

INCINERATION OF ACTINIDE TARGETS IN A PRESSURIZED WATER REACTOR SPIN PROJECT

A. Puill, J. Bergeron

Commissariat à l'Energie Atomique
DIRECTION DES REACTEURS NUCLEAIRES
Département de Mécanique et de Technologie
Service d'Etudes de Réacteurs et de Mathématiques Appliquées

Centre d'Etudes de SACLAY - 91191 Gif sur Yvette Cedex - FRANCE
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ABSTRACT - The ability of Pressurized Water Reactors (PWR) with uranium fuel to limit the inventory growth of minor actinides (²³⁷neptunium and americium) produced by the French nuclear powerplants is studied. Targets containing an actinide oxide mixed to an inert matrix are loaded in some reactors. After being irradiated along with the fuel, the target is specially reprocessed. The remaining actinide and the plutonium which is produced, added to fresh actinide, are recycled in new targets. The radiotoxicity balance, with and without incineration, is examined, considering that only the losses coming from the target reprocessing treated as waste. A scenario arbitrarily based on 18 years of operation results in a reduction of the radiotoxicity of the waste by a factor between 10 and 20, depending on the actinide considered.

I. INTRODUCTION

The management of long lived radioactive waste resulting from the stock piling of spent fuel or to losses during reprocessing is one of the main problems of the nuclear industry. Various options are being studied: Direct deep burial, with or without simple reprocessing, advanced reprocessing, incineration of plutonium, minor actinides and fission products. Our study is limited to the incineration of the main long-lived minor actinides: neptunium-237 and americium. Much work had been carried out in the past (1). After a period of reduced activity due to difficulties encountered studies have resumed especially after the initiation of the OMEGA project by Japan. Many, more or less futuristic concepts have been proposed (2) which require important research and development work. Our approach is restricted to the use of existing Pressurized Water Reactors (PWR) with little or no modifications. It is part of the framework of the general study of reactor

based incineration (3), which is itself part of a wide programme called SPIN (a French acronym meaning improved separation and incineration).

The potential for incineration in water reactors seems limited because of the value of the neutronic flux: 10^{14} neutrons/cm²/s and of the poor fission per capture ratio for the isotopes of interest: a few percent. These reactors, however, which have reached a high level of industrial maturity, represent an enormous potential of neutrons with a life expectancy of many more decades. These reactors, already used for the recycling of plutonium, can, today, contribute to limit the growth in the inventory of minor actinides pending the implementation of better performing facilities: the FBR, high flux reactors or accelerators.

Targets containing the minor actinides are irradiated in the reactor. This is the heterogeneous option. It offers advantages compared to the homogeneous option where the actinide is mixed to the fuel: The cycle is independent from that of the fuel in terms of fabrication and reprocessing, the amounts are quite small, less than two tonnes per year for a capacity of 55 GWe and the impact on the operation of the reactor is small. The target is made up of an actinide oxide mixed with an inert matrix to limit both the dose rate during handling and the heat generated after the irradiation time. The feasibility of the operation requires the qualification of the materials under flux, a good behavior in water at over 300 °C in case of cladding rupture, the possibility of reprocessing (solubility), compatibility with the cladding, a good thermal conductivity, a high melting point.

Irradiation of such actinides produces plutonium high in Pu-238 and curium. These elements have an important radiotoxicity, hence the need to reuse them. For plutonium, two options are possible: a) Recycling with the plutonium

from the fuel avoiding too high proportions of Pu-238 b) recycling in new targets: the option chosen in this study. It is not, at this time, under consideration, to recycle the curium. The main isotopes, 242 and 244, have relatively short half lives, 163 days and 18 years respectively, leading to plutonium which can be recycled. Otherwise it will be stored as is or sent through other incineration options. Neptunium and americium will be treated separately since their properties are fundamentally different (dose rate, amount produced, fission gas, conductivity, etc ...).

A parametric study of targets loaded in guide-tubes of a PWR fuel assembly for an infinite medium makes it possible to establish the influence of the actinide content and the geometry of the target on the incineration efficiency. A scenario involving the recycling of targets containing neptunium-237 in a 900 MWe PWR with four-batch core management has been studied. The targets remain 4 years in the core. The number of targets and their composition are defined such as not to disturb the operation of the reactor but an over-enrichment of the fuel is necessary to maintain the fuel cycle time. About one of every four reactors must be dedicated to incinerating neptunium and americium. Assuming that only the losses from target recycling becomes waste, the radiotoxicity is reduced by a factor of 10 to 20 depending on the actinide compared to direct storage.

