DATA UNCERTAINTY IMPACT IN RADIOTOXICITY EVALUATION CONNECTED TO EFR AND IFR SYSTEMS'

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ABSTRACT

Time-dependent sensitivity techniques, which have been used in the past for standard reactor applications, have been adapted to calculate the impact of data uncertainties in radiotoxicity evaluations. The methodology has been applied to different strategies of radioactive waste management connected with the EFR and IFR reactor fuel cycles. Results are provided in terms of sensitivity coefficients to basic data (cross sections and decay constants), and uncertainties on global radiotoxicity at different times of storing after discharge.

I. INTRODUCTION

Long-term radiotoxicity reduction appears to be the most important issue related to radioactive waste transmutation.

Because of many studies related to radioactive waste transmutation, the problem of getting better data for minor actinides and fission products has been the focus of renewed interest. Uncertainty impact evaluation and target data accuracy requirements can be used to find the most urgent data improvements to be fulfilled.

In this paper an evaluation will be made of the uncertainty of the calculated radiotoxicity connected to typical EFR (European Fast Reactor) and IFR (Integral Fast Reactor) reactor systems. Time-dependent sensitivity techniques, which have been used in the past for standard reactor applications[1,2], have been adapted to solve this problem.

II. THE RADIOTOXICITY SENSITIVITY AND UNCERTAINTY CALCULATION

We outline here the methodology proposed in Reference 3 to calculate the radiotoxicity R by means of radiotoxicity factor f_i for each nuclide i:

$$R(f_F) = \sum_i n_i(t_F) f_i = \underline{n} \cdot \underline{f}_{t=t_F} \quad (1)$$

Where $n_i(t)$ is the nuclide i density at time t_F. The time interval to be considered starts from t_o, the initial time of irradiation in a reactor. In the time interval (t_o, t_F) one has to consider the irradiation time steps, the cooling times before possible reprocessing, the fuel cycle processing time and the decay period in long-term storage.

The $n_i(t)$ are obtained by solving the standard nuclide transmutation equation:

$$\frac{dn(t)}{dt} - An(t) = -(\sigma_a^i \phi + \lambda_i) n_i + \sum_j (\sigma_{j\to i} \phi + \lambda_{j\to i}) n_j \quad (2)$$

Where ϕ is the neutron flux, a are the (one-group) neutron transmutation cross sections and λ are the radioactive decay constants with the appropriate branching ratios.

The sensitivity coefficients relevant to any variation of a parameter p_k (in our case a cross section or a decay constant) can be obtained (at first order) with a specific application of the generalized perturbation theory in the nuclide field':

$$S_k^R = \frac{dR}{R} \Big/ \frac{dp_k}{p_k} = \frac{p_k}{R} \int_{t_o}^{t_F} \underline{n}^* \cdot \frac{\delta A}{\delta p_k} \underline{n} \, dt \quad (3)$$

Where n* (usually defined as "nuclide importance vector") is obtained by solution of

$$-\frac{d\underline{n}^*}{dt} = A^* \cdot \underline{n}^* \quad (4)$$

With boundary conditions at $t = t_F$.

$$\underline{n}^*(t = t_F) = \underline{f} \quad (5)$$

Where $\underline{f}$ is the vector of components f_i defined previously.

The above formalism bas previously been applied to reactor depletion problems.[1,2]

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Another interesting parameter that can be considered is the radiotoxicity integrated over a given interval of time (e.g. the cumulative dose for prolonged exposure to the radiation source):

$$TIR = \int_{t_0}^{t_F} \underline{f} \cdot \underline{n}\, dt \qquad (6)$$

For such a case it is only necessary to replace equation (4) with the inhomogeneous equation:

$$-\frac{dn''}{dt} = A \cdot E'' + f \qquad (4')$$

Once the sensitivity coefficients of Eq. (3) are available, given a covariance matrix D_{xy} on the data r, the uncertainty I_R associated with the radiotoxicity R (or with the time integrated radiotoxicity TIR) is given by:

$$I_R = \left\{ \sum_{xy} S_x^R D_{xy} S_y R \right\}^2 \qquad (7)$$

III. CALCULATIONAL HYPOTHESIS

The main application we have considered are typical EFR and IFR reactor systems and associated fuel cycles.

The decay chain used in solving the transmutation equation is quite complex and goes from Pb-210 to Cm-248.

