NUCLEAR WASTE TRANSMUTATION :
PHYSICS ISSUES AND POTENTIAL IN NEUTRON FIELDS

M. Salvatores[*], A. Zaetta[**], M. Delpech[**], C. Girard[**], I. Slessarev[**], J. Tommasi[**]
* CEA/DRN - Cadarache
** CEA/DRN/DER/SPRC - Cadarache

ABSTRACT

1. Introduction

A deep geological repository for safe long-term storage of long-lived radioactive materials (waste) arising from nuclear fuel irradiation in reactors is a need generally accepted, whatever the strategy envisaged for further use of the irradiated fuel (e.g. : reprocessing and re-use of uranium and plutonium ; no reprocessing and final disposal).

To assess the impact on the environment of a waste repository, one is lead naturally to consider the impact of radiation on man and to define the radiotoxicity of the different isotopes.

The toxicity of the materials stored in a repository is function of time and at a given time is the sum of the activities of each radionuclide multiplied by appropriate danger coefficients.

This time dependent sum R, is a source of "potential" radiotoxicity. It has been pointed out /1/, that R does not measure "risk", which has to take into account "actual pathways and probability of radioactive release to the biosphere".

It is well understood that (e.g. in the case of spent PWR fuel) the main contributors to R are actinides, Pu being the main component (see table I). In the case of risk, the situation is by far more complex and dependent on the modelisation of different geological environments.

In the analysis made in ref. 1, the predominant role of Tc-99, I-129 and Cs-135 has been pointed out.

The same analysis also stresses that actinides will be by far less relevant with respect to the highly soluble and mobile fission products.

2. Goals and criteria

Waste management strategies are under discussion in most countries. Local conditions and constraints induce different perspectives and goals. Moreover, underlying hypothesis on the future of the fission energy option (i.e. phase-out in the next century, or extension beyond next century) imply that transuranics can be viewed as waste or as fuel.

In absence of a unique approach, R and D efforts are aimed to investigate the possible means of reduction both of the potential source of radiotoxicity and of the dose risk. Typical examples are the OMEGA program in JAPAN and the SPIN program in France (see for example papers at the GLOBAL'93 Conference, Seattle 1993).

Moreover, to evaluate the benefits of the envisageable strategies and the trade-offs in a cost / benefit analysis, and to define R and D priorities, it would be necessary to have established accepted criteria or standards in order to appreciate the impact of the potential reduction of both R and the dose risk.

This task also turns out to be difficult, and divergent opinions in this field only stress the need for more international cooperation to gain a better mutual understanding.

Among the criteria that have been put forward we can recall :

- Reduction of the long-term radioactivity of nuclear waste sufficiently so that it may be buried safety in a near-surface storage. This implies to reduce (e.g. in the case of spent PWR fuel) the activity from a ~ 5000 Ci/ton level to ~ 0.1 ÷ 0.01 Ci/ton level. This formidable task would require unrealistic decontamination factors in chemical separation processes (if a reprocessing strategy is adopted /2/), or the demonstration of the feasibility of new advanced technologies for transmutation, which have been proposed /3/, but which are far from being demonstrated. Moreover, this criterion can be dangerously seen as an alternative to a deep geological repository.

- Reduction of the long-term radioactivity of nuclear waste below the level of the mined uranium one (necessary to fabricate the fuel) "as soon as possible". This criterion, proposed as early as 1974 /4/, is still of a qualitative type and has been criticized /1/ for not being based on a valid standard for public health and safety. In fact the toxicity of mined uranium ore is due mainly to the decay daughters Ra-226 and Pb-210, which reside in above-ground mill tailings and, in the case of reprocessing, in the depleted uranium which is in principle not a waste, but a material to be re-used. However important the issue of mine-tailings (and of depleted U), their activity (~ 20 Ci/t) does not seem to represent a valid standard against which one should measure the value of a potential reduction of the radiotoxicity source or of the dose risk associated to a deep geological repository.

- Comparison of the resulting inventory of radionuclides in the repository after removal of certain fractions of the actinides (i.e. in a partition - transmutation strategy), with the release limits in the emerging US EPA standards and criteria /5,6/. Very high decontamination factors will be necessary in the chemical processes (i.e. losses) coupled with a very high efficiency in the transmutation techniques /6/. However, the authors of ref. /5/, argue that this approach in the US context could improve the licensability of the repository site.

