ASSESSMENT OF NUCLEAR POWER SCENARIOS ALLOWING FOR MATRIX BEHAVIOUR IN RADIOLOGICAL IMPACT MODELLING OF DISPOSAL SCENARIOS

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Abstract

The innovative scientific contribution of this study is to consider a third type of radiotoxic inventory: the potential radiotoxic inventory after conditioning, i.e. taking into account the containment capacity of the radionuclide conditioning matrices. The matrix fraction subjected to alteration over time determines the potential for radionuclide release, hence the notion of the potential radiotoxic inventory after conditioning. An initial comparison of possible scenarios is proposed by considering orders of magnitude for the radionuclide containment capacity of the disposal matrices and for their mobilisation potential. All the scenarios investigated are normalised to the same annual electric power production so that a legitimate comparison can be established for the ultimate waste forms produced per year of operation.

This approach reveals significant differences among the scenarios considered that do not appear when only the raw potential radiotoxic inventory is taken into account. The matrix containment performance has a decisive effect on the final impact of a given scenario or type of scenario. Pu recycling scenarios thus reduce the potential radiotoxicity by roughly a factor of 50 compared with an open cycle; the gain rises to a factor of about 300 for scenarios in which Pu and the minor actinides are recycled. Interestingly, the results obtained by the use of a dedicated containment matrix for the minor actinides in a scenario limited to Pu recycling were comparable to those provided by transmutation of the minor actinides.

1. Introduction

Under the provisions of the "separation-conditioning" option of the strategy and program defined under research topic 1 of the 1991 French radioactive waste management law, various fuel cycle scenarios will be assessed and compared [1] in terms of feasibility, flexibility, cost, and ultimate waste radiotoxic inventory. The latter criterion may be further broken down into "potential radiotoxic inventory" (the radiotoxic inventory of all the radionuclides produced) and "residual radiotoxic inventory" (the radionuclide fraction reaching the biosphere after migration from the repository).

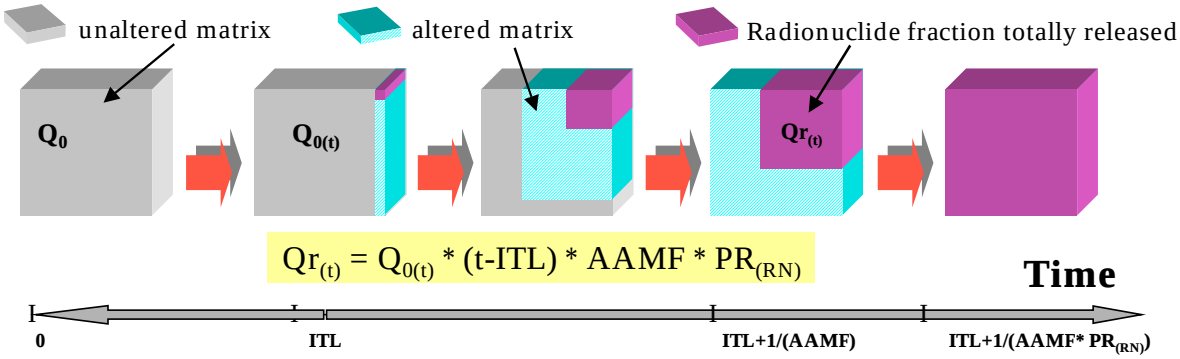
The innovative scientific contribution of this study is to consider a third type of radiotoxic inventory: the potential radiotoxic inventory after conditioning, i.e. taking into account the containment capacity of the radionuclide conditioning matrices. The source term therefore includes only the effects of the radionuclides released from the altered matrix. The matrix fraction subjected to alteration over time determines the potential for radionuclide release, hence the notion of their potential radiotoxic inventory after conditioning. The impact of the radionuclides on the human population is considered through ingestion alone, and not by inhalation.