The radiotoxicity is expressed in terms of cancer doses. The related radiotoxicity factors f_i are calculated using values provided in Ref. 5. These radiotoxicity factors assess the hazard (consequences of exposure) for the actinide elements; hazard analyses assume complete exposure to the radioactive inventory and do not evaluate mitigating effects (e.g., shielding, barriers to release, etc.). In this paper, the fatal cancer dose measure developed by Cohen[5] is utilized to quantify the hazard; this measure is based on dose exposure data from the ICRP publications[6] and cancer risk data from the BEIR reports.

To put the cancer hazard in perspective, the hazard of the spent fuel is normalized to the cancer hazard of the uranium ore required to produce the fuel. The typical fast reactor assemblies investigated here contain rough] y 20 kg of transuranics. To obtain 20 kg of TRU, 2 MT of LWR spent fuel would need to be processed; and this corresponds to roughly 10 MT of uranium ore.

The uncertainty analysis, has been performed using the typical basic data uncertainties summarized in Table I (no correlation among data has been applied). Uncertainties on radiotoxicity factors have not been included in the analysis, because we decided to limit our study to the impact of basic data like cross sections and half-lives.

Table I. Typical uncertainties on basic data applied to calculate uncertainties on global radiotoxicity

Isotope	σ_{cap}	σ_{fis}	$\sigma_{(n,2n)}$	λ
U-235	10%	5%	50%	3%
U-238	10%	8%	50%	3%
Pu-239	15%	5%	50%	3%
Pu-240	25%	20%	100%	3%
Pu-241	25%	20%	100%	3%
Others	50%	50%	300%	5%

IV. APPLICATION TO EFR

In the case of EFR, one cycle consists of 1500 days of full power irradiation with three different values of constant flux for each 500 day depletion time step.

Three different radioactive waste management strategies have been considered:

Strategy	Disposed to Repository
A	100% MA, 0.3% Pu
B	100% MA, 100% pu
C	1% MA, 0.3% Pu

Strategy A assumes all the Minor Actinides (MA) are removed in reprocessing and disposed to storage; a 0.3 % Pu removal in reprocessing is also assumed. Strategy B utilizes a once-through cycle, where all transuranics (TRU) are disposed to storage. Strategy C considers only fractional TRU removal in reprocessing.

Tables H to IV show selected sensitivity coefficients for the standard subassembly of EFR irradiated during one cycle for the global radiotoxicity calculated at different storing times (10^3, 10^5, and 10^7 years) using the three radioactive waste management strategies. We will note the large sensitivity to Pu-242 and AM-241 capture cross section for strategy A. Also noticeable are the sensitivities to λ of Pu-239 for the 10^5 years storing time (Table 111) and Np-237 λ for 10' years period (Table IV). A significant value is present for the U-238 (n,2n) cross section in the case of strategy A and 10^7 years of storing time. In general sensitivities appear to be quite low.

Table II. Variation (in %) of global radiotoxicity to 100% variation of basic data. Storing time 1000 years, Standard EFR subassembly irradiated during 1 cycle.

Isotope	Strategy	σ_{cap}	σ_{fis}	$\sigma_{(n,2n)}$	λ
U-238	A	1.6	-		
	B	23.7	-0.3	-	
	C	19.4	-0.2	-	
Pu-239	A	3.4	-1.3	-	
	B	18.3	-21.2		- 0.6
	C	15.4	-17.3	-	- 0.5
Pu-240	A	13.5	-1.3	-	- 1.1
	B	8.4	-8.3	·	- 4,5
	C	9.4	-6.9	·	- 3.8
Pu-241	A	0.1	-19.3	·	32.6
	B	-3.9	-22.6		0.3
	C	-3.1	-21.9	·	6.1
Pu-242	A	26,3	-1.9	·	
	B	2.7	0.2	·	
	C	7.3	-0.5	·	
Am-241	A	-46.9	-7.3	·	-102.5
	B	-4.7	-0.8	·	-53.1
	C	-12.9	-2.0	·	-62.7
Am-242M	A	0.1	- 2.0	-	- 8.8
	B		-0,2	-	- 0.9
	C		-0.6	-	2 . 4

Table III. Variation (in %) of global radiotoxicity to 100% variation of basic data. Storing time 100 thousand years. Standard EFR subassembly irradiated during 1 cycle.