From these few examples, one can draw the conclusion that at present it is difficult to establish relevant and generally acceptable criteria for the reduction of the radiotoxicity of wastes.

A more pragmatical approach is to investigate a large range of radiotoxicity reduction possibilities in realistic scenarios to point out priorities for R and D and to verify how a particular strategy can help to cope with uncertainties related to modelling of geological sites, with concerns related to abnormal scenarios (e.g. intrusion) in the assessment of a repository safety, and with the public perception of the waste management issue.

3. Transmutation issues from a physics point of view

In order to investigate radiotoxicity (and dose risk) reduction possibilities in realistic scenarios, one has to quantify the potential and the effects of different partitioning - transmutation approaches.

In what follows we will be concerned only with the transmutation issues.

Two main issues will be addressed, namely a) transmutation potential of different types of systems, in order to evaluate the transmutation effectiveness both for transuranics and for long-lived fission product and b) consequences on the transmutation efficiency of realistic scenarios (what reactor types for transmutation in what reactor park).

3.1 Transmutation potential

From a physics point of view, the full transmutation of one isotope in a neutron field has to be associated to the transmutation of the whole isotope family, to account for the possible build-up of other isotopes, which also have to be transmuted.

To assess the feasibility of such transmutation process in a reactor one has to consider constraints :
- neutronics : are there enough neutrons for the task of full transmutation ? What is the maximum transmutation rate and what this rate does depend on ?
- safety : how the presence of the nuclei to be transmuted does influence the core safety parameters ?
- fuel cycle : how affected decay heat, neutron sources, α activity,
- technologic : fuel fabrication and reprocessing issues to be accounted for,
- economics : the "cost" of the transmutation.

We will consider here the neutronics constraints, expressed in terms of demand D_i of neutrons per fission for full transmutation of isotope i (or isotope mixture i) in a pseudo-equilibrium state /7/.

In ref. /7/, it has been shown that D_{TRU} depends the TRU-type of isotope (or isotope vector), on the flux level (moderately) and on the spectrum type (strongly).

Typical examples of D values (neutrons / fission) are given in the table below :

TRU-nucleus	Type of n spectrum		
	Standard PWR	Standard FR	Thermalized spectrum ($\phi \simeq 10^{16}$ n/cm^2.s)
Pu-239	- 0.70	- 1.50	- 1.00
Am-243	+ 0.30	- 0.60	+ 0.20
TRU$_{PWR}$ - "vector"	- 0.17	- 1.17	- 0.60

Negative D values indicate "neutron production". Knowing that a neutron consumption of $D_c \simeq$ + 0.3 n/fission is required to compensate parasitic absorption and leakage, the previous table underlines that to transmute TRU there is a shortage of neutrons in a LWR (i.e. need to increase the fissile content), but not in a standard FR and, at a lesser extent, in a high flux thermalized neutron spectrum. As a consequence, transmutation rates are restricted by the neutron balance and by the power of the system. The latter is the only restriction in a FR.

This fact indicates the TRU transmutation is feasible in standard FRs and that, from a neutronics point of view, no extra neutron sources are needed.

As far as "long-lived dangerous fission-products" (LLDFP), their transmutation rate is proportional to the ratio :

$$r \div \frac{G}{D_{LLDFP}}$$

where G is the neutron surplus per fission available in a particular system : $G = -(D_{Fuel} - D_c)$.

For example, $D_{Tc-99} \simeq 1$ and $G \simeq 0$ for a LWR, $G \simeq 0.3$ for a FR with BR < 0.5). It is then clear that there is limited potential in standard reactors to obtain significant LLDFP transmutation rates and that an additional source of neutrons may be required. This additional source can be provided by the so-called hybrid systems (i.e. subcritical system + spallation by accelerated protons).

The maximum additional n source per fission (μ) that one can get, is dependent on the fraction f of the energy produced in the subcritical system that is used to produce the additional neutron source :

$$\mu = \frac{f}{K} \quad \text{where} \quad K = \frac{E_p}{E_{fiss} \cdot z \cdot \eta}$$

with : E_p proton energy ; z number of neutrons per proton ; η efficiency of the energy transformation into neutrons.

