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Author(s)	Chiba, Go; Honta, Keisuke
Citation	Journal of nuclear science and technology, 57(12), 1265-1275 https://doi.org/10.1080/00223131.2020.1780993
Issue Date	2020-12
Doc URL	https://hdl.handle.net/2115/83365
Rights	This is an Accepted Manuscript of an article published by Taylor & Francis in Journal of nuclear science and technology on Dec 2020, available online: http://www.tandfonline.com/10.1080/00223131.2020.1780993 .
Type	journal article
File Information	aper.pdf



Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

Go Chiba^{1*}, Keisuke Honta¹

¹*Division of applied quantum science and engineering, faculty of engineering, Hokkaido University, Kita*

13 Nishi 8, Kita-ku, Sapporo 060-8628, Japan

The passive gamma spectroscopy (PGS) is a useful technique to extract information on spent nuclear fuels without any destructive actions. This method requires a correlation between number densities (NDs) of target nuclides, and it is generally estimated by numerical simulation. Therefore, prediction accuracy of these nuclides generations is one of key issues in PGS. Nuclear data used in nuclear fuel depletion calculations is one of dominant uncertainty sources, so we quantify nuclear data-induced uncertainties of NDs of six fission product nuclides, which are important in PGS: Ce-144, Cs-134, -137, Ru-106, Sb-125 and Eu-154. Generation mechanisms of these nuclides are quantitatively investigated through sensitivities of these NDs to nuclear data, which can be calculated by the depletion perturbation theory. With the sensitivities and covariance data of nuclear data, uncertainties of NDs of these nuclides are quantified with the JENDL libraries and others. The uncertainties of Ce-144, Cs-137 and Ru-106 are less than 2%, and that of Sb-125 is around 6%. In these uncertainties, fission yields uncertainties are dominant. On the Cs-134 and Eu-154 generations, total uncertainties are around 5% and uncertainties of (n, γ) cross sections are dominant. Those calculations are carried out with BWR pincell models, but it is also confirmed that results obtained with a BWR fuel assembly model are quite similar to those in the pincell models.

Keywords: passive gamma spectroscopy, fission products nuclides, sensitivity analysis, uncertainty quantification

*Corresponding author. Email: go_chiba@eng.hokudai.ac.jp

1. Introduction

Spent (or irradiated) nuclear fuels should be composed of various radioactive nuclides including fission product (FP) nuclides. FP nuclides are generated by nuclear fission reactions, and their generation probabilities are inherently determined dependent on fissile nuclides originating the fission reaction and energy of neutron inducing the fission reaction. Thus the amounts of FP nuclides in spent fuels should include quite useful information to know the characteristics about the spent fuels. The amount of radioactive nuclides in a material can be quantified by measuring gamma-rays emitted through radioactive decay of these nuclides. If gamma-rays emitted from concerned radioactive nuclides are measurable, the amount of these nuclides can be quantified, and these can be utilized to extract the information about the material. This technique to quantify the amount of radioactive nuclides in a material through measuring emitted gamma-rays is widely known as the *gamma spectroscopy*, and if gamma-rays are measured without any actions like neutron injections to this material, this is referred to as the *passive gamma spectroscopy*.

Some FP nuclides are accumulated linearly with respect to fuel burnup, so if the total amount of these FP nuclides included in a spent fuel can be quantified, fuel burnup of this spent fuel can be estimated. Such FP nuclides are called a burnup indicator, and Cs-137 is well known one[1]. Also even if the total amount of FP nuclides cannot be quantified, if a ratio of the amounts of two FP nuclides, of which one is accumulated quadratically (linearly) and the other is linearly (almost constant) with fuel burnup, can be quantified, fuel burnup can be estimated also from this ratio because this ratio should be linearly dependent on fuel burnup. Radioactivity ratios of Cs-134/Cs-137, Eu-154/Cs-137 and Ru-106/Ce-144 are those examples[1]. The passive gamma spectroscopy focusing on these FP nuclides can be utilized to quantify fuel burnup.

The passive gamma spectroscopy can be applied also to quantification of inventories of special nuclear materials like uranium and plutonium in spent fuels from a viewpoint

of no diversion of these materials. It becomes possible by measuring gamma-ray emitted from not the target nuclides but measurable nuclides, and by using a correlation between number densities of this measurable nuclide and the target nuclide of special nuclear materials.

Generally these information such as fuel burnup and nuclides inventories can be accurately estimated by numerical simulations for spent fuels in normal nuclear power plants. However, when a severe accident accompanying core melting occurs in a nuclear power plant, it becomes quite difficult to specify irradiation history of melted fuels. In such cases, the passive gamma spectroscopy is a powerful tool, and it was applied to the decommissioning process of the Three Mile Island's unit 2 reactor[2] where FP nuclides such as Cs-134, -137, Ce-144, Eu-154, Ru-106 and Sb-125 were concerned. In addition, possibility of the application of the passive gamma spectroscopy to the decommissioning for the Fukushima Dai-ichi power plant has been recently discussed[3].

In the methods of nuclides inventory quantification with the passive gamma spectroscopy, correlations between number densities of some specific nuclides should be prepared, and results of numerical simulations are generally used. Thus, one of the important points in these methods is the prediction accuracy of inventories of the concerned nuclides. In numerical simulations for nuclides inventories, several uncertain sources should be considered, and one of the dominant sources is *nuclear data*, which describes interaction of neutrons with nuclides. Recently a lot of works about nuclear data-induced uncertainty quantification in nuclides inventories have been carried out, and relevant methods and tools have been developed. Among these activities, the work being conducted at Hokkaido University is quite unique; it is based on the sensitivity-based procedure, in which importance functions for concerned nuclides inventories are defined and calculated in a complicated system like a light water reactor (LWR) fuel assembly[4]. With the advanced methods and tools developed through these activities, it becomes possible now

to clearly understand the mechanism of nuclides generation during nuclear fuel depletion by calculating sensitivities of nuclides inventories with respect to relevant nuclear data. Furthermore, by using these sensitivities it also becomes possible to quantify nuclear data-induced uncertainties of these nuclides inventories using uncertainty information on nuclear data: *covariance data* of nuclear data.

In the present work, we focus on the generation of six FP nuclides in uranium-dioxide-loaded boiling water reactor (BWR) fuel assemblies. These nuclides are Ce-144, Cs-134, -137, Ru-106, Sb-125 and Eu-154, and are important in the passive gamma spectroscopy.

Generation mechanism is quantitatively investigated by sensitivity analyses, and by using these sensitivities and the covariance data, nuclear data-induced uncertainties of these nuclides inventories are quantified.

Section 2 describes the basic data, theory and procedure about sensitivity analyses and uncertainty quantification for nuclides number densities during nuclear fuel depletion. Section 3 presents problem specifications for nuclear fuel depletion and all the numerical results, and the conclusion is given in Section 4. Also, in the appendix, an additional study about covariance matrix generation methods of fission yields data is presented.

