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Uncertainty Analysis on Back-end Fuel-Cycle Main Parameters

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The present technology of nuclear energy production generates a long-term risk source. Several parameters are considered as indicators of this risk; among them, the external-radiation toxicity, the ingestion toxicity and the residual power. The paper analyzes the main sources of uncertainties affecting these parameters and investigates how uncertainties propagate through the computational procedure owing to methodological simplifications and assumptions. Eventually, an estimation of the overall uncertainty on a cooling-time period of one-million years is presented and discussed.

Keywords: Back-end Fuel Cycle, Nuclear Wastes, Activity, Radiotoxicity, Residual Power, Uncertainty, Decay, Actinides, Fission Products

1. Introduction

The present technology of nuclear energy production generates a long-term risk source, which is roughly proportional to the mass of fuel unloaded from power reactors and depends on spent fuel isotopic composition. Potential dangers include connected nuclear waste, among others, the residual power, the ingestion toxicity and the external-radiation toxicity. In the present work, we focus on external-radiation toxicity and residual power only, ingestion toxicity showing time behaviour and sensitivity values which are very close to residual power.

Fission-Products (FP) are the main source of spent fuel activity in the short term through gamma-ray emission that follows β decay. Their contribution to middle-term activity remains significant through ^{137}Cs and ^{90}Sr decay, but their contribution to long-term activity is almost negligible and, anyway, far lower the actinide one, ^{135}Cs, ^{129}I and ^{99}Tc being relevant contributors only.

Plutonium is the most abundant of the man-made transuranic elements present in nuclear spent fuel. Considered as a potential energy resource by several countries in regard to strategic and economic considerations, a variety of options and strategies have been proposed to extend its utilization within

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closed fuel cycles. Other countries adopt open fuel cycles where plutonium, a by-product of nuclear energy production, is simply considered as a component of nuclear waste.

Transuranics (including plutonium) widely dominate the middle and long-term external-radiation toxicity and residual power of spent fuel. Accordingly, the activity of nuclear waste is significantly modified by radioactivity decay after fuel discharge. It is generally assumed that it reaches the natural level of uranium ores again after a one-million-year decay period (see Fig 1).