For example if $E_p \simeq 1.5$ GeV, $z \simeq 50$ and $\eta \simeq 0.2$, we have $K \simeq 0.75$. This means that if all the energy produced in the subcritical system is used to feed the accelerator to produce additional neutrons, in that case $\mu_{max} = 1.3$ n/f.

The maximum additional neutron source per fission corresponds to a high subcriticality :

$$k_{eff} = \frac{\nu}{\nu + \eta}$$

As a as proton current I, we can define /7/ a simple relationship between μ and I :

$$\mu \approx \frac{z\,I}{5} \frac{mA}{MWth} \approx 0.1\,I\,\frac{mA}{MWth} \quad \text{(for } z \approx 50)$$

This relationship simply illustrates the need for an intense proton current if one wants to achieve a high value of μ (i.e. in a strongly subcritical system).

Finally we get for an hybrid system a total neutron surplus G_H :

$$G_H = G_S + \mu$$

where G_S is the neutron surplus in the subcritical system as defined previously, and which is maximum for a fast neutron spectrum, whatever the neutron flux level.

3.2   Realistic scenarios for transmutation in a reactor park - Pu recycling

If one turns now to the problem of how to achieve in practice a significant reduction of the potential source of radiotoxicity R, from the data in table I, one sees that the first priority is the Pu management. From a purely theoretical point of view, one can imagine to reduce the contribution of Pu to the waste radiotoxicity to the level of the Pu losses in reprocessing (e.g. to ≈ 0.3 %). One would obtain the results of table II. Starting from those data one could further evaluate the benefits of a selective separation and elimination of Am, Np, Cm, using theoretical decontamination factors.

However, this procedure gives only a qualitative insight. In fact, the Pu which is left out (except for losses) from the values of table II, has to be used in actual reactors. Its recycling should be feasible (core and safety constraints) but also is should be such that one would also minimize the inevitable further production (ΔMA) of minor actinides, whose contribution will more or less spoil the results shown in table II.

Moreover, if Pu utilisation is seen mainly in terms of Pu consumption (ΔPu), it is also useful to consider the ratio ΔMA/ΔPu, as an indicator of effectiveness in the use of Pu in order to minimize MA productions.

In practice, if one considers three types of 1400 MWe LWRs fully fuelled with MOX (Pu vector as issued from LWR UOX enrichment 4.5%, Burn-up 55 GWD/t) with different moderator / fuel ratios (namely $V_m/V_f = 1.1, 2, 3$), one obtains at the end of an irradiation of 55 GWd/t, after 3-years cooling time, and expressed in kg/TWhe

V_m/V_f	ΔPu	ΔMA	$|\Delta MA/\Delta Pu|$
3.0	- 68	+ 11	0.15
2.0	- 64	+ 16.5	0.26
1.1	- 54	+ 21.8	0.40
Standard UOX-PWR	+ 29	+ 3.8	/

If one considers two EFR-type of FR (without fertile blankets) and with average core enrichment e = 20% and e = 30% respectively one obtains (in kg/TWhe)

V_m/V_f	ΔPu	ΔMA	$|\Delta MA/\Delta Pu|$
e = 20%	- 17	+ 7	0.41
e = 30%	- 42	+ 6	0.14

The results of the previous two tables confirm what is expected on physical grounds /8/, namely that FRs allow to use Pu with a minimized production of MA and the interest of a LWR MOX reactor with high moderation

However, only one recycling has only marginal consequences on the radiotoxicity reduction, as it can be seen from the following table, where it is given the ratio of the radiotoxicity (R/TWhe) of the loaded MOX fuel to that of the unloaded fuel, and its evolution with time

		$t = 10$ y	10^2 y	10^3 y	10^4 y	10^5 y	10^6 y
LWR MOX	$V_m/V_f = 1.1$	1.27	0.98	96	91	87	97
	$V_m/V_f = 2.0$	1.39	0.98	92	77	70	1.01
	$V_m/V_f = 3.0$	1.25	0.76	70	61	53	88
EFR e = 30%		0.92	0.53	67	87	63	54