2. Data, theory and procedure of sensitivity analyses and uncertainty quantification

2.1. Nuclear data and their covariance data

When nuclides generations in nuclear fuels during irradiation or nuclear reactor operation are concerned, numerical calculations about nuclear fuel depletion can be employed. Nuclear data relevant to (or used in) nuclear fuel depletion calculations are enormous: neutron-nuclide reaction cross sections, reaction branching ratios, fission yields, radioactive decay-relevant data such as decay constants and decay branching ratios, etc. In the present study, uncertainties of reaction cross sections, fission yields, decay constants and

decay branching ratios are taken into account. Generally uncertainty information on these nuclear data are provided as covariance data in evaluated nuclear data libraries. We will use the evaluated nuclear data libraries JENDL-4.0[5], JENDL/FPY-2011 (revised version) and JENDL/FPD-2011[6, 7] for reaction cross sections, fission yields and decay-relevant nuclear data, respectively.

In JENDL-4.0, covariance data are evaluated for almost all the important actinoids nuclides, and those are taken into account in the present study. However, JENDL-4.0 provides no covariance data for reaction cross sections of FP nuclides, so we will use tentatively covariance data for some important FP nuclides taken from the newest version of the US library, ENDF/B-VIII.0[8]. In the following uncertainty quantification calculations, covariance data of reaction cross sections will be used together with sensitivities in multi-group representations, so covariance data in multi-group representation, obtained with the NJOY-99 code[9], are used.

In JENDL/FPY-2011, fission yields data are given to 1,299 FP nuclides. Only variance data are provided to fission yields data in JENDL/FPY-2011 due to the format limitation, but it has been pointed out by many authors that correlations in fission yields data among different nuclides should be taken into account to properly conduct uncertainty propagation calculations[6,10]. While there have been several methods to take into account the correlations, we will adopt the generalized least-square (GLS) updating procedure introducing information about chain yields and some physical constraints about fission reactions[10]. Detail of the GLS updating procedure will be described in the appendix. There is another simple method based on a single mass chain proposed by Devillers[11], and a difference between these two methods and its impact will be discussed also in the appendix. On the decay branching ratio, correlations due to the physical constraints (normalization condition) are taken into account.

2.2. Sensitivity calculations with the depletion perturbation theory

In the present work, we will handle with a sensitivity of nuclide number density with respect to nuclear data in BWR single pincell models and a BWR assembly model. When a number density of nuclide i at specific burnup and nuclear data j are denoted to as N_i and σ_j , respectively, a sensitivity of N_i to σ_j , s_j^i , is defined as

$$s_j^i = \frac{\partial N_i}{\partial \sigma_j} \cdot \frac{\sigma_j}{N_i}. \quad (1)$$

Sensitivities are very useful to understand mechanism of nuclides generation during nuclear fuel depletion. For example, on FP nuclides generation, origins of the concerned FP nuclide can be specified by observing sensitivities of this nuclide number density with respect to fission yields data.

Since quite a large number of nuclear data are used in nuclear fuel depletion calculations as mentioned above, numerical differentiation to obtain s_j^i is unrealistic. In the field of reactor physics, the generalized perturbation theory for nuclear fuel depletion problems, *depletion perturbation theory* (DPT), has been developed and well established[12,13]. Since the seminal works cited here, applicability of DPT has been enhanced, and now this theory can be applied to nuclear fuel depletion problems of LWR fuel assemblies including burnable absorbers[4].

In the present work, we will use DPT to calculate sensitivities of number densities of concerned nuclides during nuclear fuel depletion with respect to nuclear data such as neutron-nuclide reaction cross sections, fission yields, decay constants and decay branching ratios. A brief description of DPT is provided in the following.

Let us consider a nuclear fuel depletion calculation, and nuclides number densities of this fuel at time t are represented by a vector as $\mathbf{N}(t)$. We assume that an initial condition of the nuclides number density is provided at t_0 . Usually we divide the whole depletion time period into several time steps, and a burnup matrix is assumed constant during each time step. When we divide the period into I time steps, the following burnup equation

can be defined:

$$\frac{d\mathbf{N}(t)}{dt} = \mathbf{M}_i \mathbf{N}(t), \quad (t_{i-1} \leq t \leq t_i, \quad i = 1, 2, \dots, I), \quad (2)$$

where $\mathbf{M}_i$ is a burnup matrix of step i , and is defined from microscopic cross sections and neutron flux at t_{i-1} . The neutron flux at t_i , Φ_i , is defined from the following neutron transport equation:

$$\mathbf{B}_i \Phi_i = 0, \quad (3)$$

where the operator $\mathbf{B}_i$ is defined from $\mathbf{N}(t_{i-1})$.

A sensitivity of number density of nuclide j at t_I , $N_j(t_I)$, with respect to a nuclear data σ can be calculated with the following equation:

$$\frac{dN_j(t_I)}{d\sigma} = \sum_{i=1}^I \left(\int_{t_{i-1}}^{t_i} \mathbf{w}^T(t) \frac{\partial \mathbf{M}_i}{\partial \sigma} \mathbf{N}(t) dt + \left\langle \mathbf{\Gamma}_i^{x\dagger} \frac{\partial \mathbf{B}_i}{\partial \sigma} \Phi_i \right\rangle \right). \quad (4)$$

Note that it is assumed here that neutron flux normalization does not depend on any reaction cross sections for simplicity.

In Eq. (4), a vector $\mathbf{w}(t)$ represents adjoint number densities, and is defined as

$$\frac{d\mathbf{w}(t)}{dt} = -\mathbf{M}_i^T \mathbf{w}(t), \quad (t_{i-1} < t < t_i), \quad (5)$$

with a final condition $\mathbf{w}(t_I) = \mathbf{e}_j$. Note that $\mathbf{e}_j$ is a vector having the j th component of unity with all other components zero. At boundaries between subsequent time steps, discrete changes known as *jump condition* should be considered for $\mathbf{w}(t)$, but its detail is omitted here. A generalized adjoint neutron flux represented as $\mathbf{\Gamma}_i^\dagger$ in Eq. (4) is defined as

$$\mathbf{B}_i^\dagger \mathbf{\Gamma}_i^\dagger = \mathbf{S}_i^\dagger. \quad (6)$$

Detail of the source term $\mathbf{S}_i^\dagger$ is also omitted here.

Numerical calculations of nuclides number densities with DPT are generally based on numerical methods for nuclear fuel depletion calculations, which consist of neutron flux distribution calculations by solving the neutron transport equation and nuclides transmutation calculations. In the present study, all these calculations are carried out with a

reactor physics code system CBZ[14] which has been developed at Hokkaido University.