Isotope	Strategy	σ_{cap}	σ_{fis}	$\sigma_{(n,2n)}$	λ
Pb-210	A				-13.3
	B				- 4.2
	C				- 5.2
U-234	A				-12.6
	B				-4.0
	C	-	-	-	- 4.9
U-238	A	- 3.2	- 0.1	1.2	-
	B	51.5	- 0.8	0.3	-
	C	46.3	- 0.7	0.4	-
Pu-239	A	0.3	- 3.0	-	-186.3
	B	-13.4	-47.3	-	-239.3
	C	-12.0	-42.5	-	-233.4
Pu-241	A	7.8	-8.5	-	9.6
	B	2.0	-2.7	-	0.9
	c	2.6	-3.3	-	1.9
Pu-242	A	56.3	-4.8		-0.2
	B		-1.3	-	-1.7
	C	6.1	-1.7	-	-1.6
Am-243	A	-27.5	-3.6	-	-31,9
	B	-1.4	-0.2	-	-1.6
	C	-4.2	- 0 6		-4.9

Table IV. Variation (in %) of global radiotoxicity to 100% variation o'f basic data. Storing time '1O million years. Standard EFR subassembly irradiated during 1 cycle.

Isotope	Strategy	σ_{ca}	σ_{fis}	$\sigma_{(n,2n)}$	λ
Ac-227	A				-20.3
	B	-	-	-	-52.4
	C	-	-	-	-47.0
Th-229	A	-	-	-	-29.0
	B	-	-	-	-10.6
	C				
Pa-231	A				-6.4
	B				-16.5
	C				-14.8
^{235}	A	0.9	-0.3		26.4
	B	0.1	-		68.2
	C	0.2	-0.1		61.2
U-238	A	1.4	-0.2	10.0	
	B	45.3	-0.7	0.8	0.5
	C	38.0	-0.6	2.4	0.2
Np-237	A	-4.2	-0.9		-199.5
	B	-0.3	-0.1		-72.6
	C	-1.0	-0.2		-93.8
Pu-239	A	-2.4	-2.1		
	B	-5.8	-41.5		
	c	-4.4	-34.9		
Pu-240	A	12.3	-1.0		
	B	12.1	-2.2		
	c	12.1	-2.0	-	
Pu-241	A	-0.3	-17.3		29.0
	B	-2.8	-16.7	-	0.5
	c	-2.3	-16.8	-	5.3
Pu-242	A	22.8	-1.8		
	B	-1.8	-0.2		
	c	5.3	-0.5		
Am-241	A	-43.9	-6.5		
	B	-3.7	-0.6		
	c	-10.4	-1.6		

In Table V values of the global and time integrated radiotoxicities for the same cases are shown along with the related uncertainties calculated using the values of Table I. We can observe that, as expected, the large radiotoxicities are found for strategy B. Uncertainties are larger in the case of strategy A because of the importance of transplutonium data which carry larger uncertainties. In any case, the uncertainties appear to be quite acceptable with the present status of knowledge on basic data.

Table V. EFR one standard subassembly irradiated during one cycle. Global and time integrated radiotoxicity (expressed in relative cancer hazard) of radioactive waste disposed in repository.

Strategy	Radiotoxicity	Storing Time in Repository		
		10^3 Years	10^5 Years	107 Years
A	Global	3.25 X 10^1 ± 29%	2.50 X 10^{m1} ± 32%	1.77 X 10^{m} ± 30%
	Time Integrated	1.10X 10^3 ± 21%	3.11 X 10^0 ±29 %	4.41 X 10^{-2} ± 23%
B	Global	3.18 x 10^2 ± 8%	1,92 X 10^0 ± 10%	2.07 X 10^{-2} ± 9%
	Time Integrated	5.88 x 10^2 ± 9%	4,27 x 10^1 ± 6%	5.45 x 10^{m} ± 5%
C	Global	1.17X 10^0 ± 10%	1.63 x 10^{m2} ± 10%	7,40 x 10^{-5} ± 11%
	Time Integrated	9.65 x 10^0 ± 12%	2.23 x 10^{m} ± 8%	2.65 x 10^{m3} ± 7%

For Table VI values similar to the ones shown in Table V are provided for an "actinide" subassembly containing 2.5% of Am-a 2.5% of NP to model the composition where EFR is used as a MA burner. The uncertainties are quite similar.

Table VI. EFR one "actinide" subassembly irradiated during one cycle. Global and time integrated radiotoxicity (expressed in relative cancer hazard) of radioactive waste disposed in repository.