If Pu multirecycling is considered, the advantage of fast reactors becomes even more striking. In the following tables we give the evolution with multirecycling of the fissile Pu content, ΔPu and ΔMA in the three types of LWR-MOX cores indicated above

The first table concerns a pure autorecycling (the unloaded fuel is loaded back in the core) and the second table gives the same results considering the dilution of one unloaded MOX assembly with four assemblies as unloaded from a standard LWR

	$V_m/V_f = 3.0$			$V_m/V_f = 2.0$			$V_m/V_f = 1.1$		
	cycle 1	2	3	cycle 1	2	3	cycle 1	2	3
Pu fissile content (%)	4.2	7.4	9.1	6.0	7.6	8.6	8.8	9.2	9.3
ΔPu (kg/TWhe)	- 68	- 104	- 122	- 64	- 79	- 90	- 54	- 59	- 63
ΔMA (kg/TWhe)	+ 11	+ 32.5	+ 45	+ 16.4	+ 25.6	+ 32.2	+ 21.8	+ 24	+ 25.9
$|\Delta MA/\Delta Pu|$	0.16	0.31	0.37	0.26	0.32	0.36	0.40	0.41	0.42

Multirecycling with no dilution

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	$V_m/V_f = 3.0$			$V_m/V_f = 2.0$			$V_m/V_f = 1.1$		
	cycle=1	= 2	= 3	cycle=1	= 2	= 3	cycle=1	= 2	= 3
Pu fissile content (%)	4.2	4.8	5.2	6.0	6.8	7.3	8.8	9.0	9.1
ΔPu (kg/TWhe)	- 68	- 78	- 83	- 64	- 71	- 76	- 54	- 57	- 59
ΔMA (kg/TWhe)	+ 11	+ 15.8	+ 18.6	+ 16.4	+ 20.8	+ 23.2	+ 21.8	+ 23.0	+ 23.8
$|\Delta MA/\Delta Pu|$	0.16	0.20	0.22	0.26	0.29	0.30	0.40	0.40	0.40

Multirecycling with dilution

The inevitable degradation of the Pu vector in LWRs (and in particular in the high moderation spectrum $V_m/V_f = 3.0$) indicates that multirecycling should be limited as much as possible in favour of an extended mutirecycling in FRs, which do not show a comparable Pu vector degradation, and which can accept practically any Pu vector in the loaded MOX fuel, without significant penalties in the core performances.

The following table summarizes the comparison of a high moderation LWR-MOX unloaded fuel (after three cycles and with dilution, see above) and a high enrichment FR (EFR-type with $\bar{e} = 30\%$) :

	ΔPu	ΔMA	$|\Delta MA/\Delta Pu|$
LWR-MOX V_m/V_f	- 83	+ 18.6	0.22
FR EFR-type $\bar{e} = 30\%$	- 57	+ 8.4	0.15

3.3 Realistic scenarios for transmutation in a reactor park - MA recycling

In terms of reduction of the potential source of radiotoxicity R, the effect of a realistic Pu mutirecycling is given in figure 1.

We consider three types of Pu recycling.

The first one is a nuclear park where the Pu produced by LWR UOX is reused in a once through high moderated LWR-MOX.

The second one is the same but we suppose three Pu recycling in LWR-MOX.

The last one is based on the same scheme, but we use FRs to burn Pu.

We consider some losses during reprocessing of the spent fuel (after three years cooling) : 0.1 % for Pu, 100 % for MA.

Due to the fact that real Pu recycling increases the production of M.A, we observe :
- the average theoretical gain is about a factor 10 (curve A) ;
- the real gain with the once through LWR-MOX is reduced to a factor 4 (curve B). With three multirecycling the average gain is only a factor 2 ,
- the use of FRs is better, due to the lower production of minor actinides, the gain is quite closed to the theoretical about a factor 7.

If we now consider the MA actinide contribution to the radiotoxicity, qualitatively we can distinguish :
- Am . Am-241 and Am-243 are the predominant contributors to R (see table II) for $t \leq 10^4$ y ;
- Np . Predominant contributor for $t > 10^5$ y .

