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Impact of different correlation structures in cross-section covariance matrices on the inventory and inventory-related parameters

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Abstract

At present, cross section correlation data are very poorly known. This work can be helpful to evaluate the effort that the international community should devote to the improvement of correlation data for inventory prediction in nuclear systems.

Our purpose is to assess the impact of different correlation structures on the uncertainties in relevant fuel cycle and repository parameters, using fixed variances/diagonal values in the cross section covariance matrices. At this stage, only correlation in energy is taken into account, and no correlation among different types of nuclear reactions or different isotopes is considered.

To accomplish this goal, the inventory code ACAB is used to estimate the uncertainties in the actinide concentrations of the irradiated fuel, related decay heat and dose (radiotoxicity) in a representative ADS irradiation scenario. It is shown that the nature of the introduced correlations is very relevant to estimate the overall uncertainty in those parameters, and then, the need of more scientifically based correlation data is clearly justified.

1. Introduction

It is clear that to study the impact of nuclear data uncertainties on relevant parameters in nuclear systems, a realistic set of activation cross section uncertainties and their correlation (covariance matrices) is required.

A wide review/compilation of uncertainties/correlations from the most recent activation data files, general-purpose evaluated nuclear data files and bibliography proposals has been performed (Sanz et al., 2006).

- Regarding activation data files, such as EAF2007/UN (Forrest, 2007), cross section uncertainties up to 20 MeV are provided in three energy ranges (one for threshold reactions). It is assumed that errors in all the groups of a particular energy range are 100% correlated while errors in the different energy ranges are 0% correlated. Any other

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type of correlation (among reactions, etc.) is ignored.

- Regarding general-purpose evaluated nuclear data files, both multigroup covariance matrices with energy correlations and multigroup covariance matrices correlating different isotopes and reaction types can be obtained, but only for a few number of nuclides (Kodeli et al., 2006). In addition, the accuracy and validation of these data is still a major concern (Aliberti et al., 2006).
- Regarding bibliography proposals, the multigroup covariance matrices produced by ANL (Palmiotti et al., 2005) are likely the most used correlation data nowadays in ADS applications. These data are just a first approach that includes the same energy correlations for only 41 isotopes (19 actinides ranging from Th-232 to Cm-245) and 3 reactions of importance in inventory calculations ($\sigma_{n,2n}$, $\sigma_{capt,}$ $\sigma_{fission}$).

It can be concluded that correlation data are very poorly known at present. To evaluate the effort that the international community should devote to the production of more correlation data for use in inventory codes, the purpose of this work is to assess the impact of different correlation structures using fixed uncertainties/variances on the uncertainties in the computed isotopic inventory and associated nuclear related parameters. No correlation among different reactions or different isotopes has been considered at this stage, but only correlation in energy.

To carry out this work, the inventory code ACAB (Sanz, 2000) is used to propagate the activation cross section uncertainties to the actinide composition of the irradiated fuel and related responses in a representative ADS irradiation scenario. The applicability of the code to deal with ADS systems has been proved within the frame of the EU Integrated Project EUROTRANS (Eurotrans, 2005), where the performance of different methodologies to address uncertainty estimations was discussed (Sanz et al., 2007a).

2. Problem definition

One of the preliminary conceptual designs of the European Facility for Industrial Transmutation (EFIT) recently defined within the EUROTRANS project is used here for the first time in this kind of analysis (Eurotrans, 2005). Its basics characteristics are: core cooled by pure lead, thermal power 400 MW, initial total mass of actinides 2.074 tonnes (fuel initial actinide composition shown in column 2 of Table 1), and 150 GWd/tHM discharge burn-up, corresponding to an equilibrium cycle. The fuel behaviour after multiple recycling can be investigated simulating consecutive irradiation cycles inside ADS, corresponding to discharge burn-up ranging from 150 to ≈800 GWd/tHM.