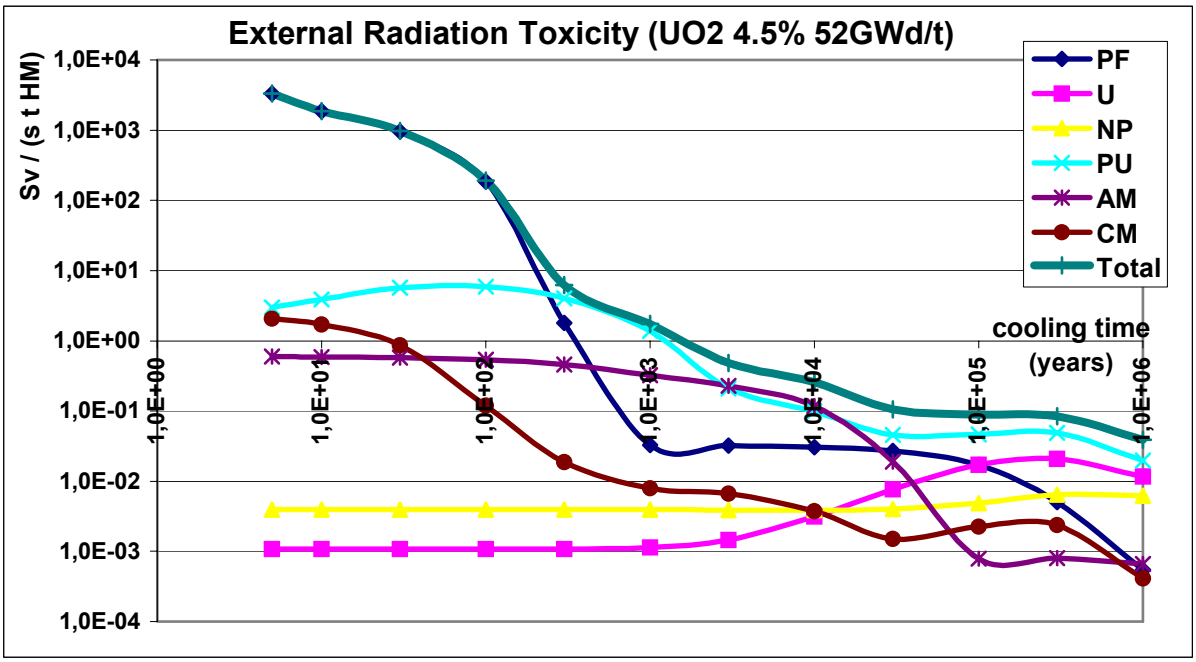

Fig. 1 - Contributions per isotope to external-radiation toxicity vs. cooling-time calculated for an ICRU sphere placed at one meter from the spent fuel (Sv per second per ton of Heavy Metal Spent Fuel (HMSF)).

The risk can be significantly reduced by adopting suitable strategies of waste handling and storage; however a large uncertainty subsists on the evaluation of the efficiency of such strategies. Many back-end fuel cycle parameters must be taken into account to evaluate the best strategy minimizing the potential dangers connected to nuclear waste.

Calculation of back-end fuel cycle is a long and difficult job which requires use of suitable data, qualified codes and a high computational capacity. A linear approach to this problem can obviously simplify and speed up calculations.

Accordingly, a simplified methodology has been developed and it is currently adopted at FRAMATOME-ANP. It allows computing the PWR fuel-mass vector of both UOX and MOX fuels within French standards, at the reactor discharge, at one million-year cooling-time and at any other intermediate cooling-time. It is based on a linearization of the fuel-life equations (the so called Bateman equations) through definition of matrix operators. An uncertainty analysis to estimate the precision of results is mandatory for any practical application.

2. Decay calculations

The basic Bateman fuel life-equations are described by the following vector differential equation [1], which holds both for reactor and spent fuel.

$$\frac{d\underline{n}}{dt} = \left[-\underline{\underline{\Lambda}} + \underline{\underline{\Gamma}}(\phi, \underline{n})\right] \cdot \underline{n} \tag{1}$$

where, at any time *t*:

- $\underline{n}$ (a vector) accounts for the nuclear density of the isotopes,
- $[\Lambda]$[(a lower-triangular matrix operator) accounts for the natural decay,
- $[\Gamma(\phi, n)]$ (a full matrix operator) accounts for the neutron entertained transmutation of nuclides (burn-up, build-up and breeding process).

The $[\Gamma(\phi, n)]$ operator also accounts for the power history of the system through, the normalization of the flux.

It is evident that, when neutron flux vanishes, the term on right of the vector equation drops to zero. This is the situation of the spent fuel discharged from reactors during the cooling period, if a zero-flux assumption within the mould is made.

2.1. The linear approach

In a linear approach, it assumed that all the back-end fuel cycle parameters *q* are linear functions of the fuel-mass vector. Accordingly, they can easily be assessed when the reactor is unloaded and at any cooling time step (*t*) via a matrix product between the fuel-mass vector m*(t)* and a suitable weighting-factor vector $\underline{h}$, as follows:

$$q(t) = \underline{m}(t)\, \underline{h} \tag{2}$$

Fuel compositions $\underline{m}(t)$ at any time step in the cooling period are obtained applying the decay matrix operator $\underline{\underline{D}}$ to the discharged fuel vector $\underline{m}(0)$, to project it to any desired down-stream state (the time-step "0" conventionally represents the core discharge time):

$$m(t) = \underline{\underline{D}}\,(t)\, \underline{m}(0) \tag{3}$$

Analytical solutions of the Bateman equations (4) can be used to compute the decay matrix $\underline{\underline{D}}\,(t)$ operator, which only depends on the time variable:

$$\begin{cases} n_i(t) = \sum_{j=1}^{i} C_{ij} e^{a_{jj} t} \\ C_{ij} = \sum_{k=j}^{i-1} \dfrac{a_{i,k}}{a_{jj} - a_{ii}} C_{k,j}; \qquad i \neq j \\ C_{ii} = n_i(0) - \sum_{k=1}^{i-1} C_{ik} \end{cases} \tag{4}$$

where a_{ij} accounts for the element (i, j) of the lower-diagonal matrix $[\underline{\underline{\Lambda}}]$.