2. Allowance for matrix containment capacity

The ultimate containment matrices in the various scenarios considered included *uranium oxide* in spent fuel and uranium ore, *glass* in waste packages containing fission products (FP) and minor actinides (MA), *new containment matrices* (NCM) or new glass compositions, and fuel assembly *structural materials* or compacted hulls and end-fittings containing structural activation products (SAP).

The potential release is taken into account by means of three parameters, as shown in Figure 1. The first parameter is the *integrity time limit* (ITL) after which matrix alteration begins; the ITL could correspond to the lifetime of the container (e.g. zircaloy cladding or steel package). The second is the matrix alteration rate, described by the *annual altered matrix fraction* (AAMF), which is constant over time; the reciprocal of the AAMF thus corresponds to the matrix lifetime. The third parameter involves the inherent behaviour of each radionuclide, as expressed by the *probability* that a particular radionuclide will be *released* from the altered portion of the matrix: $PR_{(RN)}$.

Figure 1. **Schematic representation of radionuclide release over time**



The equation postulates that the radiotoxic inventory of the radionuclides released from the matrix $Qr_{(t)}$ is a fraction of the inventory over time in a closed system $Q_{0(t)}$: the fraction is the integral over time since the ITL of the product of the annual altered matrix fractions by the radionuclide

release probabilities. The time distribution of the radionuclide inventory in a matrix is thus a uniform and constant function of ITL up to ITL + 1/(AAMF × $PR_{(RN)}$). The parameter values are indicated in Table 1.

The integrity time limit is first assumed constant for all the packages considered. The AAMF values for spent fuel and for glass reflect the minimum performance corresponding to their maximum leach rates; the figure for NCM represents a target value corresponding to 100 times better containment than currently estimated for glass. Uranium mine tailings were also added as a reference to assess the possible evolution of the equivalent of uranium ore under the same disposal conditions as the other matrices. The quantity considered was the natural uranium requirement necessary for one year of operation of a reactor population at equilibrium; the uranium was assumed to be at equilibrium with its decay products, and was taken into account here in oxide form.

With regard to their radionuclide release probability factors, the actinides characterised by their low mobility were assigned a value of 10^{-2} over duration equal to 1/AAMF. The release of the highly mobile fission products (iodine, caesium and technetium) was considered congruent with the matrix alteration, hence their $PR_{(RN)}$ value of 1. The other less characteristic chemical elements were assigned intermediate AAMF values.

Table 1. **Parameter values for the radionuclide release equation: (boldface figures are the product of AAMF × $PR_{(RN)}$)**

Matrix	**ITL (years)**	**AAMF ($year^{-1}$)**	**$PR_{(RN)}$**							
			Pu (10^{-2})	Am (10^{-2})	U (10^{-2})	I (1)	Tc (1)	Cs (1)	Other FP (10^{-1})	SAP (10^{-1})
Uranium oxide	300	10^{-4}	10^{-6}	**10^{-6}**	**10^{-6}**	**10^{-4}**	**10^{-4}**	**10^{-4}**	**10^{-5}**	
Glass	300	10^{-5}	**10^{-7}**	**10^{-7}**	**10^{-7}**	**10^{-5}**	**10^{-5}**	**10^{-5}**	**10^{-6}**	
New containment matrices	300	10^{-7}	**10^{-9}**	**10^{-9}**	**10^{-9}**	**10^{-7}**	**10^{-7}**	**10^{-7}**	**10^{-8}**	
Structural materials	300	10^{-4}	**10^{-6}**	**10^{-6}**	**10^{-6}**	**10^{-4}**	**10^{-4}**	**10^{-4}**	**10^{-5}**	**10^{-5}**

3. Application to fuel cycle scenarios

3.1 Scenarios

All the scenarios considered were normalised with respect to an electric power production of 400 TWh/year. The scenarios also assumed quasi steady-state operation to allow valid comparisons of the ultimate waste production over one year of operation in each case. The twelve scenarios taken into account are briefly described below, and can be considered as belonging to four major types:

- *Scenarios resulting in large quantities of plutonium and minor actinides in the waste materials.* Open-cycle scenario, and once-through-Pu scenario in which plutonium is recycled once as MOX fuel without further reprocessing.
- *Scenarios eliminating the plutonium from the waste materials.* FNR-Pu, PWR/FNR-Pu, and PWR-Pu scenarios, in which plutonium is recycled repeatedly either as MOX fuel in pressurised water reactors (PWR) or fast neutron reactors (FNR), or as

PWR MIX (MOX with enriched uranium) fuel; a variant with isotopic separation of ^{242}Pu was also considered: IS-Pu242.