In nuclear fuel depletion calculations, generation and transmutation of FP nuclides should be numerically simulated. Since there are a large number of FP nuclides over 1,000, it is computationally heavy to treat all these FP nuclides explicitly in nuclides transmutation calculations especially in a complicated system like LWR fuel assemblies. Furthermore, this issue about computational burden becomes serious when the DPT calculations are carried out since a huge amount of data during nuclear fuel depletion should be stored. In order to solve these problems, two types of fuel depletion calculations are carried out in the present study. The first one is to use a detail nuclides chain model in which all the FP nuclides are explicitly treated, and the second one is to use a simplified nuclides chain model in which a limited number of important FP nuclides are treated. In the second type of calculations, we use a chain model consisting of 197 FP nuclides; 193 nuclides of which are chosen in one of the FP transmutation chain model adopted in the SRAC-2006 code[15] and the following four nuclides are added: Nd-149, Sn-125, I-133 and Gd-153. This nuclide chain is one of generally-used ones in LWR fuel depletion analyses with CBZ.

For resonance calculations, the advanced Bondarenko method[16] is adopted, and medium-wise 107-group cross section data are generated. In fuel assembly calculations, background cross sections required in the advanced Bondarenko method are calculated from the Dancoff factors evaluated with the whole assembly model[17]. Neutron transport calculations with the 107-group cross sections including generalized adjoint neutron flux calculations for DPT are performed with a neutron transport calculation module MEC based on the method of characteristics, and reaction rates are calculated for all the media containing nuclear fuels. Scattering anisotropy is taken into account by the P0 transport approximation.

Nuclides transmutation calculations are carried out with the reference detail chain

model or the simplified chain model as mentioned above. Nuclides transmutation equations including adjoint problems in DPT are solved by the matrix exponential method with the mini-max polynomial approximation method[18,19]. For LWR assembly calculations, the predictor-corrector method is employed.

2.3. Uncertainty quantification with sensitivities and covariance data

Nuclear data-induced uncertainties of nuclides number densities can be easily quantified if sensitivities of nuclides number densities with respect to nuclear data are available. Nuclear data-induced uncertainty of N_i (a variance of N_i), V_{N_i} , can be calculated with the following equation:

$$V_{N_i} = \sum_j \sum_{j'} s_j^i s_{j'}^i \text{cov}(\sigma_j, \sigma_{j'}), \quad (7)$$

where $\text{cov}(\sigma_j, \sigma_{j'})$ is a covariance between σ_j and $\sigma_{j'}$, and a covariance matrix covering all the nuclear data considered is defined as $\mathbf{V}_\sigma$. When we define a sensitivity vector $\mathbf{s}^i$ as $\mathbf{s}^i = (s_1^i \ s_2^i \ \cdots \ s_J^i)^T$, where J is the total number of concerned nuclides number densities and T is for vector and matrix transposition, using $\mathbf{V}_\sigma$ and $\mathbf{s}^i$, V_{N_i} can be rewritten as

$$V_{N_i} = \mathbf{s}^{iT} \mathbf{V}_\sigma \mathbf{s}^i. \quad (8)$$

Covariance between N_i and $N_{i'}$, $\text{cov}(N_i, N_{i'})$, can be also calculated as

$$\text{cov}(N_i, N_{i'}) = \mathbf{s}^{iT} \mathbf{V}_\sigma \mathbf{s}^{i'}, \quad (9)$$

and a covariance matrix covering all the concerned number densities, $\mathbf{V}_N$, can be calculated as

$$\mathbf{V}_N = \mathbf{S}^T \mathbf{V}_\sigma \mathbf{S}, \quad (10)$$

where $\mathbf{S} = (\mathbf{s}^1 \ \mathbf{s}^2 \ \cdots \ \mathbf{s}^I)$, where I is the total number of nuclear data considered.

When the simplified chain model is used, sensitivities of nuclides number densities to nuclear data are calculated within the framework of this simplified chain model, so in

uncertainty quantification calculations with sensitivities and covariance data, covariance data which is consistent with this simplified chain model should be used. This issue has been discussed, and the numerical method and tool have been already established in our previous work[20]. Its detail is omitted here, but in the present work, we will follow this previous work.

3. Numerical results

3.1. *Problems specification*

In the present study, we focus on a set of BWR single pincell models and a BWR fuel assembly model.

We use two of the BWR pincell models developed for cross section data library generation for the ORIGEN code[21]: a model based on the STEP-1 assembly and a model based on the STEP-3 assembly. Geometric specifications and U-235 enrichments are different from each other as shown in **Table 1**. Power density is set to 25.6 MW/tU, and 0% void ratio for coolant is assumed. To see impact of void condition on calculation results, 70% void ratio condition is also considered for the STEP-3 model. Therefore totally three conditions will be treated as the BWR pincell models.

[Table 1 about here.]

About a BWR fuel assembly model, we will use the model developed through the OECD/NEA burnup credit benchmark phase-IIIC[22]. This model is based on a typical 9×9-type BWR assembly. Its horizontal geometrical specification is shown in **Figure 1**. The power density is set to 25.3 MW/tU, and three different void ratio conditions, 0%, 40% and 70%, are treated.

[Figure 1 about here.]

3.2. *Sensitivity calculations with pincell problems*

First, in order to specify the origins of the target FP nuclides generations, sensitivities of nuclides number densities at several fuel burnups to fission yields are calculated in the pincell problem of STEP-3 with 0% void ratio. In order to ease understanding, the simplified chain model consisting of 197 FP nuclides is used here, so contributions of short-lived FP nuclides are taken into account indirectly via their daughter nuclides treated in the simplified chain model.

Among six target nuclides, no neutron-nuclide reactions contribute to the generations of Ce-144, Cs-137, Ru-106 and Sb-125: those nuclides are accumulated by a fission reaction and subsequent radioactive decays in the specific decay chain. About the Cs-134 generation, contribution of (n, γ) reaction of Cs-133 is dominant rather than contribution from the decay chain to which Cs-134 is belonging; this means that the Cs-134 generation has a large sensitivity to fission yields of FP nuclides in the decay chain to which Cs-133 is belonging. Whereas these generation mechanisms are quite simple and have been well known, the generation mechanism of Eu-154 is much more complicated than the others. These will be described later in this section.

Figure 2 shows sensitivities of number densities of the target six nuclides at several fuel burnups to fission yields data. This figure shows sensitivities summed up over all the FP nuclides for each fissile nuclide occurring fission reactions. Generally, FP nuclides generated by U-235 have large sensitivity at the beginning of the depletion, but contribution of Pu-239 becomes large as fuel burnup increases. The Pu-239 contributions are relatively large in the cases of Ru-106 and Sb-125.

[Figure 2 about here.]

As mentioned above, only Eu-154 generation shows complicated sensitivity profiles for fission yields data. **Figure 3** shows sensitivities of Eu-154 number density to FP nuclide-wise fission yields. This figure shows that fission yields of FP nuclides whose mass number

is different from 154 have large sensitivities, and sensitivities of FP nuclides with low mass number become large as fuel burnup increases. This means that multiple (n,γ) reactions are important in the Eu-154 generation.

[Figure 3 about here.]