Strategy	Radiotoxicity	Storing Time in Repository		
		103 Years	105 Years	107 Years
A	Global	5.87 x 10^1 ± 31%	4.32 x 101 ± 25%	5.08 x 10^{m3} *2 1%
	Time Integrated	2.17 x 10^2 ± 19%	5.56 X 10^0 ± 17%	9.36 X 10^{m2} ± 16%
B	Global	3.29 x 10^2 ± 9%	5.38 X 10^0 ± 9%	2.33 X 10^{m2} ± 10%
	Time Integrated	8.11 x 10^2 ± 10%	4.62 X 10' ± 6%	6.18 x $1o^{m}$ ± 5%
C	Global	1.39 x 10^0 ± 14%	1.91 x 10^{-2} ± 10%	1.06 x 10^{m} ± 1570
	Time Integrated	2.01 x 10^1 ± 13%	3.38 X 10^{m} ± 9%	4.12 x 10^{m3} ± 6%

The self-recycling management has also been taken into consideration. In this case 7 cycles of 1500 days of full power irradiation are followed by 2 years of cooling time (one of them is related to fabrication) in order to reach the equilibrium. At the end of each individual cycle the removal specified in strategy C is disposed to the repository. The results for the global radiotoxicity are shown in Table VII. Of course, the global radiotoxicity has larger values then the corresponding of one cycle irradiation stragegy C. That is due to the larger quantity of waste going to the repository. Uncertainties remain comparable.

Table VII. EFR one standard subassembly irradiated for self-recycling (7 cycles of irradiation, each cycle followed by 2 years of cooling time). Global radiotoxicity, (expressed in relative cancer hazard) of radioactive waste disposed in repository.

Storing Time in Repository		
10^3 Years	10^5 Years	10^7 Years
$6.56 \times 10^{0} \pm 9\%$	$7.65 \times 10^{-2} \pm 10\%$	$3.70 \times 10^{-7} \pm 12\%$

V. APPLICATION TO IFR

In the typical IFR fuel cycle model, a fuel driver subassembly resides in the core for 4 one-year cycles at 80% capacity factor corresponding to a 292 full power days cycle. At the end of the irradiation 0.1 % of the TRU isotopes are lost in the pyroprocessing and disposed to storage. Isotopic depletion was computed using a *900* MWt core and an equilibrium recycle model with 5% of the rare-earths (excluding Y, Eu and Sm) carried in the TRU stream.

In Table VIII selected sensitivity coefficients for the global radiotoxicity are shown for different storing time in the repository. In Table IX values of global and time integrated radiotoxic cities are shown along with their related uncertainties. As expected, radiotoxicity is well below corresponding values of EFR because only 0.1 % of TRU are removed during the reprocessing. Uncertainties are also lower because of less sensitivity for IFR to such isotope basic data as Pu-242 and Am-241. These lower sensitivities are attributed to two effects:

1. Both minor actinides and plutonium have a 0.1 % processing removal; thus, ratio of MA/Pu is much smaller than strategies A and C described in Section IV.

2. The hard neutron energy spectrum associated with the metal IFR fuel leads to less production of higher actinides.

Table VIII. Variation (in %) of global radiotoxicity to 100% variation of basic data. IFR subassembly irradiated during 4 cycles.

Isotope	Storing Time In Repository (Years)	σ_{cap}	σ_{fis}	$\sigma_{(n,2n)}$	λ
AC-227	10^3				
	10^5				-0.2
	10^7				-61.4
Th-229	10^3				
	10^5				-0.2
	10^7				-6,3
Pa-231	10^3				
	105				
	107				-19.3
U-235	10^3				
	10^5				0.2
	10^7				79.9
U-238	10^3	9.6	-0.1	-	-
	10^5	23.3	-0.2	0.1	-
	10^7	20.0	-0.2	0.5	0.1
Nr-237	10^3	-	-	-	-
	10^5	0.3			0.2
	10^7	-0,6	-0.2	-	-43,1
Pu-239	10^3	8.8	-17.3	-	-1.0
	10^5	-7.0	-41,8	-	-273.0
	10^7	-4.6	-36.1	-	
Pu-240	10^3	7.1	-5.9	-	-4,6
	10^5	0.6	-		-0.6
	10^7	6.1	-0.9	-	
Pu-241	10^3	-1,1	-7.5	-	
	10^5	0.3	-0.4	-	0.1
	10^7	-0.6	-4.3	-	0.1
Pu-242	10^3	0.2	-	-	-
	10^5	0.1	-0.1	-	-
	10^7	0.1	-	-	-
Am-241	103	-1.7	-0.5	-	-36.1
	10^5	0.5	-0.1	-	
	10^7	-1.1	-0.3	-	

Table IX. IFR one subassembly irradiated during four cycles. Global and time integrated radiotoxicity (expressed in relative cancer hazard) of radioactive waste disposed in repository.