Fuel compositions at the core discharge $\underline{m(0)}$ are, in turn, computed applying the fuel-life matrix operator $\underline{\underline{T}}\ (R, BU)$ to the fresh fuel vector $\underline{m_{fresh\ fuel}}$ to project it to the desired end of cycle state:

$$m(0) = \underline{\underline{T}}\ (R, BU)\ .\ \underline{m_{fresh\ fuel}} \tag{5}$$

The three steps described here above can be collapsed into a compact expression, as follows:

$$q(t) = [\ \underline{\underline{D}}\ (t)\ \underline{\underline{T}}\ (R, BU)\ .\ \underline{m_{fresh\ fuel}}\ ]\ \underline{h} \tag{6}$$

Assuming the fresh fuel compositions are known with zero uncertainty, every step in the linear approach above here (Equations (2), (3) and (5)) generates a contribution to the overall uncertainty that affects any parameter $q(t)$.

Even though, in this paper, decay and depletion steps only (Equations (3) and (5)) are fully discussed, it is essential to account for uncertainty generated by assumptions made in Equation (2). Therefore, the final results presented here account for all uncertainties affecting the back-end cycle parameters.

The following methodology has been adopted to evaluate the impact of uncertainties on final results:

- uncertainties connected to the linearization of decay calculations and those related to the linearization of depletion calculations are analysed separately,
- to find out the global uncertainty affecting final results individual uncertainties are cumulated both linearly and in a quadratic way,
- uncertainty calculations are repeated for several back-end cooling time-steps of interest.

2.2. Algorithm used for numerical computation

Problem of spent fuel decay in zero-flux conditions can be solved analytically. Matrix $\underline{\underline{D}}(t)$ is computed starting from matrix $\underline{\underline{\Lambda}}$, which contains nuclear features of isotopes of interest. This means that the matrix can be made with all fuel unloaded from any same-technology reactor system. Matrix $\underline{\underline{D}}(t)$ depends on adopted time-steps and on $\underline{\underline{\Lambda}}$ values only. Thus it is unique for a given time-step t.

Analytical solutions allow computing the isotopic composition vector $\underline{n(t)}$ at any time t starting from the beginning of life vector $\underline{n(0)}$, the generic component of this vector being given by the following equation:

$$\underline{n}(t) = \sum_{j=1}^{i} C_{ij} e^{a_{ij} t} \equiv \underline{f}(\underline{n}(0), t) = \underline{\underline{D}}(t) \cdot \underline{n}(0) \tag{7}$$

Analytical solutions evaluated that way depend on both time-steps and initial conditions. If it is more convenient to obtain solutions depending on time-step only, they can be obtained for any isotope, looping on isotopes as follows:

$$\underline{n}_i(0) = \begin{bmatrix} 0 & \cdots & 0 & 1 & 0 & \cdots & 0 \end{bmatrix}^T \quad \Leftrightarrow \quad \begin{cases} n_{ij}(0) = 1 & i = j \\ n_{ij}(0) = 0 & \forall i \neq j \end{cases} \tag{8}$$

$$\left[\underline{n}_1(0)\middle|\underline{n}_2(0)\middle|\cdots\cdots\middle|\underline{n}_N(0)\right]=\begin{bmatrix}1 & 0 & \cdots & \cdots & 0\\ 0 & 1 & & & \vdots\\ \vdots & & \ddots & & \vdots\\ \vdots & & & \ddots & 0\\ 0 & \cdots & \cdots & 0 & 1\end{bmatrix}=\underline{\underline{I}} \tag{9}$$