Next sensitivities of Eu-154 number density to one-group (n,γ) cross section are shown in **Fig. 4**. Note that the present DPT calculations yield sensitivities of nuclides number densities to cross sections in 107-group representation. To summarize this, one-group cross section sensitivity, which is obtained by summing up sensitivities of all the energy groups, is presented here. This figure shows that (n,γ) cross sections of several nuclides such as Eu-153, -154 and Sm-152 have large sensitivities. Note that among these three nuclides, covariance data are not evaluated for Eu-154 (and Sm-150 also) in ENDF/B-VIII.0. This will be discussed later. Based on these sensitivities, the generation mechanism of Eu-154 can be simply presented as **Fig. 5**. In this figure, FP nuclides which are explicitly treated in the simplified chain model are shown with bold-line boxes, and those which are neglected are shown with dashed-line boxes. Half-lives of unstable nuclides are also presented inside the boxes.

[Figure 4 about here.]

[Figure 5 about here.]

Sensitivities of number densities of Ce-144, Cs-134, -137, Ru-106 and Sb-125 at 40 GWD/t to one-group (n,γ) cross sections are shown in **Table 2**. This table shows that (n,γ) cross sections of these nuclides do not affect their own number densities, and that only Cs-133 (n,γ) cross section is important among them.

[Table 2 about here.]

3.3. *Uncertainty quantification calculations*

Uncertainties of nuclides number densities during nuclear fuel depletion are calculated from sensitivities and covariance data with the pincell models. In this calculation, the detail chain model is used, and three different conditions are considered. Total uncertainties are shown in **Figure 6**. Uncertainties of Ce-144 and Cs-137 are small and less than 0.5% during nuclear fuel depletion. Uncertainty of Ru-106 is around 1.5%, and those of the other nuclides, Cs-134, Sb-125 and Eu-154 are around several %. Generally, uncertainties are not significantly dependent on the conditions including fuel burnup. Only the uncertainty of Eu-154 is significantly dependent on fuel burnup.

[Figure 6 about here.]

Next the uncertainties of nuclides number densities in the model of STEP-3 with 0% void ratio are decomposed to nuclear data-wise uncertainties. This kind of evaluation can be easily realized in uncertainty quantification using sensitivity profiles. Results are shown in **Figure 7**. Fission yields data are dominant uncertainty sources in the number densities of Ce-144, Cs-137, Ru-106 and Sb-125, and reaction cross sections data are dominant in those of Cs-134 and Eu-154.

The fission yields-induced uncertainty in Sb-125 number density is much larger than the others, and this can be understood by comparing uncertainties given to the cumulative fission yields in JENDL/FPY-2011 as shown in **Table 3**. Note that fissile-wise contributions in fission-yields-induced uncertainties are presented in the appendix.

[Figure 7 about here.]

[Table 3 about here.]

On the Cs-134 number density, Cs-133 (n, γ) cross section is a dominant contributor to the total uncertainty. On the Eu-154 number density, Eu-153 and Sm-152 (n, γ) cross sections are dominant contributors to the total uncertainty. It should be reminded here that the covariance data for Eu-154 (n, γ) cross section are not evaluated in ENDF/B-

VIII.0 whereas this cross section is quite sensitive to the Eu-154 number densities. To quantify an impact of the Eu-154 (n, γ) cross section uncertainty on the Eu-154 number density, covariance data of this cross section are taken from the TENDL-2019 library[23], and uncertainty of Eu-154 number density is recalculated. Result is shown in **Figure 8**. When the covariance data of Eu-154 (n, γ) cross section are considered, uncertainty of Eu-154 number densities becomes much larger than the preceding results. **Figure 9** shows nuclide-wise uncertainties of Eu-154 number densities induced by (n, γ) cross sections. This suggests a possibility that the uncertainty of Eu-154 (n, γ) cross section has a large impact on the Eu-154 number densities.

[Figure 8 about here.]

[Figure 9 about here.]

Correlations are also calculated among different conditions and different burnups for each target nuclide. If strong correlations can be observed among different conditions/burnups, uncertainty quantification calculations can be done for one representative model, and this result can be used for extrapolation to different conditions/burnups. On the other hand, if correlations are weak, results should be dependent on used covariance data and the adequate extrapolation cannot be expected. Results are shown in **Fig. 10**. Note that the TENDL-2019 covariance data are used in this calculation. Parameters whose indices are from 1 to 8 are number densities of the STEP-3 model with 0% void ratio, those from 9 to 16 are of the STEP-3 model with 70% void ratio, and those from 17 to 24 are of the STEP-1 model with 0% void ratio. The parameters 1, 2,..., 8 are number densities at 5 GWD/t, 10 GWD/t, ..., 40 GWD/t and these arrangements are the same as for the parameters from 9 to 16, and those from 17 to 24. Generally there exist positive correlations among number densities of the same nuclide in different conditions and fuel burnups since a limited and specific nuclear data contribute to the uncertainties as mentioned above. Correlations among different conditions are generally quite strong, so one

representative model can be chosen and used to uncertainty quantification calculations of number densities of these nuclides. As for the correlations among different fuel burnups, number densities of Ce-144, Cs-134, -137 and Ru-106 are generally strong since dominant contributors to total uncertainties are not significantly dependent on fuel burnup. On the other hand, the uncertainties of number densities of Sb-125 and Eu-154 are significantly dependent on fuel burnup, so these should be paid attention in the actual uncertainty quantification work.

[Figure 10 about here.]

Finally, uncertainty quantification calculations of number densities of these six target nuclides are carried out with the BWR fuel assembly model. Here, averaged number densities over the whole assembly are concerned, and uncertainties are quantified at different fuel burnups and void ratios. In the whole-assembly fuel depletion calculations including DPT calculations, use of the reference detail chain model is unrealistic from a view point of computational time and memory requirement, so the simplified chain model and corresponding covariance data are adopted. Total uncertainties of the assembly-averaged number densities are shown in **Figure 11** with those of the pincell model of STEP-3 with 0% void ratio. The impact of void ratios on uncertainties of assembly-averaged number densities is very small, and difference in the uncertainties between the pincell model and the assembly model is negligible. These results suggest that a simple single pincell model can be utilized for uncertainty quantification calculations of number densities of the target six nuclides rather than the assembly model which requires heavy computational burden.

[Figure 11 about here.]

4. Conclusion

The passive gamma spectroscopy is a simple and useful technique to extract important information such as fuel burnup from spent nuclear fuels without any destructive actions.

This method requires a correlation between number densities of target nuclides, and it is generally estimated by numerical simulation. Therefore, prediction accuracy of these nuclides generations during nuclear fuel depletion is one of key issues in the passive gamma spectroscopy. Nuclear data used in nuclear fuel depletion calculations is one of dominant uncertainty sources. In the present work, we have quantified nuclear data-induced uncertainties of number densities of six fission product nuclides, which are important in the passive gamma spectroscopy: Ce-144, Cs-134, -137, Ru-106, Sb-125 and Eu-154.