The problem chosen as reference for this study consists of the uncertainty estimation of the spent fuel actinide inventory and decay heat and dose for 500 GWd/tHM discharge burn-up. In the calculations, a constant neutron environment is assumed for all the irradiation period, both a neutron spectrum (average energy <E>= 0.3749 MeV) and flux intensity ($3.12 \cdot 10^{15}$ n/cm^2·s). With this irradiation conditions we reach the requested burn-up at 3250 days. The assumed neutron flux and spectrum are representative of the equilibrium cycle and were taken from detailed neutronic calculations corresponding to a given cell in the interior of the core (Alvarez et al., 2007).

3. Methodology

Concerning to actinides, the ANL covariance matrices produced by Palmiotti et al. (2005) are probably the most used data set at present for uncertainty estimates in actinide inventory prediction. The matrices are in a 15-multigroup structure (between 20 MeV and thermal energy), and consist of a set of uncertainties (diagonal values) and a set of energy correlations. The same correlation matrix is assumed for all isotopes and reactions, under the form of full energy correlation in 5 energy bands (see Fig. 1).

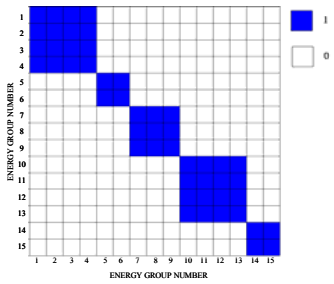

Fig. 1. Energy group structure and proposed energy correlation by ANL. Groups given in decreasing energy.

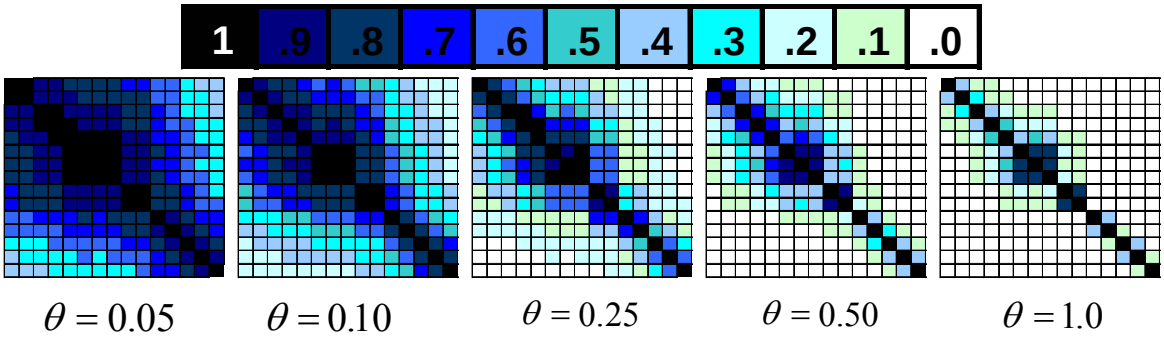

Fig. 2. Parametric correlation matrices in 15 groups generated from the proposed correlation model based on parameter θ.

In addition to use the ANL covariance matrices, we will use covariance matrices generated with the fixed ANL variance/diagonal data set but with different correlation structures.

We propose a simple correlation model to generate several parametric correlation matrices. These matrices are produced in the ANL 15-group structure. The rational followed in the generation of these matrices is as follows. Let us assume that the energy range is divided in G groups and let [E_1, E_2, …, E_G] be the energy mean values or centre points of each group. The correlation among the groups with energies E_i and E_j is defined by the equation

$$c_{ij} = \exp\left[-\theta\left|\log E_i - \log E_j\right|\right] \qquad (1)$$

where θ is a positive parameter between 0 and ∞, that can be considered as a energy-correlation range parameter (see Fig. 2). When θ is large, the correlations are low and *vice versa*, when θ is small, the correlations are high. In the extreme cases, θ = ∞, all the coefficients c_{ij} are zero (no correlation) and when θ = 0, all the coefficients are 1 (full correlation). For a given value of θ, the correlation c_{ij} decreases as the groups are more distant.