The complete matrix is to be constructed combining individual contributions.

$$\begin{aligned}&\left[\underline{n}_1(t)\middle|\underline{n}_2(t)\middle|\cdots\cdots\middle|\underline{n}_N(t)\right]=\left[\underline{f}\left(\underline{n}_1(0),t\right)\middle|\underline{f}\left(\underline{n}_2(0),t\right)\middle|\cdots\cdots\middle|\underline{f}\left(\underline{n}_N(0),t\right)\right]=\\&=\left[\underline{\underline{D}}(t)\underline{n}_1(0)\middle|\underline{\underline{D}}(t)\underline{n}_2(0)\middle|\cdots\cdots\middle|\underline{\underline{D}}(t)\underline{n}_N(0)\right]=\underline{\underline{D}}(t)\cdot\underbrace{\left[\underline{n}_1(0)\middle|\underline{n}_2(0)\middle|\cdots\cdots\middle|\underline{n}_N(0)\right]}_{\underline{\underline{I}}}=\underline{\underline{D}}(t)\end{aligned} \tag{10}$$

Although this approach is fully analytical, two main sources of uncertainty exist:

- the impact of spontaneous sources of neutrons on the spent fuel behavior,
- the propagation of errors through the nuclide chains.

2.2.1. Neutron source in spent fuel

A weak fission-independent neutron source subsists in spent fuel, due to spontaneous fissions, alpha-n and (n, 2n) reactions. It strongly depends on the spent fuel features and composition. Amplified by the system according to its sub-criticality level, such a source sustains a fission neutron flux which can be quite high and generate transmutations in the fuel. Events like these can slightly modify spent fuel isotopic composition in time compared to pure decay. This spontaneous source has systematically been neglected in decay calculations, as the connected nuclide-transmutation operator has never been taken into account.

To check the pertinence of such an a priori assumption, a comparison was made between a conventional computation and an equivalent one, where spontaneous neutron source was accounted for.

Analysis of final storage strategies is currently underway in different countries, thus actual technological solutions are still unknown. Accordingly, two different extreme situations, representing the two most commonly retained waste-disposal strategies, have been analyzed:

- French solution: the fuel unloaded from power reactors is reprocessed to separate uranium and plutonium isotopes from Minor Actinides and Fission Products. Uranium and Plutonium can be re-cycled, MA and FP are stored separately. Therefore MA and FP only have been accounted for when evaluating neutron-induced reactions in the glass vitrified waste;
- US solution: all the unloaded fuel is considered as waste. It must be stored without reprocessing after cooling in the cooling pool. In the present study, all the discharged fuel (including plutonium, uranium, MA and FP) has been calculated as waste.

After defining the container geometry, the composition of the glass mould and the nuclear waste concentration within, all necessary information is available to solve neutron transport and Bateman equations, also to compute isotopic composition of radioactive wastes for any cooling-time period.

It is clear from Bateman equations (Eq. 11) that errors should be higher for isotopes sharing strong competition among nuclear induced absorption rates and radioactive decay:

$$\frac{dn^i}{dt} = \underbrace{\sum_{j \neq i} \sigma^{j \to i} \phi n^j}_{\substack{\text{creations due} \\ \text{to other isotopes} \\ \text{reactions}}} + \underbrace{\sum_{j \neq i} \lambda^j \gamma^{j \to i} n^j}_{\substack{\text{creations due to} \\ \text{other isotopes} \\ \text{decay}}} - \underbrace{\lambda^i n^i}_{\substack{\text{disintegration} \\ \text{due to natural} \\ \text{decay}}} - \underbrace{\sigma^i_{tot} \phi n^i}_{\substack{\text{disintegration} \\ \text{induced by neutron} \\ \text{flux}}} \qquad (11)$$

Cross-sections and fluxes in Eq. 11 above here are all one-group. Cross-sections are generally collapsed from multi-group values with suitable procedures where neutron fluxes are used as weighting functions. These fluxes are computed by solving the neutron transport equation on appropriate geometrical samples with either probabilistic or deterministic methodologies. At FRAMATOME-ANP, both a deterministic transport code and a probabilistic Monte-Carlo code are used.