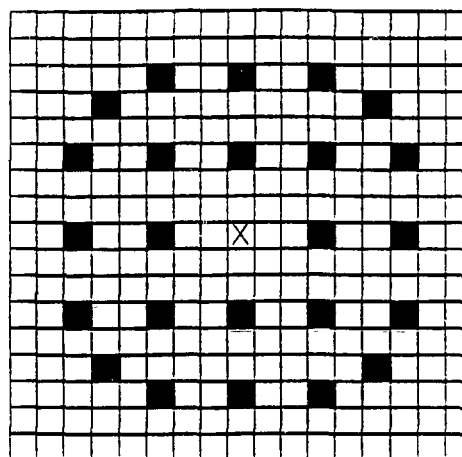
Another option has been studied: The introduction of an all-actinide assembly in the center of the core where the uranium assembly begins its fourth irradiation cycle. This assembly is composed of americium pins at the center and neptunium pins at the periphery to limit the dose rate during handling. This configuration allows increasing the reactivity of the central region. Its stay in the core is limited only by the corrosion of the cladding and the release of fission gases. High incineration rates can be achieved with a high percentage of fission.

II - PARAMETRIC STUDY

A cluster of 24 rods containing actinide is loaded into the guide tubes of a fuel assembly managed on a four-batch basis. Enrichment of the fuel is 3.70% (Figure 2.1).

The actinide core consists of neptunium oxide NpO_2 (or americium oxide AmO_2) combined with an inert matrix of alumina Al_2O_3 (other matrices could be used). The transport calculation was carried out with APOLLO (4) on one fuel assembly.

In the fission chain common to the target and the fuel, 20 heavy nuclei, of which 10 are fissile,



- UO_2 rod (264)
- Guide tube with target (24)
- Instrumentation thimble (1)

Figure 2.1 - 900 MWe PWR fuel assembly with actinide cluster

and 77 fission products are specified. The neptunium-237 and the americium-241 are self-shielded and derived from the JEF2 library. The other isotopes originate from the CEA 86 procedure (5).

A year of irradiation (one PWR cycle) results in depletion which varies, depending on the initial NpO_2 content between 10% (content 100%) and 25% (content 5 to 10%). Reducing the radius of the target core increases depletion but the effect of this parameter remains slight, at least under the conditions of the calculation where a single point in the actinide is considered. The results for neptunium-237 are given in Table 2.1.

The results for americium (americium-241 to 243, isotope contents corresponding to pressurized water reactor fuel with 42,000 MWd/t irradiation and cooled for 5 years) are given in Table 2.2. A single core radius is considered (0.413 cm). A year of irradiation results in depletion varying between 17% (content 70%) and 41% (content 20%). The range of initial AmO_2 content was intentionally limited due to problems relating to fabrication (dose rate) and irradiation (conductivity, swelling, etc.) of the targets.

The calculations were carried out with a fuel enriched with uranium-235 by 3.70%. In view of the annual incineration factors, the targets will need to stay in the core for a number of years.

The significant differences in the rate of disappearance (a factor of 2 for a content of 20%) can be explained by the value of the neutron parameters of the two materials. Table 2.3 shows the microscopic absorption cross-section of neutrons at 0.0253 eV and the absorption resonance integral between 0.5 eV and the fission neutron energy.

**Table 2.1 - Effect of initial NpO₂ content and radius of target on efficiency of depletion of neptunium-237 (Al₂O₃ matrix)
APOLLO code - Fuel assembly with a 24-rod cluster**