The best-estimated/mean cross-section values have been taken from the EAF-2007 library (Forrest, 2007), and processed to the required ANL 15-multigroup structure. Selecting as cross section values those of EAF seems a reasonable option since it is the most complete activation library, which includes all the nuclides and reactions of interest.

To study the effect of the different covariance structures in the actinide inventory and related responses, the Monte Carlo (MC) uncertainty approach in the code ACAB has been used. It is able to deal with multigroup covariance matrices in any arbitrary group structure and with any kind of correlations. The MC approach is based on using a log normal joint probability distribution of the cross sections for each reaction. Here the results are obtained for a 1000 history-sampling.

There is no a well-founded justification to apply the ANL covariance matrices to a different best-estimated cross section library. However, the objective of this work is not to provide a confident set of uncertainty estimates of relevant parameters of a nuclear system, but to assess how those uncertainties would be affected by the correlation structure.

4. Results: uncertainty evaluation

The predicted nominal values (no uncertainties considered) of the actinide concentrations at the end of the 3250-day irradiation period are given in the third column of Table 1.

4.1. Uncertainties in the actinide concentrations at end of irradiation

The different set of results concerning actinide concentrations are provided in Table 1. In the first set, only the "diagonal" values of the ANL covariance matrices are used (any kind of correlations is neglected). The results obtained are given in column 4 of Table 1.

In the second set, the full ANL covariance matrices are considered, i.e., including the energy correlation assigned in (Palmiotti et al., 2005). The results are provided in column 9.

The third set uses a series of correlation parametric matrices, generated according with Eq.

(1) for the following values of θ: θ=1, θ=0.5, θ=0.25 and θ = 0. The results are provided in columns 5-8.

Table 1
Relative errors (ratio between standard deviation and the mean, in %) for actinide concentrations at the end of 3250-day irradiation, using ANL uncertainties with no correlation (diagonal values, column 4) and different correlation matrices (parametric correlations, columns 5-8, and ANL correlation, column 9). Initial and final concentrations for an initial actinide mass of 2.074 ton are shown in columns 2 and 3.

Isotope	Initial (10^{24} at.)	Final (10^{24} at.)	Relative error of the final concentrations (%)					
			No correl	Theta (θ) 1	0.5	0.25	0	ANL correl
^{234}U	76.7	45.7	3.6	4.7	5.7	6.7	8.5	5.7
^{235}U	18.3	14.1	3.5	4.5	5.6	6.7	8.8	5.4
^{236}U	25.4	21.8	2.9	3.9	4.8	5.8	7.5	4.8
^{237}U	0.000002	0.04	3.4	4.5	5.6	6.7	8.7	5.6
^{238}U	0.1	0.1	1.1	1.5	1.9	2.2	3.0	1.9
^{237}Np	224.5	33.6	6.5	8.4	10.5	12.5	16.0	10.3
^{238}Np	0.000006	0.1	2.8	3.7	4.5	5.3	6.7	4.5
^{239}Np	0.0003	0.0003	2.7	3.4	4.3	5.2	7.0	4.3
^{238}Pu	426.0	221.0	5.1	6.8	8.5	10.3	13.7	8.1
^{239}Pu	521.4	133.7	4.1	5.3	6.7	8.0	10.4	6.3
^{240}Pu	1726.0	803.3	3.0	4.0	5.0	6.1	8.1	4.8
^{241}Pu	312.8	188.3	4.1	5.5	7.0	8.5	11.4	6.5
^{242}Pu	749.8	467.3	2.7	3.5	4.4	5.3	6.9	4.2
^{243}Pu	-	0.03	2.5	3.3	4.1	4.9	6.5	4.0
^{244}Pu	0.2	0.2	1.5	1.9	2.4	2.9	3.9	2.4
^{241}Am	350.2	73.0	5.4	7.2	9.0	10.8	14.1	8.7
^{242}Am	0.0004	0.04	2.4	3.2	4.0	4.9	6.5	3.9
^{242m}Am	29.6	4.8	8.3	11.0	13.8	16.7	22.0	12.5
^{243}Am	314.3	190.4	4.4	5.7	7.2	8.7	11.6	7.1
^{242}Cm	0.3	8.6	2.5	3.3	4.1	5.0	6.6	3.9
^{243}Cm	3.1	2.0	21.6	28.2	35.4	42.8	56.5	33.9
^{244}Cm	267.1	272.3	8.1	10.4	12.8	15.4	20.3	13.1
^{245}Cm	78.2	74.3	14.5	19.3	24.5	29.8	39.9	23.3
^{246}Cm	52.0	51.7	9.9	13.1	16.6	20.2	27.0	15.9
^{247}Cm	11.2	11.0	7.0*	9.3*	11.8*	14.4*	19.3*	11.3*
^{248}Cm	8.3	9.8	1.8*	2.4*	3.0*	3.7*	5.1*	2.9*
^{249}Bk	-	0.4	1.4*	1.9*	2.4*	2.9*	4.0*	2.3*
^{249}Cf	-	0.8	0.8*	1.0*	1.3*	1.6*	2.2*	1.3*
^{250}Cf	-	0.5	0.5*	0.7*	0.9*	1.1*	1.5*	0.9*
^{251}Cf	-	0.1	0.3*	0.4*	0.5*	0.6*	0.9*	0.5*
^{252}Cf	-	0.006	0.2*	0.3*	0.3*	0.4*	0.6*	0.3*