$$\phi_0 = \int_{E_{\min}}^{E_{\max}} \phi(E) dE \qquad \sigma_0 = \frac{\int_{E_{\min}}^{E_{\max}} \sigma(E) \cdot \phi(E) dE}{\phi_0} \qquad (12)$$

In the present application, the transport equation has been solved with MCNP Monte-Carlo code [2] on a cylindrical sample model made of a homogeneous glass mould with a volume of about 250 litres, with 7 kg of radioactive waste distributed uniformly inside. MCNP calculations could then be made for criticality level and flux of the sample.

Comparison between neutron-induced rates and decay rates on UOX reprocessed fuel after ten-year decay period demonstrated that the natural decay component remains higher than the neutron-sustained one. Such conclusions can be generalized to any practical case according to the very conservative assumptions on decay time, neutron source and isotopic composition of waste (reprocessed vs. un-reprocessed ones).

2.2.2. Propagation of errors affecting nuclide base-data through the nuclide chains.

In decay calculations, the only nuclide data that really matter are those concerning half-lives (or decay constants) of nuclides.

In the field of fuel back-end studies, errors affecting decay constants have no impact on the *q(t)* parameters if the half-life is either very short (less than one second) or very long (more than one billion years).

Generally, half-lives between one second and one billion years are fairly accurate. Therefore, the purpose of the present analysis is to discover whether those low errors are amplified through nuclide chains.

The numerical approach adopted is as follows:

1. main back-end parameters are computed for a given spent-fuel composition and for all relevant time steps with usual decay constants (reference values),
2. assuming that decay constants are known with a relative error not greater than *p*, a relative random perturbation uniformly distributed in the interval *[-p, p]* is applied to any decay constant,
3. back-end parameters are computed at relevant time steps with perturbed decay constants,
4. differences between *q(t)* reference and so-perturbed values are observed and analyzed,

5. looping on perturbations allows increasing statistical distribution of the derived discrepancies at each time step,

6. the largest absolute values of discrepancies are adopted as an estimate of the overall uncertainty.

As a general conclusion, it can be stated that any statistical perturbation of decay constants of relative amplitude p and uniform distribution generates a perturbation lower than p on any $q(t)$ parameter at any cooling-time between 5 and one million years.

Therefore, there is no amplification of the uncertainties through chains of errors affecting decay constants. As these errors are very low (<2%) for all decay constants corresponding to half-lives of interest, we assume uncertainties on final results due to errors affecting nuclide base-data can be ignored.

3. Depletion calculations

As said (Eq. 5), fuel compositions at the core discharge are obtained applying the fuel-life matrix operator to the fresh fuel vector to project it on the desired downstream state:

$$\underline{m}(0) = \underline{\underline{T}}(R, BU) \cdot \underline{m}_{fresh\ fuel}$$