Radio toxicity	Storing Time in Repository		
	10^3 Years	10^5 Years	10^7 Years
Global	$1.82 \times 10^{-1} \pm 4\%$	$3.68 \times 10^{-3} \pm 10\%$	$1.52 \times 10^{-5} \pm 6\%$
Time Integrated	$2.22 \times 10^{0} \pm 7\%$	$4.44 \times 10^{-2} \pm 4\%$	$5.44 \times 10^{-4} \pm 4\%$

w . CONTRIBUTION FUNCTIONS

The function:

$$Ci(t) = n_i(t) \cdot n_i^*(t)$$

provides the contribution, at each time t, of each isotope i to the functional under study (e.g., the global radiotoxicity). This quantity can be exploited to indicate optimum strategies for radiotoxicity reduction.

In Figs. 1 to 6 we show the main isotope contributes for the global radiotoxicity related to the standard EFR subassembly for strategy A and C at different storing times in the repository (contribution functions for strageties B and C have very similar shapes).

As an example, if we look to Fig. 2, we will observe that Pu-241 and Am-241 functions are almost symetric after going to the repository (this time corresponds to the discontinuity point in the plot at 4.1 years). This is due to the transmutation of Pu-241 to Am-241. Eliminating the Pu-241 at the end of the irradiation will eliminate the big Am-24 1 contribution to the global radiotoxicity after 1000 years of storing in the repository. On the contrary the Pu-239 and Pu-240 contribution remain almost constant during all the time. Similar considerations can be done for other isotopes and different cases.

