium. Fission products are negligible.

Case studies are made based on available information: recycling of plutonium in PWR, in FBR, incineration of neptunium and of americium.

FBRs come closer to the theoretical scenario than PWRs. Furthermore, they accept the degraded isotopies of plutonium, may be adapted to a higher consumption of plutonium and are better suited for the incineration of minor actinides in the sense that they produce fewer higher isotopes, owing to a higher ratio of fission cross sections to capture cross sections for different isotopes and a better economy of neutrons in the core.

The CAPRA programme was launched to better evaluate these performances and to master the technology of an incinerator FBR.

Hierarchisation remains very difficult in the case of residual toxicity which brings out the fission products I, Cs, Tc, Zr, Pd.

The Interim Storage - Disposal and Storage Safety programmes must define the radionuclides that can return to the biosphere.

Technical feasibility of separation

The separation of plutonium is a reality and has been industrially mastered. The PURETEX programme will allow a maximum loss of 0.1% to be guaranteed.

The separation of neptunium may be mastered in the PUREX process itself by enhancing its extraction during the first cycle and its separation during the second uranium cycle. The accessible performance remains to be quantified (greater than 70%).

The separation of americium and of curium is much more difficult, especially if totally incinerable solvents are to be used. The DIAMEX process (under development) uses amides, extracts 99% of the americium and curium with the lanthanides. The SESAME process aims at selectively extracting the americium alone with an oxidized valency. Other fundamental studies are undertaken to separate these minor actinides.

Iodine is presently separated in the reprocessing process.

Technetium and zirconium are fission products with a behaviour already known in reprocessing. Some studies are performed on the solubility and separation of technetium which is greater than 70% for a standard PWR fuel. The main effort has concerned cesium for which a specific macrocycle extractant of the calixarene type has been defined at the fundamental research laboratory scale.

Palladium is part of the dissolution sludges. Its behaviour is unknown.

Technical feasibility of transmutation

The basic nuclear data are not all available and experiments are necessary, either for transmutation in a fission reactor (integral experiments to validate the European base, complementary data for technetium), or for hybrid system calculations (intermediate energy data and cascade codes).

Data are introduced into the calculation models used to predict the behaviour of radioelements in power reactors and the acceptable limits for these reactors without consequences on safety.

In a first analysis, the following indications may be given:

- the Increased Moderation Array PWRs have the best performance of all the PWRs in reducing the production of minor actinides and increasing the consumption of plutonium.

- FBRs give better performances than PWRs. They enable increased consumption of plutonium and incinerate the minor actinides with a lower production of higher isotopes.

- Various calculations have shown consumptions of minor actinides Am, Np from 7 to 8 kg/TWh_e.

- The homogeneous mode (diluted in all the rods) is the most promising method for the recycling of neptunium, the heterogeneous mode (as targets) is preferable for the recycling of americium.

These calculations have to be backed up by irradiation experiments:

The result of neptunium transmutation calculations with limited burnup has been validated in a Superfact 1 (Phenix).

Superfact 2 (Phenix) is planned for the transmutation of neptunium and americium with a higher burnup.

A demonstration will be made in Superphenix.

ACTINEAU is an experiment programmed in the OPERA (PWR) loop to constitute a database on the transmutation of neptunium.

In the case of fission products, the calculations and experiments are not as advanced. Technetium may be transmuted into stable ruthenium in a fast reactor (technetium loses 50% of its mass in 3 years in radial blanket and thermalised flux).

The accelerator-sub-critical medium hybrid systems show that these systems offer no supplementary potential for the transmutation of minor actinides. Their use for the transmutation of fission products requires additional studies.

III. FIVE-YEAR PLAN

The SPIN programme is planned over fifteen years with a more detailed definition over a five year rolling plan.

In financial terms (MF):

	1994	1995	1996	1997	1998
SPIN	250	300	300	300	290
including (Investments)	30	64	70	55	40

PURETEX	Years of R&D
• Hull fusion	92-98
• Hull compacting	93-97
• Elimination of the bitumen	92-96
• Sodium glass	94-96
• Decontamination of the wastes	92-97
• Reduction of releases	95-?