NpO ₂ content (%) Radius of target core (cm)	5	10	20	42	70	100
0.280	60	120	190	350	550	790
	45	92	197	490	1050	2190
	25.7	24.6 %	22.3	18.4	14.7	11.1
	40.2	38.4 %	35.2	29.3	23.6	18.0
	83.5	81.1 %	76.8	69.1	58.0	46.8
0.310	70	140	200	420	650	900
	55	113	242	597	1285	2680
	25.0	23.8 %	21.6	17.7	14.2 %	10.9
	38.8	37.1 %	34.0	28.3	22.8 %	17.5
	80.0	77.7 %	73.6	66.2	55.6 %	44.9
0.413	100	160	260	650	980	1350
	97	201	428	1058	2276	4760
	24.6	23.4 %	20.8	16.4 %	12.3 %	10.2
	38.4	36.7 %	33.2	26.3 %	20.0 %	16.6
	79.6	76.9 %	71.8	62.1 %	50.7 %	42.1

Table 2.2 - Effect of initial AmO₂ content on efficiency and depletion of total americium - Target core radius - 0.413 cm

AmO ₂ content (%) Al ₂ O ₃ matrix	20	30	38	70
1	690	860 pcm	990	1330
2	403	691 mg/cm	930	2285
3	41.4	35.2 %	30.8	17.0
4	60.0	53.6	47.7	31.0
5	98.0	94.9	89.0	72.5

- 1 - Average initial efficiency of rod (pcm) Uranium-235 = 3.70%
 2 - Initial load of Np-237 in mg/cm of length Soluble boron = 600 ppm
 3 - Depletion to 12,000 MWd/t, 1 year. % 5 - Depletion to 60,000 MWd/t, 6 years. %
 4 - Depletion to 20,000 MWd/t, 2 years. %

Table 2.3 - Effective cross-sections and absorption resonance integrals of minor actinides (in barns) - Reference: BNL 325 (1984)

	Np237	Am241	Am243	Natural boron
$\sigma_a(0.0253 \text{ eV})$	176 ± 3	587 ± 12	75 ± 2	767 ± 8
$I_a(>0.5 \text{ eV})$	650 ± 50	1425 ± 100	1820 ± 70	345

III - MULTI-RECYCLING OF TARGETS IN A PRESSURIZED WATER REACTOR

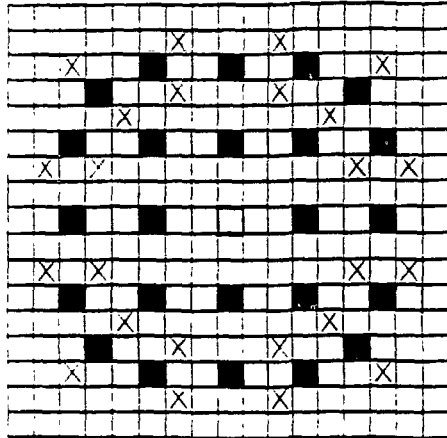
In the parametric study, a cluster of 24 actinated rods is loaded in the guide-tubes of the pressurized water reactor fuel assembly. For a 43 GWd/tHM irradiation of the fuel assembly, linear power levels of 130 W/cm are attained in the target (fission of the plutonium produced). With the current pressurized water reactor guide-tube design, cooling of the rod is not guaranteed under all operating conditions.

Without rejecting the previous solution which has advantages as regards handling of the targets but requires a new guide-tube design, the study is continued by loading targets in fuel assembly in

lieu of uranium dioxide rods, with the same geometry as the latter. The target rods are removed after unloading the fuel assembly and sent to a special reprocessing plant. The feasibility of dismantling pressurized water reactor fuel assemblies has been demonstrated, but is nevertheless a complicated operation.

The position of the 24 targets in the fuel assembly is shown in Figure 3.1. The 24 guide-tubes and the instrumentation tube were retained. Enrichment of the fuel with uranium-235 is increased to 4.5% to ensure an equilibrium cycle length of around 280 equivalent full-power days. Another possibility would be retaining 3.70% enrichment associated with three-batch core measurement: poisoning is compensated by the

loading of additional fresh fuel assemblies. To obtain a useful degree of incineration and to keep enrichment of uranium below 5% (the current limit of the enrichment plant), the concentration of actinide is limited: 70% for NpO_2 and 30% for AmO_2 . The latter has a high equivalent dose rate and presents problems of swelling in the flux due to the alpha decay of the produced curium. The inert matrix used in the calculations is Al_2O_3 .