Generation mechanisms of these target nuclides have been quantitatively investigated through sensitivities of these number densities with respect to nuclear data, which can be calculated efficiently by the depletion perturbation theory. Generally these nuclides are accumulated through a series of radioactive decays except Cs-134 and Eu-154. Cs-134 is accumulated mainly by (n,γ) reaction of Cs-133, and a decay series including Cs-133 is important in the Cs-134 generation. On the Eu-154 generation, several different decay chains and (n,γ) reactions contribute, and generation mechanism is much more complicated than the other nuclides.

With the sensitivities and the covariance data of nuclear data, uncertainties of number densities of these target nuclides have been quantified. Nuclear data-wise contributions to total uncertainties have been also quantified, and it has been found that fission yields data uncertainties are main contributors in the generations of Ce-144, Cs-137, Ru-106 and Sb-125. The uncertainties of the first three are less than 2%, and that of Sb-125 is around 6% due to large uncertainties in fission yields data. On the Cs-134 and Eu-154 generations, uncertainties of (n,γ) cross sections are dominant. On Eu-154, one of important uncertainty sources, (n,γ) cross section of Eu-154, is not evaluated in the recent nuclear data libraries such as JENDL-4.0 and ENDF/B-VIII.0, so it is strongly recommended to evaluate this uncertainty in future revision of these libraries.

The above calculations have been carried out with the single pincell models, and it

has been confirmed that results obtained with the fuel assembly model are quite similar to those in the pin cell models. This suggests that sensitivity and uncertainty analyses for these nuclides number densities are possible with a simple pin cell model.

In the present work, we have tentatively used covariance data for reaction cross sections of some FP nuclides which are inconsistent with the nuclear data used in the nuclides generation calculations. If consistent covariance data for these nuclear data are available, it becomes possible to evaluate more reasonably nuclear data-induced uncertainties for all these target nuclides. If the uncertainties are larger than that expected (or required) in actual use of the passive gamma spectroscopy, further effort to reduce these uncertainty is necessary. New nuclear data measurement or development of more advanced nuclear model is desirable, and also the uncertainty reduction can be attained by adopting nuclear data adjustment technique using integral data such as post-irradiation examination data.

Acknowledgement

A part of this work is supported by The Secretariat of the Nuclear Regulation Authority (S/NRA) in Japan. The authors greatly appreciate for Dr. Toshihisa Yamamoto and Dr. Tatsuya Fujita of S/NRA who have provided valuable comments on this work. One of the authors (G.C) appreciates also for Dr. Yasushi Nauchi of Central Research Institute of Electric Power Industry who have provided useful advice about the passive gamma spectroscopy.

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Appendix: Impact of covariance matrix generation methods for fission yields data

In the present work, we have applied the GLS updating method to generate the covariance matrix of fission yields data. In the GLS updating method, we have considered five constraints; chain yield, the number of protons, the number of neutrons, the number of total fission fragments and the number of fission fragments belonging to a peak of the light mass. In the first constraint, chain yields, which are equivalent with cumulative fission yields of stable FP nuclides at the end of each decay chain, are restricted within the variance of the cumulative yield evaluated in JENDL/FPY-2011. Note that the alpha-decay FP nuclides with long half-lives such as Rb-87, Cd-113, In-115, Te-123, La-138, Nd-144, Sm-147, -148 and Gd-152 are regarded as stable nuclides. On the second to fifth constraints, it is assumed that the corresponding quantities are well known and their uncertainty is negligible. Taking the second constraint for example, covariance matrix of the fission yield is updated based on the new information that the following quantity is preserved:

$$\sum_i z_i y_i = \text{Const.}, \quad (\text{A}\cdot 1)$$

where z_i and y_i are the number of protons and independent fission yield of FP nuclide i . In these constraints, it is also assumed that fission yields of light nuclides, whose mass number is less than fifteen, have no uncertainties.

On the other hand, there is another simple method proposed by Devillers[11]. In this method, decay processes other than the β^- decay are neglected, so decay chains are simplified to mass chains. In a sequential simple mass chain, the constraint on the mass yield can be analytically considered, and the resulting covariance matrix of the fission yields data can be analytically defined. This Devillers's method has been utilized in some previous works[6].

Figure 12 shows nuclides number densities uncertainties induced only by fission yields data. These number densities are those at 40 GWD/t of the STEP-3 pincell model with 0% void ratio. The covariance matrix of the fission yields is generated by Devillers' method and two variants of the GLS updating method. In the first GLS method denoted to as GLS-1 in this figure, only the first constraint about the chain yield is considered, and in the second GLS method denoted to as GLS-2, all the constraints are taken into account. Generally differences among these three are small, and this suggests that Deviller's method is practical since the covariance matrix can be analytically prepared. A slight difference is observed in the Cs-137 number density uncertainty between Deviller's method and GLS-1. This would be due to the β -delayed neutron emission of I-137. Note that this kind of comparison, in which slight differences should be detected, is possible with our sensitivity-based procedure.

[Figure 12 about here.]

Figure 13 shows fissile nuclide-wise contributions to the number densities uncertainties.

[Figure 13 about here.]

Table 1 Specifications of BWR single pincell models

Item	STEP-1	STEP-3
Pin pitch [cm]	1.63	1.44
Fuel pellet radius [cm]	0.529	0.490
Cladding outer radius [cm]	0.615	0.560
Cladding thickness [cm]	0.086	0.070
U-235 enrichment [wt%]	3.0	4.1

Table 2 Sensitivities of nuclides number densities to one-group (n,γ) cross section

Target	Nuclear data	Sensitivity
Ce-144	Ce-144 (n,γ)	-0.0004
Cs-134	Cs-133 (n,γ)	+0.8376
	Cs-134 (n,γ)	-0.0483
Cs-137	Cs-137 (n,γ)	-0.0005
Ru-106	Ru-106 (n,γ)	-0.0001
Sb-125	Sb-125 (n,γ)	-0.0080

Table 3 Relative standard deviations of cumulative fission yields in JENDL/FPY-2011 (unit: %)

	U-235-induced	Pu-239-induced
Ce-144	0.8	0.7
Cs-137	0.5	0.5
Ru-106	1.4	2.0
Sb-125	41.8	8.0

Figure Captions

Figure 1 Geometrical specification of the BWR assembly model

Figure 2 Sensitivity of nuclides generation to fissile nuclide-wise fission yields
(STEP-3, 0% void ratio)

Figure 3 Sensitivity of Eu-154 generation to FP nuclide-wise fission yields
(STEP-3, 0% void ratio)

Figure 4 Sensitivity of Eu-154 generation to FP one-group (n,γ) cross sections
(STEP-3, 0% void ratio)

Figure 5 Nuclides transmutation chain relevant to Eu-154 generation

Figure 6 Relative standard deviations of nuclides number densities during nuclear fuel depletion with several conditions

Figure 7 Component-wise relative standard deviations of nuclides number densities (STEP-3, 0% void ratio)

Figure 8 Relative standard deviations of Eu-154 number densities with and without the TENDL-2019 covariance data (STEP-3, 0% void ratio)