- *Scenarios eliminating the plutonium from the waste materials, with separation and specific conditioning of the minor actinides.*
  These are variants of the preceding scenarios with implementation of enhanced separation and conditioning (SC) techniques. The fuel is reprocessed in an enhanced reprocessing plant using the new DIAMEX and SANEX processes to separate americium (Am) and/or curium (Cm) for incorporation in a new containment matrix (NCM) with very high radionuclide retention performance (Table 1). The vitrified waste therefore contains only fission products (except for process losses). These scenarios are designated: PWR-Pu/MA-SC, FNR-Pu/MA-SC, and PWR/FNR-Pu/MA-SC.

- *Scenarios eliminating the plutonium and some or all of the minor actinides from the waste materials for transmutation in PWRs or FNRs.*
  These are the PWR-Pu/MA, PWR-Pu/NpAm (Cm is sent to vitrification), FNR-Pu/MA, and PWR/FNR-Pu/MA scenarios in which the minor actinides are transmuted in homogeneous mode, and the PWR/FNR-AmCm-target and PWR/FNR-Am-target scenarios in which the actinides are transmuted in heterogeneous mode as once-through targets; after irradiation, 90% of the minor actinides are transmuted into fission products.

3.2 Fuels

The burn-up is assumed equal to 60 GWd·t^{-1} for all the PWR fuels, and approximately 140 GWd·t^{-1} for the fast neutron reactor fuels. The U and Pu reprocessing losses are assumed equal to 0.1%. All the fuel compositions and their annual flows are determined by neutronic feasibility analysis, calculated by the Reactor and Fuel Cycle Physics Department of the CEA's Nuclear Reactor Division (DRN/SPRC) [2]. The comparisons were performed under steady-state conditions based on the total annual production.

The potential radiotoxic inventory of the ultimate wasteforms over time were calculated by multiplying the activities of each radionuclide (determined by decay using the JEF2.2 data [3]) by the dose-per-unit-intake factors (Sv·Bq^{-1}) from ICRP72 [4].

4. Results

4.1 Mass balance and "raw" potential radiotoxic inventory

The mass balance was established for the fission products, minor actinides and plutonium released from the waste (Table 2). The structural activation products have a lower radioactive and radiotoxic impact. The reprocessed uranium is not considered as an ultimate waste form, unlike the uranium reprocessing losses (estimated at 0.1%).

Table 2. **Annual heavy nuclide contribution (kg/year) to ultimate waste form for each scenario**

Recycling policy	**No recycling**	**Partial Pu**	**Pu recycling**			
Scenario	Open cycle	Once-through Pu	Isotopic separation ^{242}Pu	MIX Pu	FNR Pu	PWR/FNR Pu
U	754 736	79 105	743	747	238	440
Pu	10 332	3 175	2 021	17	56	35
Np	746	683	632	674	172	499
Am	645	1 229	1 852	1 853	1 425	1 406
Cm	113	286	360	951	112	196
Total (excl. U)	11 835	8 373	4 865	3 495	1 776	2 136