- UO_2 rod (240) - e U235 = 4.5 %
- Guide Tube (24)
- Actinide target (24)
- Instrumentation thimble (1)

Figure 3.1 - Pressurized water reactor assembly with actinide targets replacing fuel rods

At equilibrium, 40 fresh 4.5% enriched fuel assemblies are loaded into the core each year. Twelve of them contain 24 actinide targets. At equilibrium, in four-batch management, 48 fuel assemblies with targets are present in the core. Every year, 12 x 24 targets are unloaded. The

two-dimensional diffusion calculations were carried out rod-by-rod using the CRONOS 2 program (6).

The unloaded targets are sent to a special reprocessing plant. The fission products and the curium are eliminated. Fresh actinide is added to the resulting products (neptunium, americium, plutonium) and new targets are fabricated for further irradiation over four cycles. In our study, all the plutonium formed by transmutation is recycled in the targets.

Three successive phases of incineration in a pressurized water reactor are examined. Each lasts four years, followed by successive reprocessing and fabrication stages lasting two years. Each phase is considered to be in equilibrium with the annual loading/unloading of 12 x 24 targets per reactor. The annual balance per pressurized water reactor is shown in Table 3.1 for neptunium and Table 3.2 for americium.

The average depletion of neptunium per year reaches 30% of the quantity loaded. The isotopic composition of the plutonium formed varies only slightly between the second and third cycles (16% fissile plutonium and 80% plutonium-238). Unlike MOX fuel, the quantity of plutonium does not drop during multiple cycles. The fissile plutonium content increases relative to all heavy nuclei from 3% to more than 6%.

Annual americium consumption at equilibrium is roughly 60% (70% for Am-241) but also produces plutonium and curium. Only the plutonium is recycled in the targets. The curium produced (20% of the americium loaded) is a short lived waste which will have to be managed. An incineration scenario with in core storage of minor actinides can be conceived.

Table 3.1 - Annual balance per pressurized water reactor for the Neptunium - 70% of NpO_2 in the targets (Phases at equilibrium, annual throughput of 12 x 24 targets)

Phase (4 years of irradiation + 2 years ex-core)		1	2	3
Np-237 loaded	(kg)	236	274	310*
NpO_2 concentration	(%)	70	66	72
Total Pu loaded	(kg)	0	79	72
Np-237 unloaded	(kg)	149	190	218
Pu unloaded	(kg)	81	147	150
Fissions	(kg)	6	16	14
Np-237 consumption	(%)	36,8	30,6	29,7
$\text{Pu}_{\text{fiss}} / (\text{Pu}_{\text{tot}} + \text{Np})$ at beginning of phase	(%)	0	3,0	3,0
$\text{Pu}_{\text{fiss}} / (\text{Pu}_{\text{tot}} + \text{Np})$ at end of phase	(%)	4,7	6,9	6,3
$\text{Pu}_{\text{fiss}} / \text{Pu}_{\text{tot}}$ at end of cycle	(%)	13,3	15,7	15,7
NPu rod max power (W/cm) at beginning of phase		0	102	105
NPu rod max power (W/cm) at end of phase		138	205	194

* Target doubled

Table 3.2 - Annual balance per pressurized water reactor for the Americium - 30% of AmO₂ in the targets (Phases at equilibrium, annual throughput of 12x24 targets)

Phase (4 years of irradiation + 2 years ex-core)		1	2	3
Am 241 loaded	(kg)	51,9	69,1	76,7*
Am 243 loaded	(kg)	19,6	33,0	34,9
AmO ₂ initial concentration	(%)	30	36,9	39,7
PuO ₂ initial concentration	(%)	0	10,1	9,6
Total Pu loaded	(kg)	0	27,9	27,2
Total Am added	(kg)	0	82,3	91,8
Am 241 unloaded	(kg)	9,3	20,0	23,6
Am 241 consumption	(%)	82	71	69
Am 243 unloaded	(kg)	10,5	19,8	21,2
Total Pu unloaded	(kg)	28,2	55,1	57,5
Total Pu production	(kg)	28,2	27,2	30,3
Total Cm production	(kg)	14,2	19,8	21,0
Pu fission/heavy nucl. at beginning of phase	(%)	0	3,0	3,0
Pu fission/heavy nucl. at end of phase	(%)	6,3	7,4	7,0
Max power in target at beginning of phase	(W/cm)	0	66	67
Max power in target at end of phase	(W/cm)	135	193	194

* Target doubled

It begins at year zero with a 58 GWe installed power (25% of the power coming from reactors recycling MOX fuel representing a third of the total load). The annual production of actinides estimated to be 600 kg of neptunium-237 and 900 kg of americium is completely absorbed by a number of incinerating reactors representing 25% of the installed power.