Figure 9 Nuclide-wise contribution in relative standard deviations of Eu-154 number densities induced by (n,γ) cross section uncertainties (STEP-3, 0% void ratio)

Figure 10 Correlation matrices of nuclides number densities among different con-

ditions and different burnups

Figure 11 Relative standard deviations of assembly-averaged nuclides number densities in the BWR fuel assembly

Figure 12 Relative standard deviations of nuclides number densities induced by fission yields data uncertainties

Figure 13 Fissile-wise contributions in relative standard deviations of nuclides number densities induced by fission yields data uncertainties

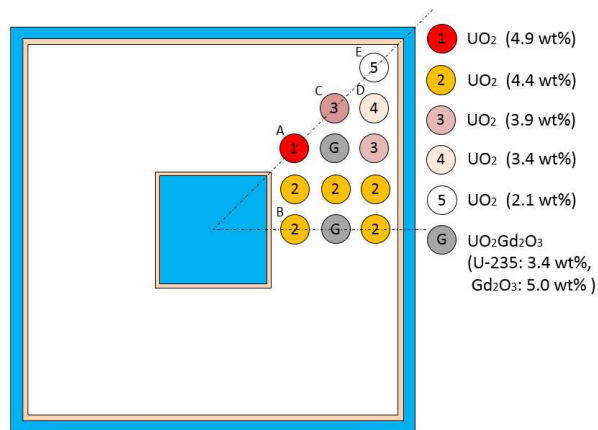


Figure 1 Geometrical specification of the BWR assembly model

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

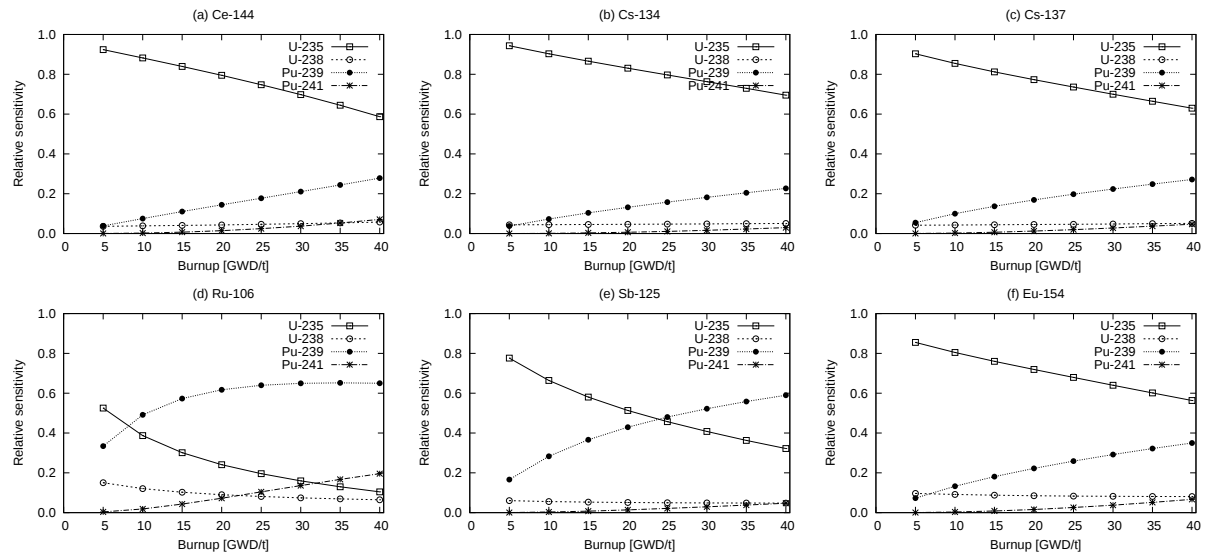


Figure 2 Sensitivity of nuclides generation to fissile nuclide-wise fission yields (STEP-3, 0% void ratio)

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

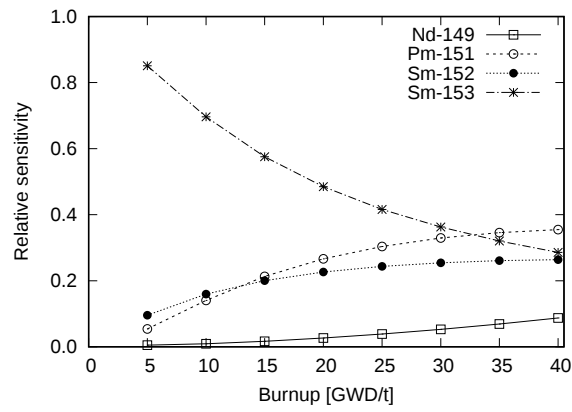


Figure 3 Sensitivity of Eu-154 generation to FP nuclide-wise fission yields (STEP-3, 0% void ratio)

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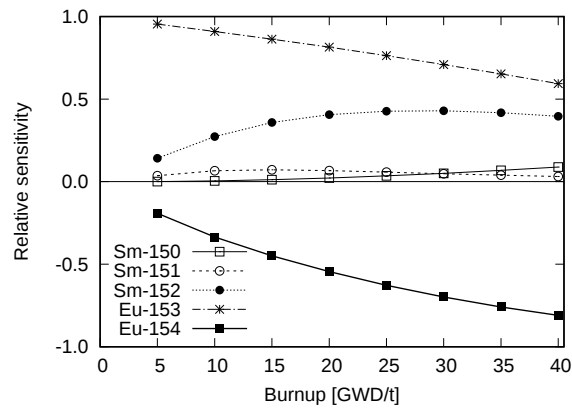


Figure 4 Sensitivity of Eu-154 generation to FP one-group (n,γ) cross sections (STEP-3, 0% void ratio)