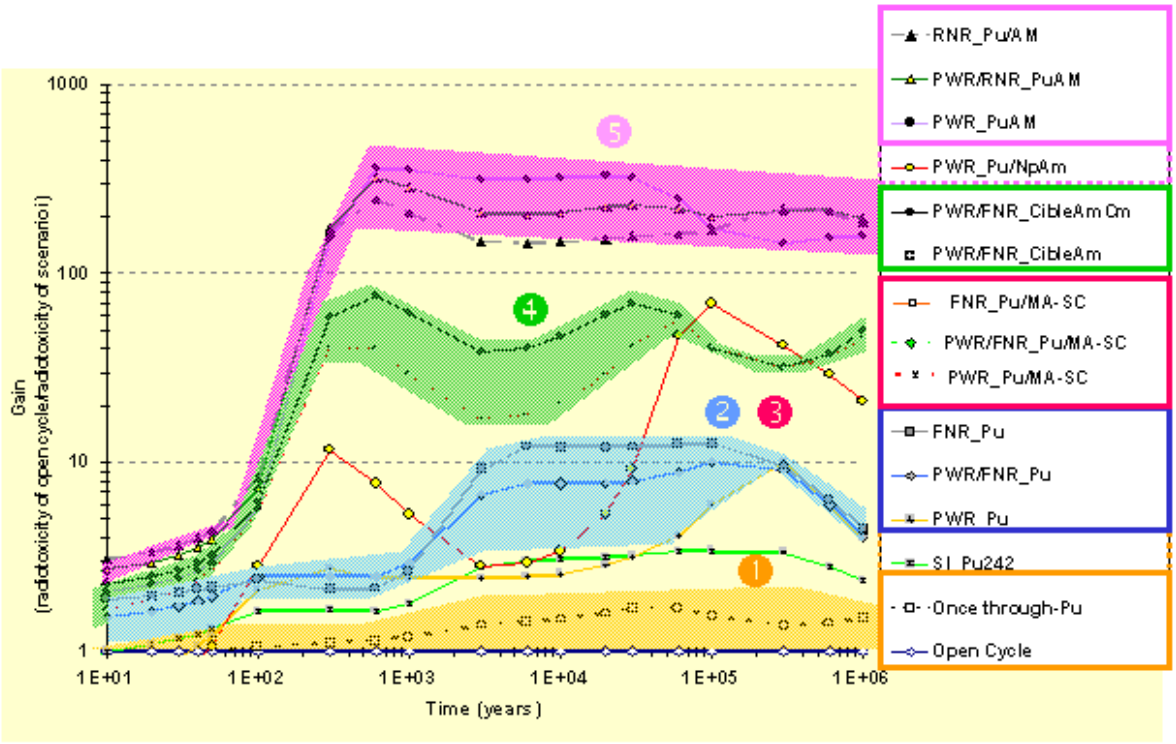
Recycling policy	**Pu + MA recycling**					
Scenario	MIX Pu + MA	MIX Pu + AmNp	PWR/FNR Am/Cm targets	PWR/FNR Am targets	PWR/FNR Pu + MA	FNR Pu + MA
U	735	720	437	438	378	236
Pu	25	22	96	95	39	58
Np	1	1	1	1	1	1
Am	3	3	16	15	2	2
Cm	3	1 668	94	252	1	1
Total (excl. U)	32	1 694	207	363	43	62

When the raw radiotoxic inventory results are plotted relative to the "open-cycle" reference scenario (Figure 2), three main categories of scenarios can be distinguished.

- The first includes the scenarios with significant quantities of residual plutonium (open and once-through cycles).
- The second, with a potential radiotoxic inventory 4 to 10 times lower, comprises the multiple plutonium recycling scenarios (MIX-Pu, FNR-Pu, and PWR/FNR-Pu).
- The third includes the scenarios in which both plutonium and the minor actinides are recycled (MIX-Pu/MA, FNR-Pu/MA, and PWR/FNR-Pu/MA), resulting in a potential radiotoxic inventory some 100 times lower than in the reference scenario.

Note: From the standpoint of the potential radiotoxic inventory, there are no differences between the basic scenarios and the variants involving a separation and conditioning strategy.