* Because of the lack of ANL uncertainty information from Cm-246 to Cf-252, the marked uncertainty values are meaningless. These uncertainty estimates are only due to the cross section uncertainties for actinides from U-234 to Cm-245.

In discussing results of Table 1, we have to take in mind that ANL data include uncertainty information only for 19 actinides in the range from Th-232 to Cm-245. In consequence, the obtained results are meaningful for the actinides within that range. Results for Cm-246 can be also of some meaning, since it is mainly generated from Cm-245. For the other nuclides in the table (from Cm-247 to Cf-252), the estimates are meaningless because of the lack of uncertainty information.

It is worth noting that regarding target accuracy on actinide concentration prediction (mainly related with the ADS transmutation potential), the assigned value is ± 5% (Sanz et al., 2007b).

Results given in columns 4-9 show that the nature of the introduced correlations is very relevant to estimate the uncertainty in the nuclide concentration. Assuming no correlation, most of the nuclides exhibit concentration uncertainties below the target value of 5%; there are three nuclides with uncertainty values a little higher than 5% (in this group the nuclide with a higher uncertainty in the concentration is Am-242m); and there are two nuclides that clearly show a concentration uncertainty far above 5% (Cm-243 and Cm-245). However, assuming full correlation ($\theta = 0$), it can be seen that all the actinides (except one) exhibit uncertainties above the required target accuracy, ten of them with uncertainties higher than 10%. In summary, depending on whether correlations are accounted for or not, the final uncertainties on the actinide inventory can vary by a factor of ~2.

It can be also seen that the uncertainties in concentrations obtained assuming the ANL energy correlation matrix are very similar to those obtained assuming a correlation range parameter θ around to 0.5 (only a little bit higher than 0.5).

4.2. Uncertainties in the response functions

The decay heat in ADS is of interest mainly related with reprocessing and waste conditioning and storage. The data for decay heat predictions have been taken from the EAF-2007 decay library (Forrest, 2007). For our reference problem, loaded with minor actinides, we have found that actinide contribution to decay heat plays the major role from one-year cooling time, in particular due to the presence of Cm-244.

The uncertainties in the actinide decay heat value, for an initial actinide mass of 2.074 ton, for the different cross-section covariance matrices have been evaluated along cooling time. Results are summarized in Fig 3. It can be seen that as the correlation increases, the relative errors become higher. The maximum uncertainty value appears after ~5 years of cooling time for all the correlation structures, going from 6.6% for the uncorrelated case to 16.4% when assuming full correlation ($\theta = 0$). It can be concluded that the correlation structure effect in the decay heat predition is very significant. If a decay target accuracy of ± 10% is required (Sanz et al., 2007b), depending on the correlation structure, the decay heat can exhibit uncertainties far above or below the target value.