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

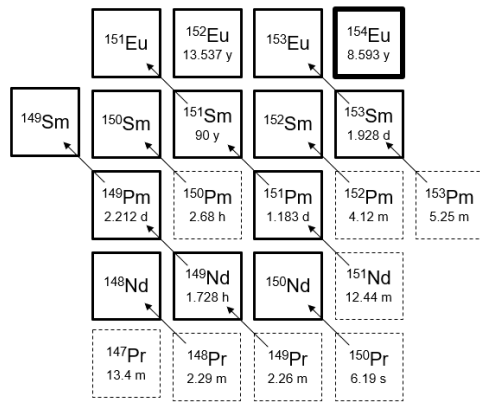


Figure 5 Nuclides transmutation chain relevant to Eu-154 generation

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

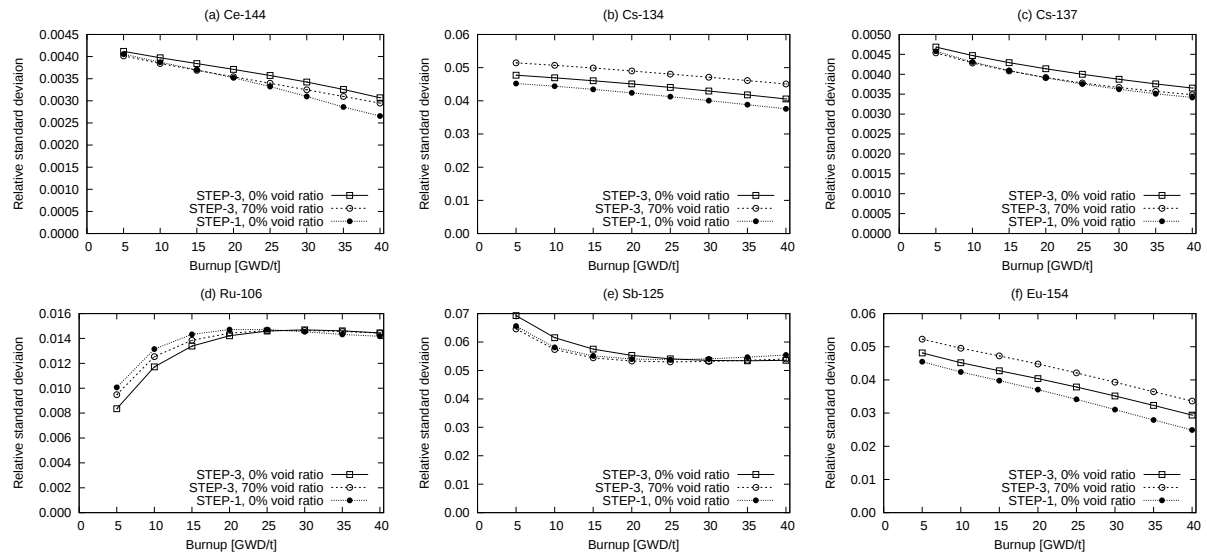


Figure 6 Relative standard deviations of nuclides number densities during nuclear fuel depletion with several conditions

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

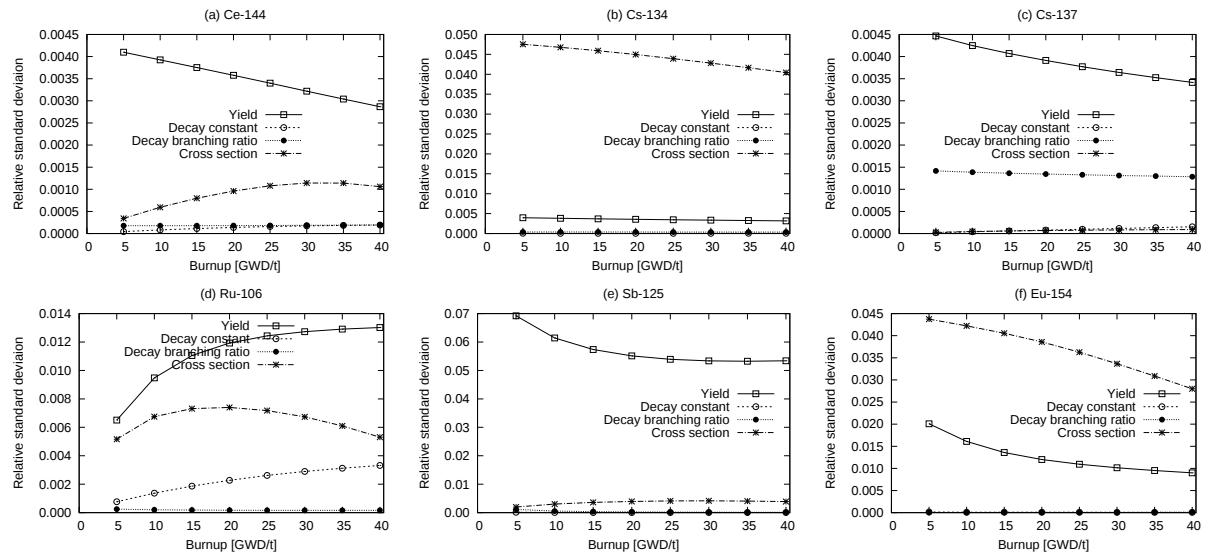


Figure 7 Component-wise relative standard deviations of nuclides number densities (STEP-3, 0% void ratio)

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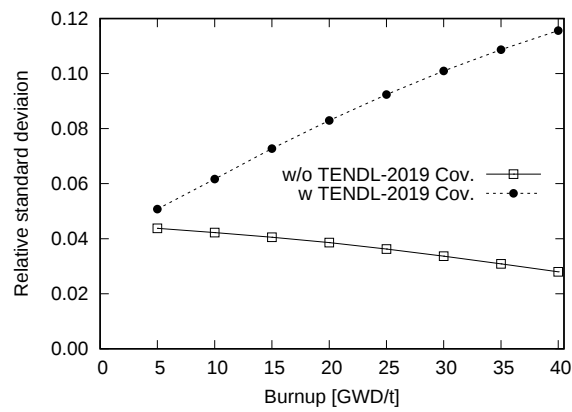


Figure 8 Relative standard deviations of Eu-154 number densities with and without the TENDL-2019 covariance data (STEP-3, 0% void ratio)

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

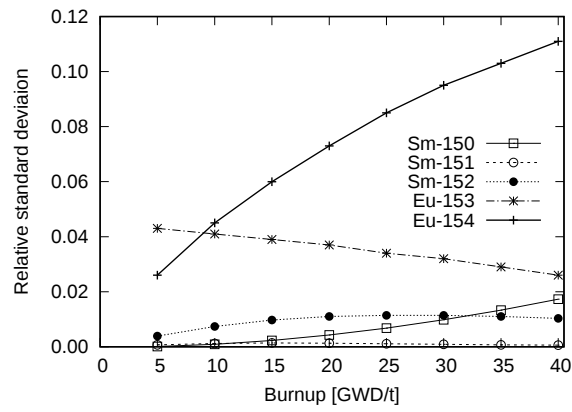


Figure 9 Nuclide-wise contribution in relative standard deviations of Eu-154 number densities induced by (n,γ) cross section uncertainties (STEP-3, 0% void ratio)