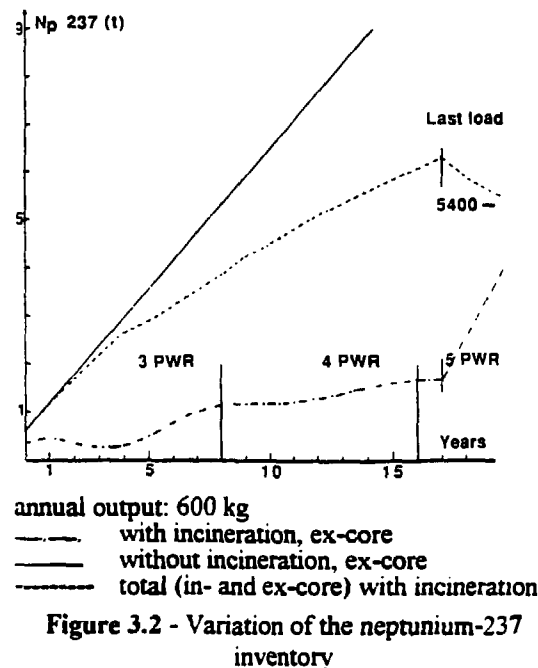
In the scenario adopted for the study, the last loading of the targets takes place at the beginning of year 17, and is followed by a gradual return to conventional management. From this point, the materials produced are directed towards the other means of incineration. However, the targets present in the cores continue to be irradiated. The last batch of targets is unloaded at the end of the 20th year of the operation. The quantity of unrecycled neptunium remaining is 5400 kg as compared to the 11,400 kg loaded between years 0 and 17. Thus approximately 53% is consumed (6000 kg).

For americium, the efficiency is better, since the targets are less loaded and the absorption parameters are higher: 76% of the total americium produced is consumed during this period. Figures 3.2 and 3.3 show the variation of the inventories of neptunium-237 and americium, with and without incineration in and out of the core.

The scenario could of course be extended beyond the 18th year with targets of the fourth generation or more. Figures 3.2 and 3.3 show how the ratio between total neptunium with incineration and without incineration diminishes over the years.

The efficiency could be further increased by reducing the target load, which results in

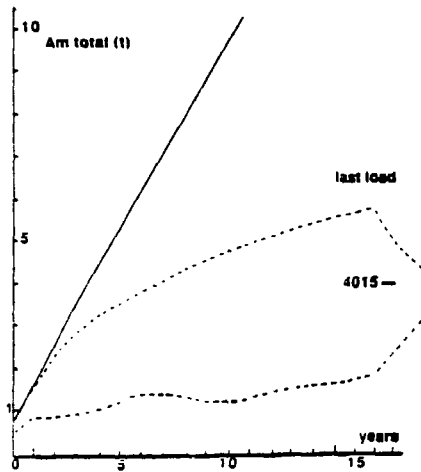
reduction of enrichment, which is also beneficial, but requires more reactors for a given output of material. A financial optimum is to be determined.



IV - OPERATION AND SAFETY

The relatively high absorption of the actinides requires additional enrichment of the fuel from 3.7% to 4.5% to preserve the cycle length. However, the loss of reactivity is reduced relative to a bare fuel assembly as plutonium is formed.

The increase in enrichment with uranium-235 reduces the absolute value of the Doppler coefficient, but the effect is slight (less than 7%)



annual output: 900 kg
 ——— total (in- and ex-core) with incineration
 - - - - without incineration, ex-core
 with incineration, ex-core

Figure 3.3 - Variation of the total americium inventory . 12 PWR

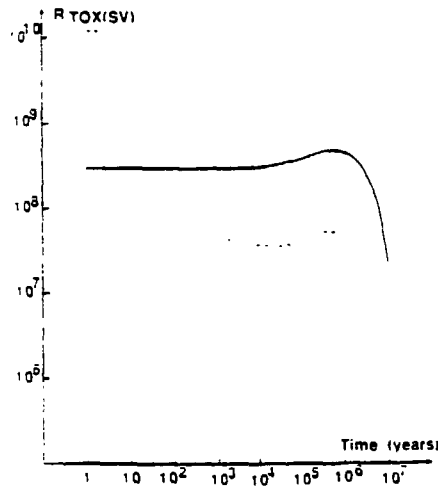
The presence of absorbent targets replacing the uranium makes the moderation coefficient more negative but the values remain within safety limits.