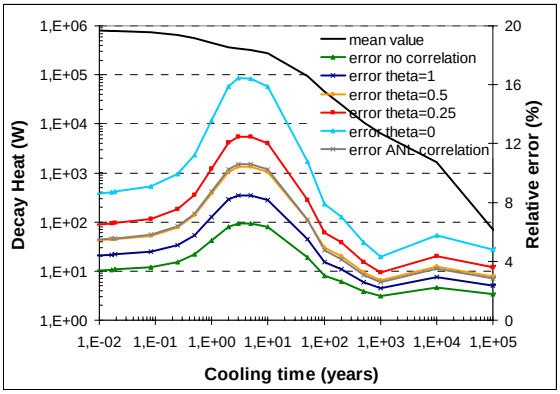

Fig. 3. Uncertainties in the decay heat due to actinides along cooling time, for an initial actinide mass of 2.074 ton and 500 GWd/tHM burnup.

To give some indication of the potential biological hazard of actinides, the EAF-2007 library of dose coefficients (Forrest, 2007) has been used to compute the committed effective dose equivalent (CEDE) by inhalation and ingestion.

In Fig. 4 the uncertainties in actinide CEDE by inhalation due to different cross-section covariance structures are illustrated. The results show again the importance of the correlations if a radiotoxicity target accuracy of ± 10% is required (Sanz et al., 2007b). Similar tendencies are observed when analyzing the CEDE by ingestion.

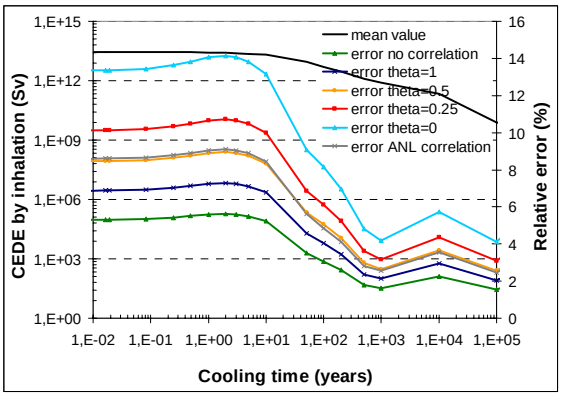

Fig. 4. Uncertainties in the CEDE by inhalation due to actinides along cooling time, for an initial actinide mass of 2.074 ton and 500 GWd/tHM burnup.

The actinide neutron emission is also an important response function when analyzing ADS fuel cycles, particularly related to the ADS fuel fabrication. To compute the neutron source from (α,n) reactions and spontaneous fission, the EAF-2007 libraries of decay data, differential ranges and cross sections for α particles have been used (Forrest, 2007). For the fuel composition of our reference problem, the (α,n) neutron source is found to be mainly due to the presence of Mg-isotopes (targets). Regarding to the spontaneous fission neutrons, the major contributors are Cm-246, Cm-248 and Cf-250 (relative contribution ~80% for all cooling times). Because of the lack of ANL uncertainty information for the mentioned isotopes, it is meaningless to evaluate the impact of the proposed covariance matrices in the neutron emission. Once that information becomes available, this analysis could be carried out.

5. Conclusions

Different kinds of energy correlations in cross-section covariance matrices, with the same variance/diagonal data set, have been considered: uncorrelated, ANL correlation and parametric correlations based on a parameter θ (θ=1, θ=0.5, θ=0.25 and θ=0). The impact of the different correlations on the actinide inventory and related parameters has been assessed. The results show that the correlation structure effect is very important in the prediction of the concentrations at the end of the irradiation period, as well as in the prediction of the decay heat and dose along the cooling time. Depending on whether correlations are accounted for or not, the final uncertainties on the actinide inventory and related responses can vary by a factor of ~2. In consequence, the parameters can exhibit uncertainties far above or below the corresponding required target accuracies.

The need of more scientifically based correlation data is clearly justified, concluding that more effort in defining correlation data for actinide prediction is needed.

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