ACTINEX

•	Separation of the actinides	92-98
•	Separation of the fission products	94-99
•	Basic data for transmutation	93-97
•	Transmutation studies in fission reactor	93-97
•	Fuel studies and transmutation experiments	93-98
•	Hybrid transmutation system studies	93-97

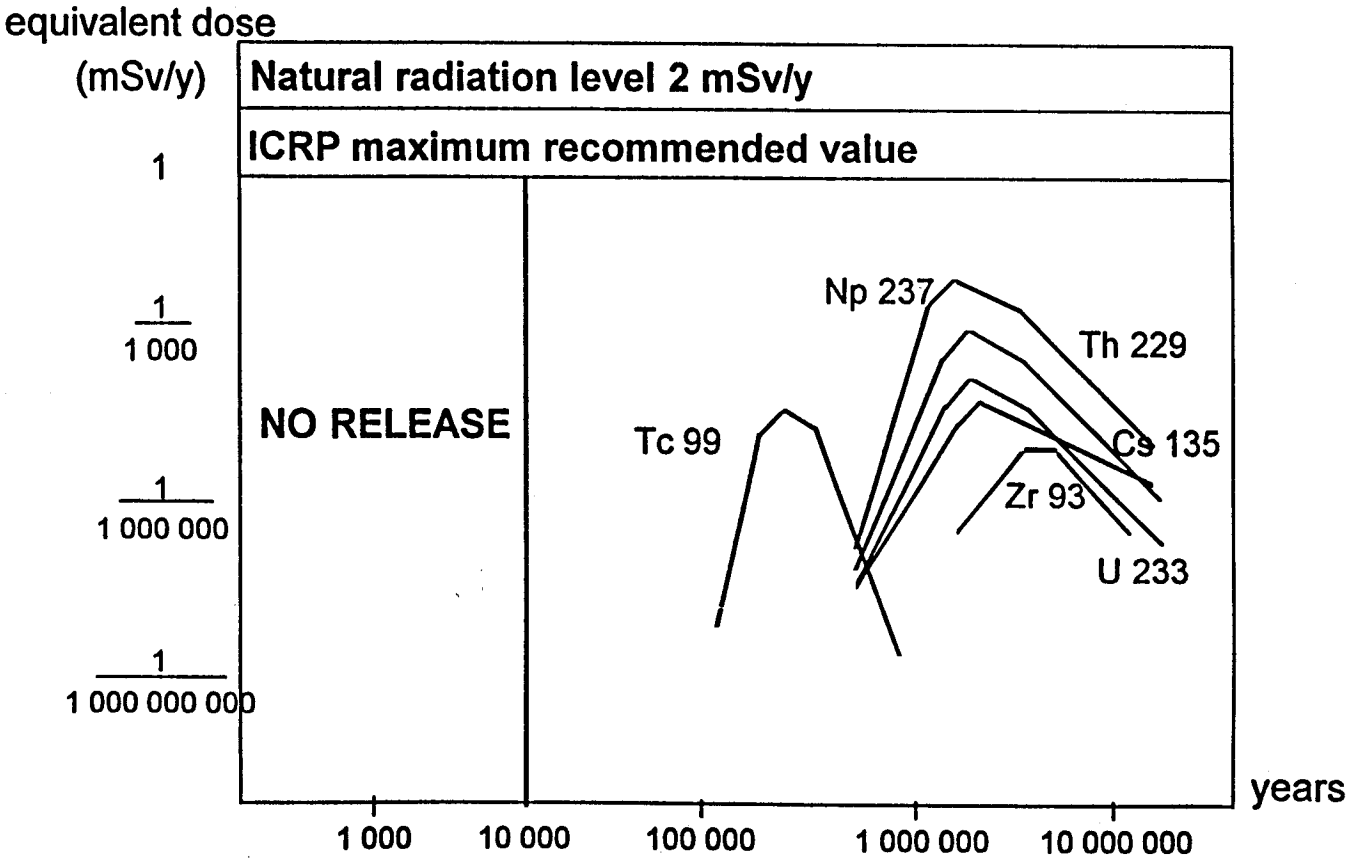
Table 1 - POTENTIAL RADIOTOXICITY OF FUEL

SOURCE TERM AND ITS COMPONENTS
EVOLUTION VERSUS TIME

	10^3 years	10^4 years	10^5 years
TOTAL (Sv/TWhe)	$3.1\ 10^8$	$7,7\ 10^7$	$4.2\ 10^6$
COMPONENTS %			
Pu	90	97	88
Np	/	/	1,3
Am	9.2	2.5	2.7
Cm	0.3	0.4	/
P.F.	$6.0\ 10^{-4}$	$2.4\ 10^{-3}$	$3.2\ 10^{-2}$

RESIDUAL RADIOTOXICITY

Fig.1 AURIAT site (concept A) standard scenario



Volumes of Final Residues Generated in UP3 (m3/tU)
(Long-lived waste after conditioning)