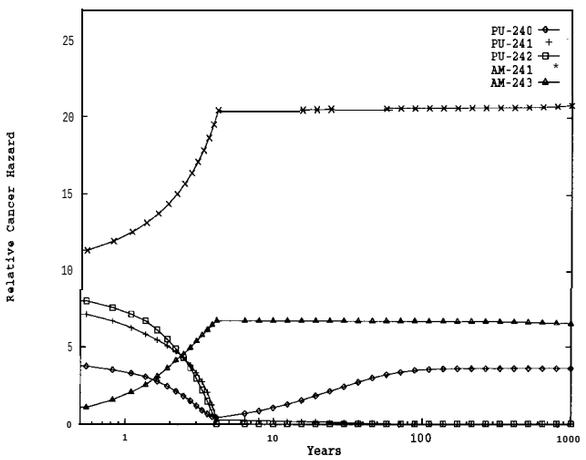

Fig. Strategy A 1000 Years EFR Standard Subassemblies

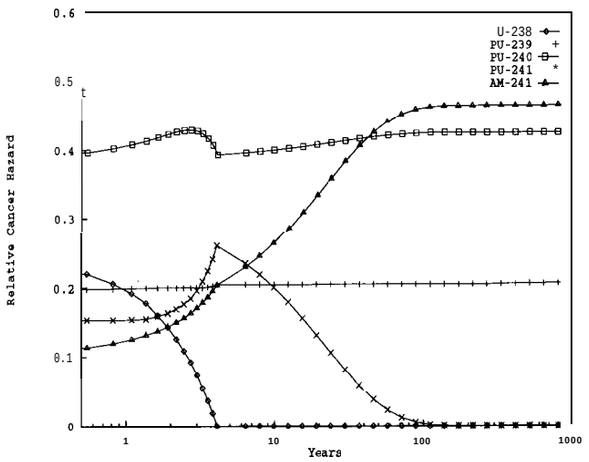

Fig. 2. Strategy C 1000 Years EFR Standard Subassemblies

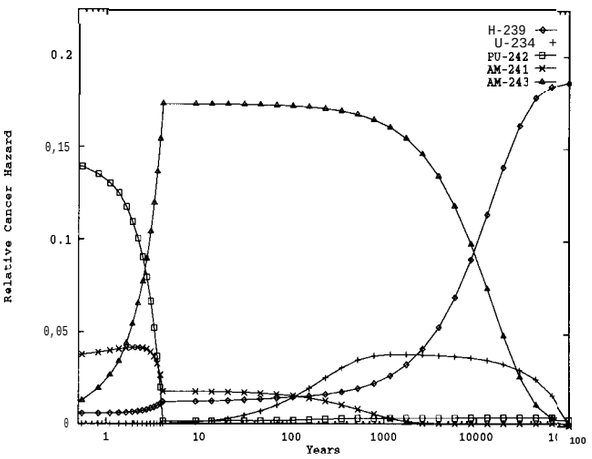

Fig. 3. Strategy A 100000 Years EFR Standard Assemblies

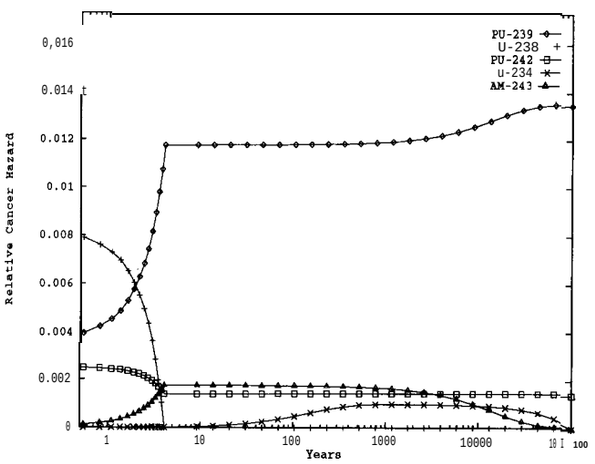

Fig. 4. Strategy C 100000 Years EFR Standard Assemblies

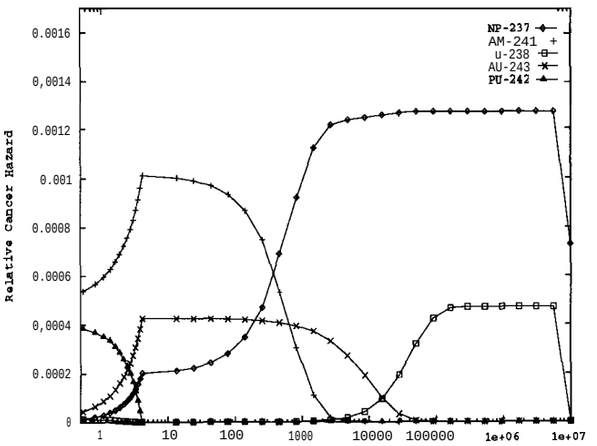

Fig. 5. Strategy A 10000000 Years EFR Standard Assemblies

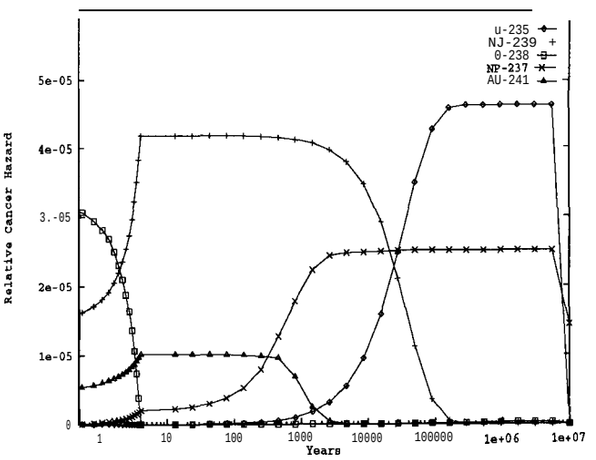

Fig. 6. Strategy C 10000000 Years EFR Standard Assemblies

VII. CONCLUSIONS

A sensitivity and uncertainty analysis on radiotoxicity evaluation has been conducted for such applications as EFR and IFR reactor systems. The main result indicates quite low sensitivities to basic data, except for the Pu-242 and Am-241 capture cross section and some selected half-1 ife constants.

The present knowledge of basic data lead to quite acceptable (of the order of 30% in the worst case) values for uncertainties related to global radiotoxicity at different storing time in repository. The methodology employed can be useful to establish optimum strategies for radiotoxicity reduction. Finally, a similar analysis can be done taking into account the long-lived fission products, but for the scope of this work we have limited our application to actinides.

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