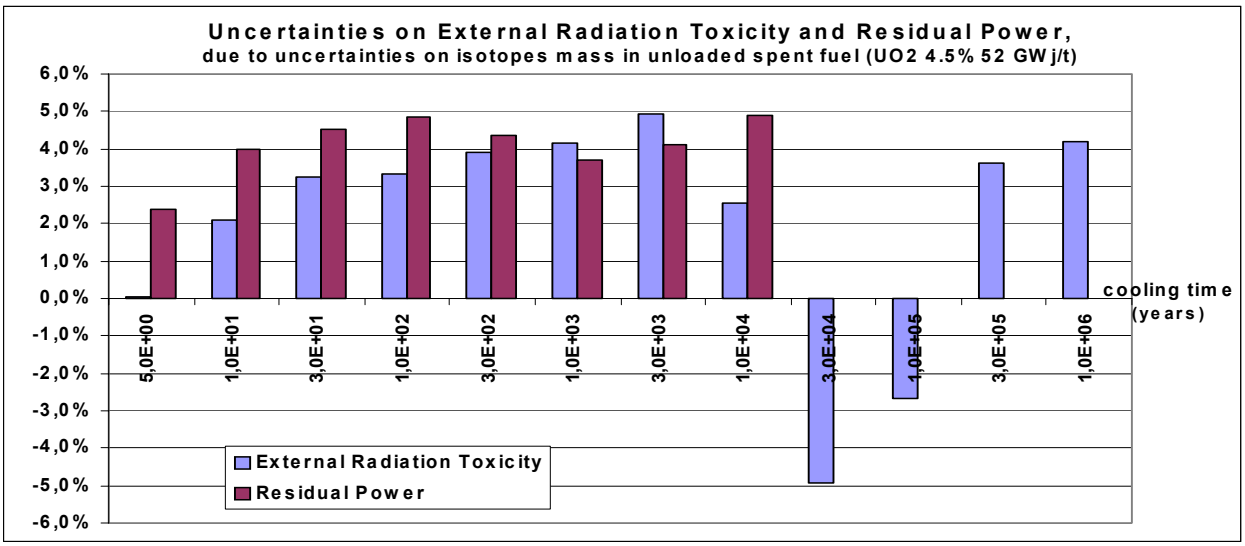

Fig. 2 - Uncertainties on external-radiation toxicity and residual power, due to uncertainties on isotope mass in unloaded spent fuel (UOX 4.5% - 52 GWd/t)

The transmutation matrix $\underline{\underline{T}}(R, BU)$ is computed using the one-dimension SCALE-ORIGEN chain [3] for a given enrichment (R) and burn up (BU) in an infinite medium homogeneous geometry approximation.

A partial validation of fuel compositions computed with the deterministic geometry-simplified SCALE-ORIGEN chain against the probabilistic geometry-detailed MCNP-MONTEBURNS-ORIGEN (MMO) [4] chain was performed.

Through this validation, an estimation of the sensitivity of $q(t)$ parameters to uncertainties in mass values calculated by the SCALE-ORIGEN chain (Fig. 2) is obtained.

Several main sources of uncertainty affect the linear approach adopted in depletion calculations:

a) The linearity vs. burn-up assumed in building-up neutron-sustained-process matrix operators. This assumption impacts the precision of the whole depletion process (the in-core burn-up, build-up and breeding neutron-sustained events) through the coupling among computational time-steps, nuclide densities and core flux-level.

Every matrix $\underline{\underline{T}}(R, BU)$ being evaluated at a given burn-up, it is obvious that the number of available matrices is limited to burn-up steps. If one wants to compute $q(t)$ at a BU which does not correspond to a computational step, a linear interpolation between the edging matrices is made. In the present study, it was demonstrated that this approximation remains acceptable within time-steps lower than 8 GWd/t.

b) The limited set of enrichments adopted to construct matrices and the SCALE-ORIGEN computational basic assumptions. Every matrix has been evaluated for several different enrichments of the fuel (3 Uranium , 8 Plutonium enrichments). Any enrichment range is connected to a given uncertainty level. Using such matrices for fuel enrichments outside the definition range generates an uncertainty on the computed results. Low overall uncertainties show that the methodology adopted is quite fine and robust (Fig. 3).