- Cm   . Significant contributor for t $\simeq 10^4$ y (due to the decay of Cm-244 into Pu-240).

In order to have a quantitative estimation of the potential reduction of the R source term, we have made the following hypothesis on the partitioning process :

- The Np can be separated with a decontamination factor in the range 20÷50 and routed to the Pu stream.
- Am can be separated from the other trivalent components (Cm, Lanthanides) with a decontamination factor in the range 20÷50. The effect of possible contamination with small amounts of Cm and Lanthanides has not been taken into account at this stage.
- Cm is left to decay mixed with Lanthanides. Its recycling in reactors is not envisaged. Eventually, the Pu-240 built-up from decay will be retrieved for mixing and re-use with the major Pu stream.

As far as fuel, the Np is considered homogeneously mixed to the standard MOX fuel. Am is both considered homogeneously mixed to the standard MOX fuel, or loaded in the reactor in form of targets with an inert support matrix (20 % AmO_2 in a MgO matrix). In the case of targets (heterogeneous recycling), the present hypothesis is only conceptual since detailed fuel studies are underway on this subject /9/.

As far as reactor type for MA recycling, a scenario based on previous studies /10/ has been defined which privileges FRs. In fact applied studies /11/, have confirmed the results of the physical analysis, summarized in paragraph 3.1 and detailed in Ref. /8/ : the recycling of MA in MOX fuelled LWRs gives rise to significant demands for increased Pu content, to a worsening of the reactivity coefficients and to significant increase of n sources and activity in the fuel cycle, and this for whatever V_m/V_f ratio (even if cores with a high V_m/V_f seem to be preferable).

In the present case the chosen scenario is as follows.

The nuclear park is constituted by :
- LWR UOX (70 % of the total power of the park),
- LWR MOX (9 % of the total power).
  The Pu produced by the LWR UOX is recycled in LWR-MOX (once through).
- FRs burner (21 % of the total power). The Pu issued from the LWR-MOX is recycled in FRs with all the M.A produced in LWR UOX and MOX
  * Np is recycled homogeneously in the core,
  * Am is recycled in the FRs blanket with Cm 245,
  * Cm 242, 243, 244 are left to decay on Pu isotopes and are then recycled in the Pu flux.

We consider an equilibrium state and some losses during reprocessing : 0.3 % for Pu, 1 % for Np, Am, Cm.

The consequences on the radiotoxicity source term reduction are shown on figure 2.

Here again one can observe that a realistic scenario implies a modification of the theoretical reduction factors, which are associated to theoretical decontamination factors.

The benefits of the MA recycling and of the proposed strategy (including the intermediate storage of Cm) are evident at any time on the R source term and are comprised between a factor of 40 and 80.

3.4   Realistic scenarios for transmutation in a reactor park - LLDFP transmutation

We have indicate previously (paragraph 3.1) that there is a limited potential of standard fission reactors to transmute massively LLDFP. Some results were reported in Ref. /13/, where the concept of moderated sub-assemblies at the periphery of a standard FR was exploited, to benefit both of the excess neutrons leaking out of the core and of the increase of the $\bar{\sigma}_c$ with moderation.

In order to explore the potential of a hybrid system, according to the analysis of paragraph 3.1 (see also Ref /12/), preliminary studies have been performed of a system composed of a proton

accelerator (E_p = 1.6 GeV) coupled to a molten salts subcritical core. The fuel for this core is in the form of $\left[\{TRU_{UOX-LWR}\}\,Cl_3 + Pb\,Cl_3\right]$, which acts both as spallation source and multiplying medium. For a core power of 3000 MWth, K_{eff} = 0.95, proton beam current I = 75 mA and f (fraction of energy used to feed the accelerator) = 0.2, a transmutation rate of Tc-99 (introduced as targets in the core) of 0.7 tons/year has been found, using standard methods and codes (HETC cascade code to produce the n source coupled to a 2D neutron transport code). More studies are under way to further detail the potential of hybrid systems for a larger range of LLDFP.