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

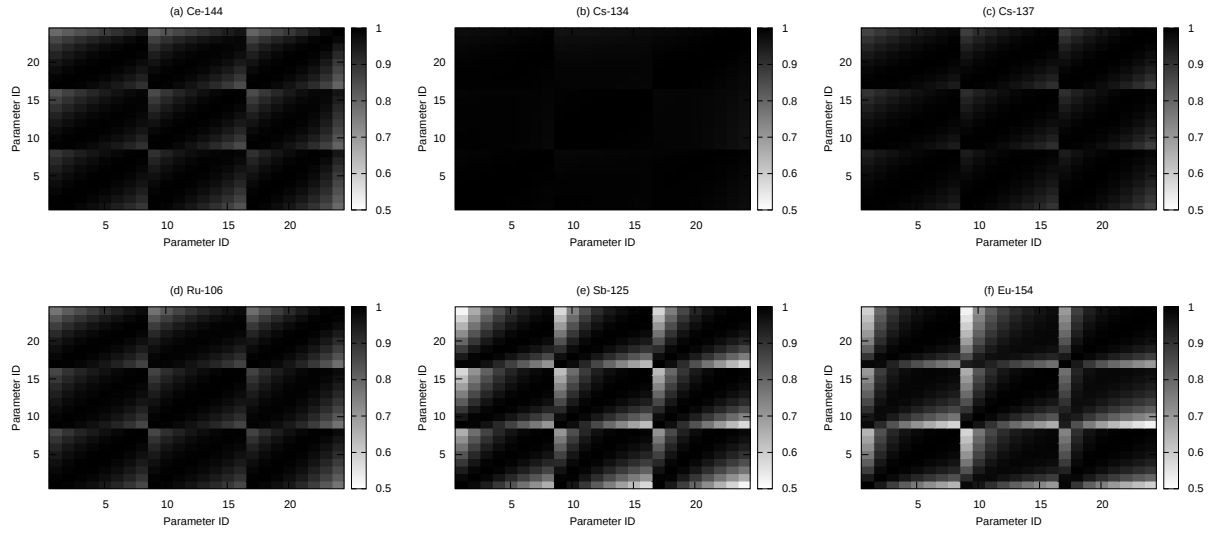


Figure 10 Correlation matrices of nuclides number densities among different conditions and different burnups

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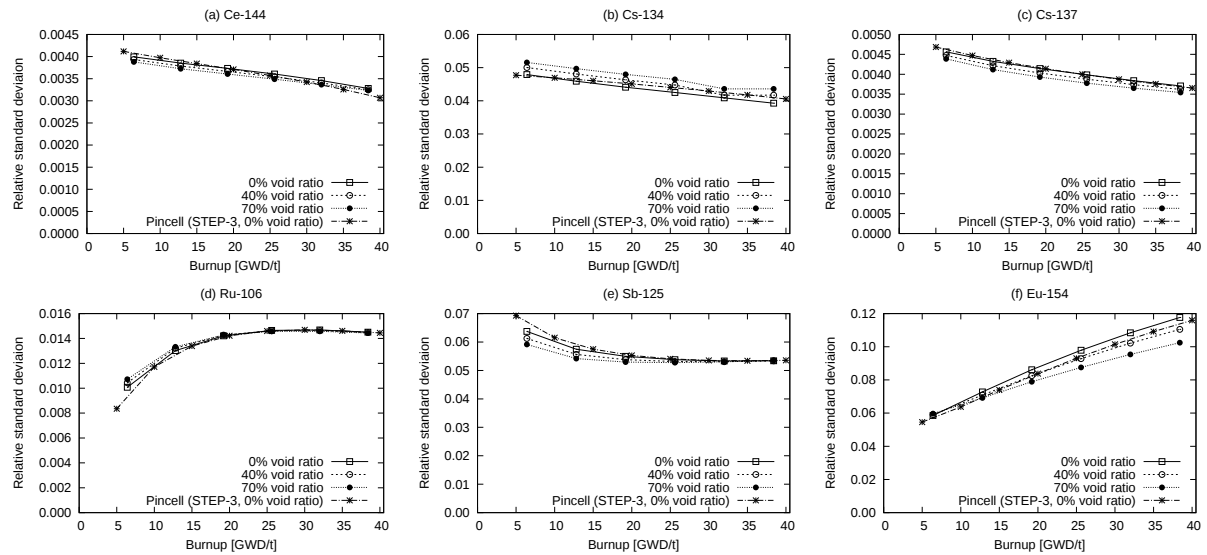


Figure 11 Relative standard deviations of assembly-averaged nuclides number densities in the BWR

fuel assembly

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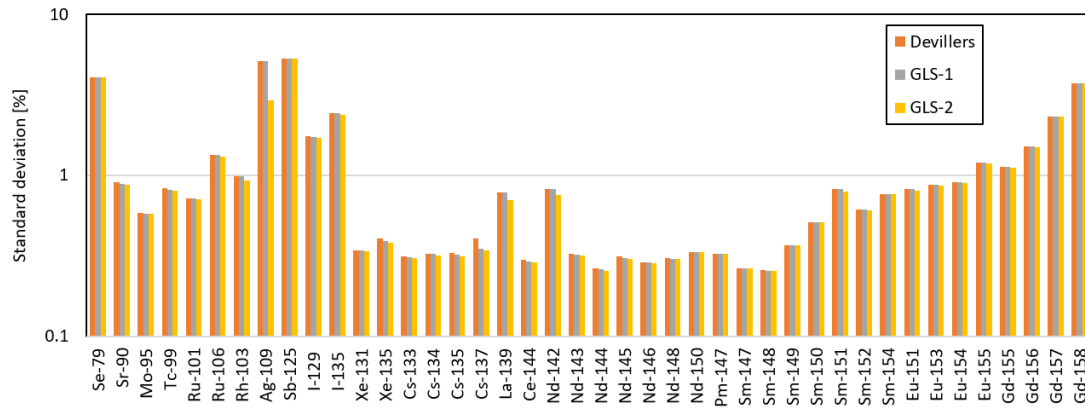


Figure 12 Relative standard deviations of nuclides number densities induced by fission yields data uncertainties

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy

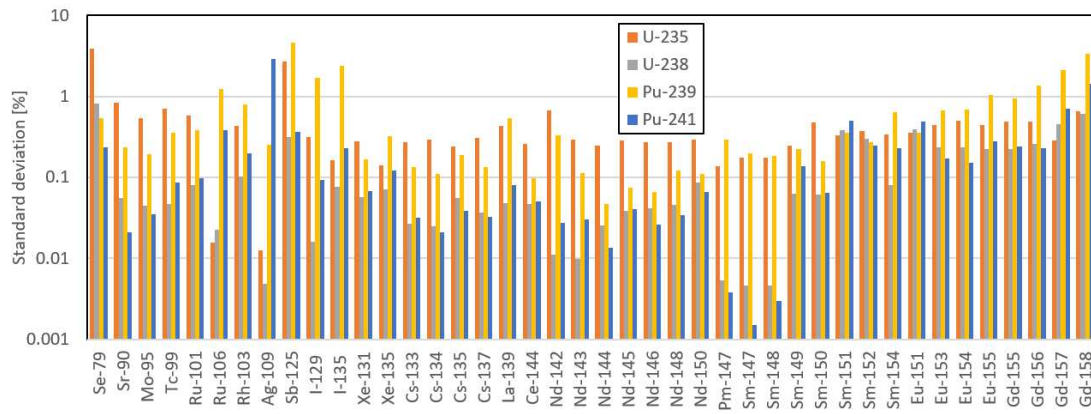


Figure 13 Fissile-wise contributions in relative standard deviations of nuclides number densities induced by fission yields data uncertainties

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Sensitivity and uncertainty analyses of fission product nuclides inventories for passive gamma spectroscopy