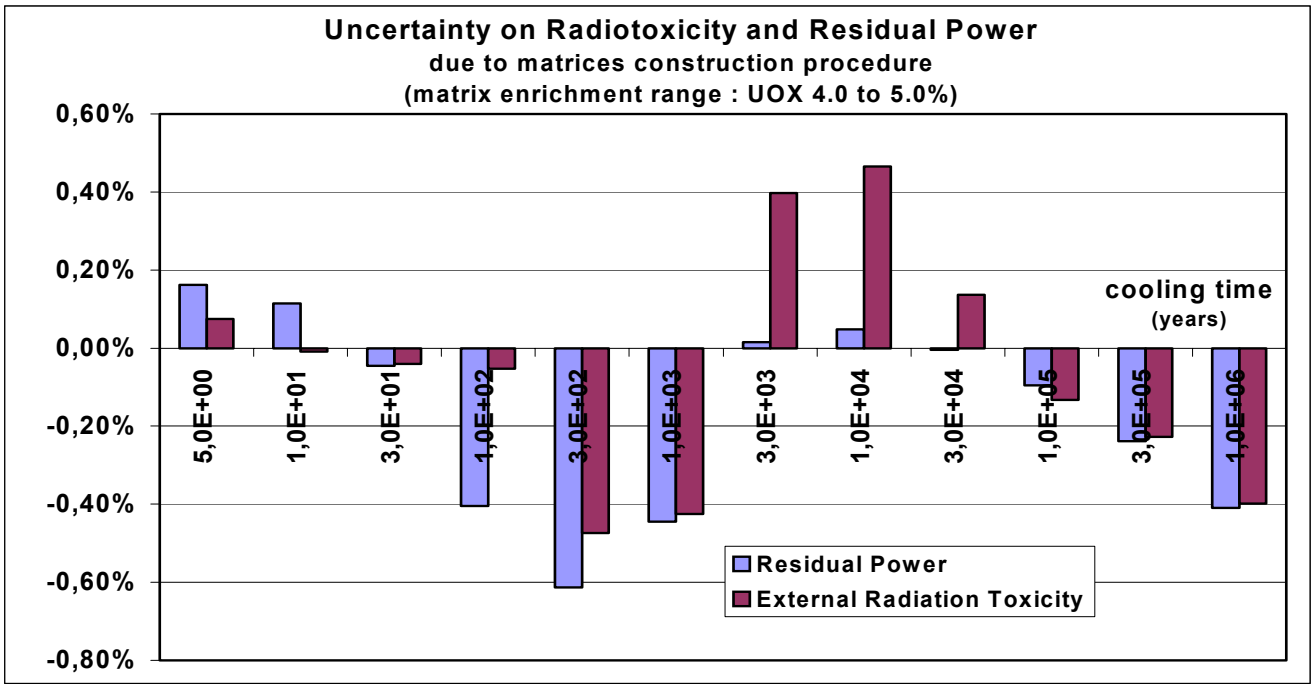

Fig. 3 - Uncertainties on external-radiation toxicity and residual power vs. cooling-time period, due to matrices construction procedure. Matrix enrichment range: UOX 4.0 to 5.0 %.

c) The power history. Nuclear reactors can release energy according to very different strategies, which depend on exploitation policies, demand and grid features. An evaluation of the sensitivity of parameter uncertainty to the power history was made heuristically by running a series of nine independent ORIGEN-S depletion calculations, where the same target of fuel burn-up at reactor discharge was obtained according to different exploitation strategies, including re-fuelling, long-

period exploitation at low power and stretching-out. Crossed result comparison allowed estimating uncertainty connected to this important exploitation parameter.

d) The heterogeneity of burn-up among assemblies at the discharge. Burn-up heterogeneity is a complex function of operation and control options, re-loading strategy, fuel burn-up target at reactor discharge and actual burn-up. Burn-up matrices are bounded to an applicability range vs. enrichment within a given burn-up interval. According to the quite large dispersion of fuel burn-up values at reactor discharge, *q(t)* values are sensitive to the way this dispersion is accounted for in depletion calculation. A sensitivity analysis was made to evaluate uncertainty resulting from by burn-up dispersion by comparing multiple burn-up realistic cases to conventional averaged burn-up solutions.

Table 1 summarizes main results of the uncertainty analysis on external-radiation toxicity and residual power vs. cooling-time. Arithmetic (linear) and squared combinations of individual contributions to uncertainty are presented here. It must be emphasized that uncertainties on toxicity and residual power remain remarkably low throughout the whole cooling-time period considered of one-million years.

Table. 1 – Maximum values of uncertainties of external-radiation toxicity and residual power in the whole cooling-time period considered in the study.