The formation of plutonium in the targets results in a void coefficient penalty. It is checked that this remains negative in the event of draining.

The analysis covers targets containing 70% of NpO_2 and 30% of AmO_2 .

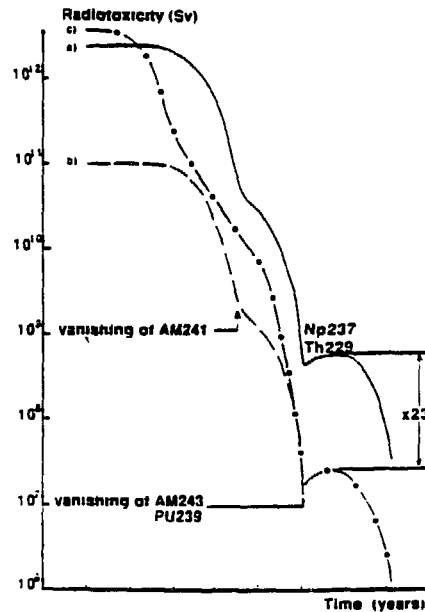
V - RADIOTOXICITY

An assessment of the radiotoxicity reduction factor (activity x ingestion dose factor) resulting from incineration is carried out. Loading of targets is stopped at the end of the 18th year. The targets present in the core continue to be irradiated and will be reprocessed. A comparison is made between the case in which all the actinides produced are stored and that of incineration in which only the losses (neptunium: 10%, plutonium: 0.3%) resulting from reprocessing of the targets are stored. The materials remaining at the end of the in-core operation (i.e. neptunium, americium and plutonium) are processed using new methods of incineration. The difference in radiotoxicity in the two processes is shown in Figures 5.1 and 5.2. The reduction due to incineration of neptunium corresponds to a factor of around 10 beyond 1000 years, assuming that only the losses are considered as waste and the other means of incineration take over for the remaining materials.



——— 11400 kg of Np 237
 - - - - 1140 kg of Np 237
 22 kg of Pu (80 % Pu 238)

Figure 5.1 - Variation of radiotoxicity - Neptunium-237



a) 17100 kg of total Am ———
 b) 704 kg of total Am - - - -
 + 28 kg of Pu (64 % Pu 238)
 c) 704 kg of total AM
 32 kg of Pu
 1498 kg of cm 244

Figure 5.2 - Variation of radiotoxicity - Americium

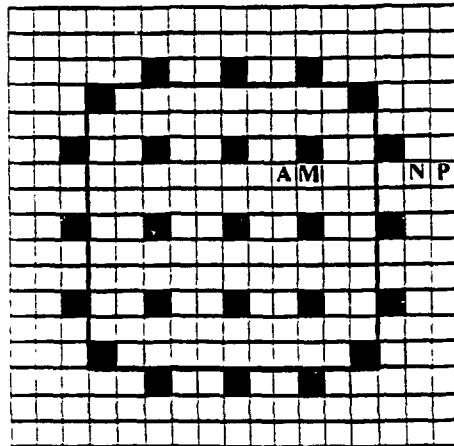
Two options are proposed for americium as a function of the type of curium management:

- Temporary surface storage of the curium pending a new incineration process. In this case it is not included in the deep storage radiotoxicity inventory (figure 5.2 - curve b)

- Direct storage of the curium produced with losses from the target recycling. The analysis of the decay of the isotopes shows that no more curium-242 remains at year 25 (it is transformed into 940 kg of Pu-238) but 1500 kg of Cm-244 remains (as well as 680 kg of Pu-240). Curve c takes this curium into account. The plutonium produced, minus 0.3% due to losses, is destined for the new incineration processes. The radiotoxicity of the americium component of the waste coming from advanced recycling is reduced by a factor of 20 (after 200 years, taking into account the curium produced) assuming that only the losses from target recycling are destined for deep storage.