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TABLE 1
CONTRIBUTION OF EACH ISOTOPE TO RADIOTOXICITY (%)
IRRADIATED PWR FUEL (33.000 MWd/t)
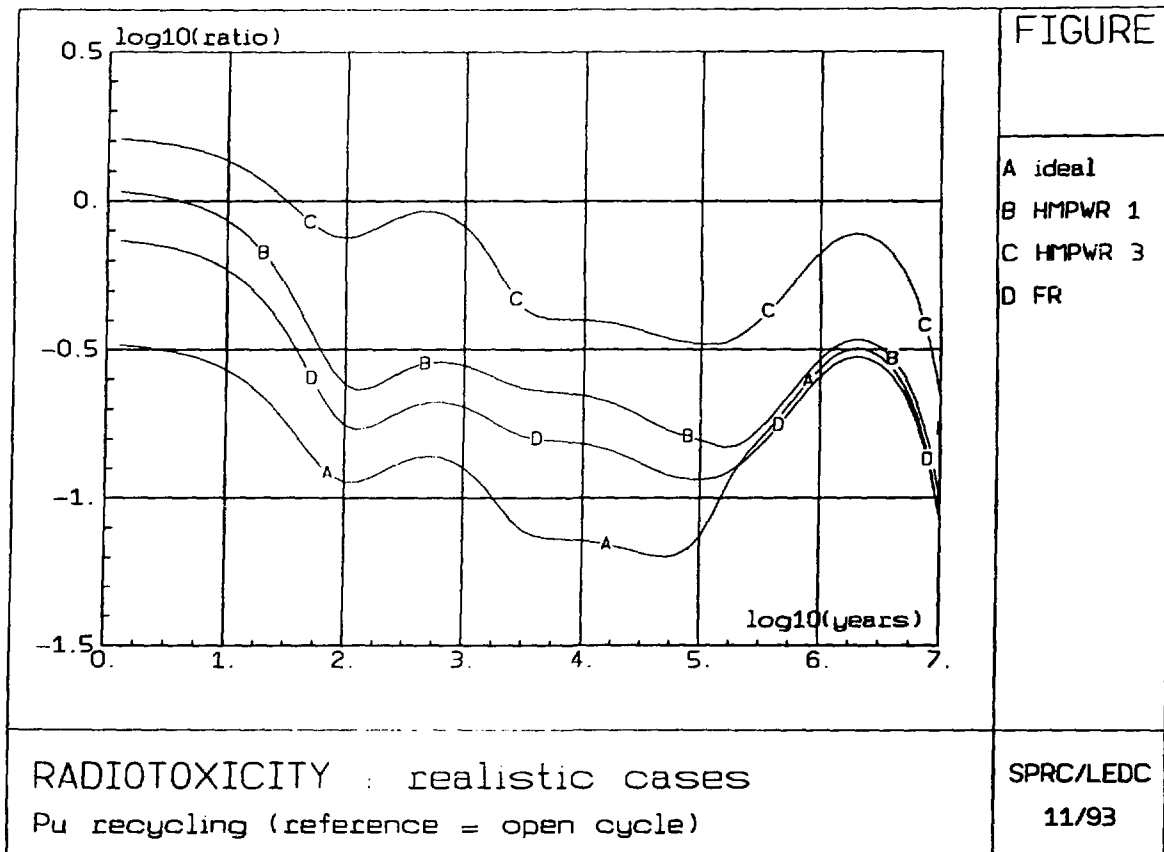*3 years cooling - 100 % Pu, Np, Am, Cm (open cycle)*

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)		$7.37\ 10^9$	$2.07\ 10^9$	$4.75\ 10^8$	$2.6\ 10^7$	$2.57\ 10^6$	$1.90\ 10^5$

TABLE 2

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)	10^2	10^3	10^4	10^5	10^6	10^7
				TIME (y)			
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.	$7.9\ 10^2$	$7.9\ 10^2$	$7.9\ 10^2$	$7.8\ 10^2$	$5.9\ 10^2$	$3.9\ 10^1$

FIGURE 1

RADIOTOXICITY : realistic cases
Pu recycling (reference = open cycle)

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FIGURE 2

(Pu + Np + Am + Cm) BALANCED SCENARIO

RADIOTOXICITY EVOLUTIONS
coherent Pu + M.A. strategy