PWR UOX 4.5% 52GWd/t	**Ext. Rad. Toxicity (1)**	**Residual Power (2)**
Arithmetic combination	20.4%	11.1%
Squared combination	13.1%	8.4%
PWR MOX 8.65% 48 GWd/t	**Ext. Rad. Toxicity (1)**	**Residual Power (2)**
Arithmetic combination	12.1%	4.7%
Squared combination	10.3%	3.5%

(1) Sv per second and per ton of Heavy Metal of Spent Fuel (Sv / s.t(HMSF))

(2) W per ton of Heavy Metal of Spent Fuel (W / t(HMSF))

Fig. 4 presents a detailed history of the uncertainty on external-radiation toxicity vs. cooling time. The short- and intermediate-term peaks which can be observed in the draw are mainly due to the Cs chain and the ^{241}Pu – ^{241}Am chain, respectively.

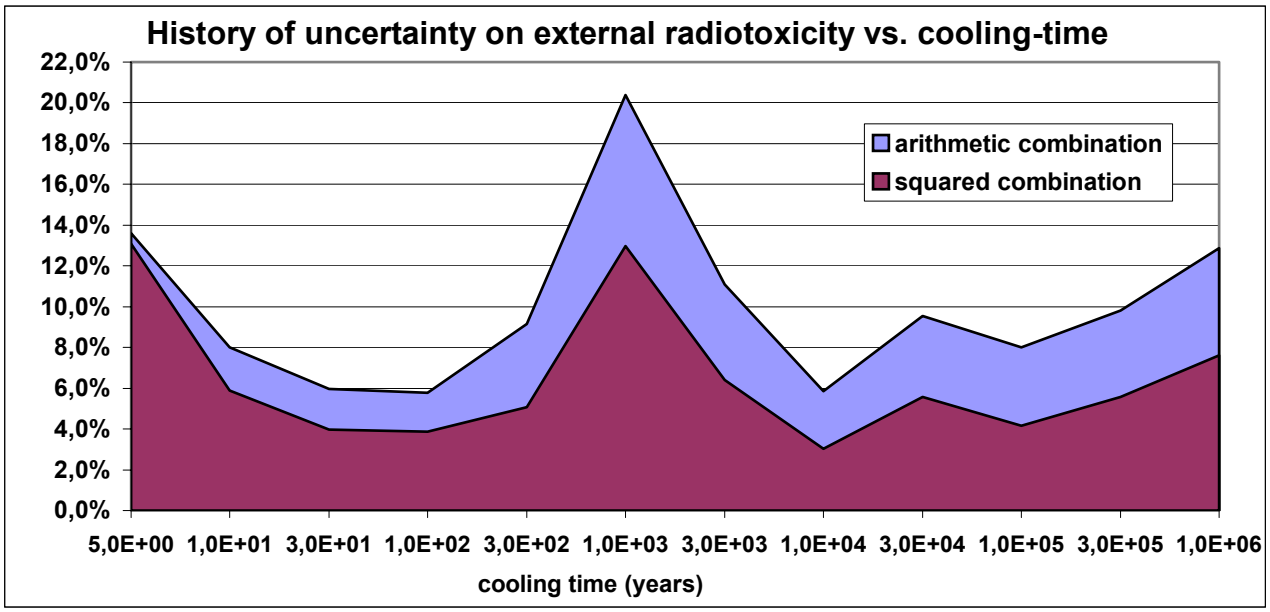

Fig. 4. - History of uncertainty on external-radiation toxicity vs. cooling time.

Results obtained on uncertainty analysis give an indirect validation of the computational procedure adopted, which appears to be quite performing and robust.

4. Conclusion

The present technology of nuclear energy production generates a long-term risk source, which is roughly proportional to the mass and composition of discharged nuclear waste. Several parameters are considered as indicators of risk, such as the external-radiation toxicity and the residual power.

The paper analyzes the main sources of uncertainties affecting these parameters and investigates how uncertainties can raise and propagate through the computational procedure.

The paper also gives an estimation of the overall uncertainties of the evaluated external-radiation toxicity and residual power, which turn out to be bounded at 20% peak over a cooling-time period spanning one-million years.

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