VI - DEDICATED ALL-ACTINIDE FUEL ASSEMBLY

Another process has been investigated, involving a dedicated fuel assembly exclusively consisting of actinide rods. A mixed fuel assembly containing americium rods in the centre and neptunium rods at the periphery is postulated (Figure 6.1).



156 rods with 20 % NpO₂
 108 rods with 30 % AmO₂
 ■ Guide Tube (24)

Figure 6.1 - Mixed central assembly

The neptunium rods are less active in handling and mask the dose rate of the americium rods. The geometry of the rods is identical to that of fuel rods. The number of rods of each category and the actinide contents have been selected to obtain similar masses and incineration rates. This fuel assembly is loaded in the centre of the core, at the position of the uranium fuel assembly beginning a fourth cycle. The objective is to demonstrate incineration in a pressurized water reactor without disturbing operation of the reactor. This fuel

assembly can stay in the core for six years (limited only by corrosion of the zircaloy cladding and the release of fission gases, a longer stay being conceivable with stainless steel cladding and expansion chambers). 3.7% enrichment is retained, and the presence of this fuel assembly results in a penalty of approximately 7 equivalent full-power days on the length of the cycle.

The gradual formation of plutonium results in an increasing release of power as a function of irradiation: 75 watts/cm in the hot neptunium rod. The material balance for the two types of targets is shown in Table 6.1.

Table 6.1 - Mixed central fuel assembly. Material balance after 6 years of irradiation

Americium targets: 108 rods	
(30 % AmO ₂ + 70 Al ₂ O ₃)	
Am-241 loaded	19,5 kg
Total Am loaded	26,9 kg
Am-241 unloaded	6,2 kg
Total Pu unloaded	9 kg
Pu-238 unloaded	6,3 kg
Fission	2,3 kg (12 %)
Am-241 incineration	13 kg (68 %)
Neptunium targets: 156 rods	
(20 % NpO ₂ + 80 % Al ₂ O ₃)	
Np-237 loaded	24,3 kg
Np-237 unloaded	8,2 kg
Total Pu unloaded	12,1 kg
Pu-238 unloaded	9,2 kg
Fission	3,9 kg (16 %)
Incineration	15,1 kg (67 %)

The masses loaded (Am + Np) correspond to the annual production of two pressurized water reactors.

In six years, consumption is 70% but combined with the formation of plutonium (40% of the initial mass) and curium, which are materials which have to be subsequently dealt with.

VII - CONCLUSION

Contrary to current opinion, it is shown that multiple recycling of actinide targets in a given number of pressurized water reactors (25% of installed power) should, today, limit the increase in the volume of matter as well as the radiotoxicity of the waste destined for storage.

The amount of matter as well as the number of targets (5000 per year) are limited. The process is part of a separate cycle independent from the fuel cycle.

The rates of disappearance at fuel assembly level are relatively high, being 10 to 15% per year for neptunium-237 and 15 to 40% for americium. All the plutonium formed is recycled in the targets.

A scenario carried out over 18 years of operation for 58 GWe installed capacity leads to a 53% (6000 kg) consumption of neptunium-237 and of 76% (13000 kg) of americium produced during that time. Assuming that only the losses from the target recycling are to be stored as waste, the potential radiotoxicity is reduced by factors of 10 (neptunium) to 20 (americium) after a few hundred years. Continuing the process beyond 18 years would lead to a smaller and smaller increase in the global actinide inventory.

Although slightly degraded, the reactivity coefficients remain within the safety limits. The study assumes the feasibility of industrial separation of the long-lived actinides in spent fuel, fabrication of targets with satisfactory in-core characteristics, and the reprocessing of high-activity materials containing large proportions of plutonium-238. Investigations are being continued, particularly with a view to improving the efficiency of incineration of neptunium-237. Also noteworthy is the aspect of interim storage in-core of a large proportion of the material, which is advantageous in terms of radiological protection. Despite the complexity of the operations, the multi-recycling of actinide targets in a pressurized water reactor represents a real contribution to solving the problems of long-lived waste, and one which can be implemented in the near future. The irradiation of targets in the OPERA loop of the OSIRIS pile at the CEA, the ACTINEAU experiment represents the first step.

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