Figure 2. **Reduction of potential raw radiotoxic inventory for each scenario compared with open cycle**



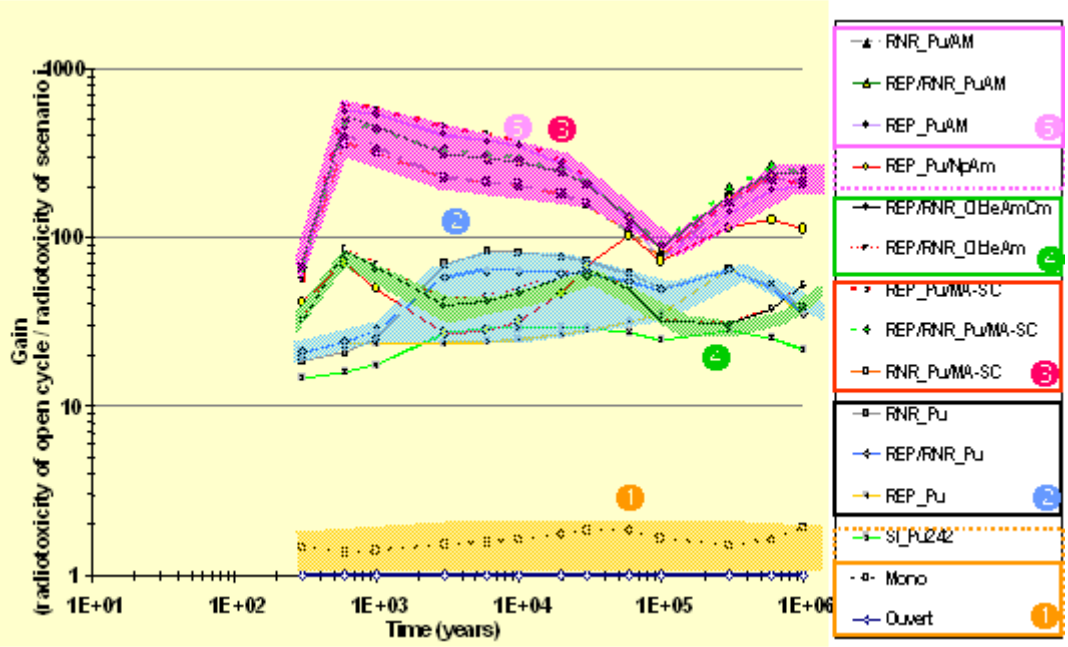
4.2 Potential radiotoxic inventory after conditioning

Allowing for the containment capacity of the conditioning matrix, the logical outcome of the processes implemented in spent fuel reprocessing, provides a new basis for comparing the possible scenarios.

The matrix containment performance has a decisive influence on the final impact of a given scenario or group of scenarios. The Pu recycling scenarios provide a gain by a factor of about 50 over the open cycle in terms of the potential radiotoxic inventory after conditioning; the scenarios in which both plutonium and minor actinides are recycled result in a gain by a factor of about 300.

It is interesting to note that specific conditioning of the minor actinides in high-performance containment matrices in a scenario in which Pu alone is recycled would be as effective as transmuting the minor actinides. Moreover, the effectiveness of the scenarios in which the minor actinides are recycled as once-through targets would be no better, under the hypothetical conditions of this study, than recycling plutonium alone, as the glass matrix provides better containment than the unreprocessed targets.

Figure 3. **Reduction of potential radiotoxic inventory after conditioning for of each scenario compared with open cycle**



5. Conclusion

The notion of the radiotoxic inventory after conditioning, by taking into account the respective containment properties of each ultimate wasteform, provides a means for distinguishing three categories of fuel cycle management routes according to the potential release of the radiotoxic inventory.

Fuel cycle management strategies in which plutonium is recycled partially or not at all yield the poorest performance; multiple plutonium recycling strategies are about 50 times more effective in this respect, and multiple recycling of plutonium and the minor actinides is even more effective (some 300 times more than the open cycle).

Enhanced reprocessing together with the use of dedicated matrices having a containment capacity 100 times better than glass taken into account in this study would result in performance factors equivalent to those of an enhanced reprocessing/transmutation cycle without requiring the use of burner reactors.

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