Assessment of neutronic parameter's uncertainties obtained within the reactor dosimetry framework: development and application of the stochastic methods of analysis

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Abstract

One of the main objectives of reactor dosimetry is the determination of the physical parameters characterizing the neutronic field in which the studied sample is irradiated. The knowledge of the associated uncertainties represents a significant stake for nuclear industry as shows the high uncertainty value of 15% (k=1) commonly allowed for the calculated neutron flux (E> 1MeV) on the vessel and internal structures. The study presented in this paper aims at determining then reducing uncertainties associated with the reactor dosimetry interpretation process. After a brief presentation of the interpretation process, input data uncertainties identification and quantification are performed in particular with regard to covariances. Then uncertainties propagation is carried out and analyzed by deterministic and stochastic methods on a representative case. Finally, a Monte Carlo sensitivity study based on Sobol indices is achieved on a case leading to derive the most penalizing input uncertainties. This paper concludes rising improvement axes to be studied for the input data knowledge. It highlights for example the need for having realistic variance-covariance matrices associated with input data (cross sections libraries, neutron computation code's outputs,…). Lastly, the methodology principle presented in this paper is enough general to be easily transposable for other measurements data interpretation processes.

KEYWORDS: Reactor dosimetry, uncertainty propagation, sensitivity analysis, Sobol method.

1. Introduction

One of the main objectives of reactor dosimetry is the determination of the physical parameters characterizing the neutronic field in which the studied sample is irradiated. These physical parameters, from neutron spectrum to reaction rates, are used in experimental reactor facilities to carry out the follow-up of the irradiation and to qualify the neutron calculation scheme. In industrial power reactors, they are also used for the follow-up of the neutron induced damages of vessel and internal structures.

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These values are derived from dosimeter's activities of which constitutive isotopes have adapted nuclear reactions (nuclear cross sections, g or X ray emissions,…). These activities are treated using nuclear data, neutron computation results from device modelling and irradiation conditions. This treatment constitutes a complex process because there are many entries more or less correlated and many stages inducing potential non-linearities (neutron spectrum unfolding). The assessment of the output uncertainties requires the implementation of rather complex methods.

However, the knowledge of these uncertainties represents a significant stake for nuclear industry as shows the high uncertainty value of 15% (k=1) commonly allowed for the calculated neutron flux (E> 1MeV) on the vessel and internal structures. Thus, the study presented in this paper aims at determining then reducing uncertainties associated with reactor dosimetry interpretation process.

After a brief presentation of the interpretation process, data uncertainties identification and quantification are performed in particular with regard to covariances. Then uncertainties propagation is carried out and analyzed by deterministic and stochastic methods on a representative case. Finally, a Monte Carlo sensitivity study based on Sobol indices is achieved on a case leading to derive the most penalizing input uncertainties. The paper concludes rising improvement axes to be studied for the input data knowledge.

2. Dosimetry process

2.1 Submission of Papers, Size of Pages

Dosimeters are selected with the sights of their response function on the studied neutron spectrum (nuclear cross section) and of their characteristics with respect to their activity's measurement (nature and energy of the emitted particles, radioactive half-life). Table 1 gives a typical list of used isotopes.

Table 1 : typical list of used isotopes

Isotope	^{197}Au	^{59}Co	^{55}Mn	^{115}In	^{237}Np	^{103}Rh	^{238}U
Reaction	n,γ	n,γ	n,γ	n,γ	n,p	n,n'	n,p
Threshold in MeV	Th	Th	Th-epi	Th-epi	0.6	0.7	1.0

Isotope	^{93}Nb	^{115}In	^{58}Ni	^{54}Fe	^{63}Cu	^{56}Fe	^{27}Al	^{24}Mg
Reaction	n,n'	n,n'	n,p	n,p	n,a	n,p	n,a	n,p
Threshold in MeV	1.2	1.3	2.7	2.8	6.1	6.1	7.3	6.7

These dosimeters are laid out within the irradiation device according to required information: gradient and level of neutron flux, neutron spectrum… With the end of the irradiation, dosimeters are sent to the LDCI laboratory, part of the Reactor Studies Department (DER) located at the CEA-CADARACHE centre (France).

The fast neutron flux (resp. fluence), $\phi(E>1MeV)$ given in n/cm²/s (resp. $\Phi(E>1MeV)$ given in n/cm²), is defined to be the flux (resp. fluence) of neutrons which energy is higher than 1MeV.

The fluence of each dosimeter is then evaluated according to a four stages process, using measured activities, calculated neutron spectra, dosimetric cross sections libraries [1] and irradiation conditions:

1) Activity Measurement and estimation of its uncertainty for each dosimeter
2) Determination of thermal and epithermal neutron fluxes using the ACTIGE neutron activation code ACTIGE [2] which treats the activation chain for the selected isotope.
3) For each dosimeters, determination of ϕ(E>1MeV) and Φ(E>1MeV) using ACTIGE code with respect to the corrections induced by the previously calculated thermal and epithermal fluxes.
4) Determination of the neutron spectrum and neutron fluence at the measurement point using STAY' SL code [3] which carries out the spectrum unfolding.

This stage uses in entry the reaction rates drawn from stage 3, a representative calculated neutron spectrum and multigroup nuclear cross sections drawn from the libraries [1], as well as associated covariances. STAY' SL Code provides a solution by a direct least squares method seeking to minimize the following $\chi 2$ function:

$$\chi^2 = {}^t\left(A^0 - A\right)\cdot\left(\Gamma_A + \Gamma_A 0\right)^{-1}\cdot\left(A^0 - A\right) \qquad (1)$$

Where

A^0 : matrix of reaction rates drawn from stage 3

A : matrix of calculated (adjusted) reaction rates

$\Gamma_A 0$: variance-covariance matrix of reaction rates drawn from stage 3

Γ_A : variance-covariance matrix of calculated reaction rates

3. Uncertainties determination

The uncertainties determination on data process generally includes 3 phases: input data uncertainties identification, uncertainties propagation to determine the output data uncertainties and finally sensitivity analysis. The latter makes it possible to determine most influential input data uncertainties on output data uncertainties. Thus, one identifies the input data on which uncertainty reduction is the most interesting.

3.1 Data uncertainty determination

An exhaustive data input analysis of each stage has allowed the characterization of the associated uncertainty by, a statistic distribution law Λ, a standard deviation Δ and a safety factor k. These parameters are necessary to any statistical study. When information is not available, laws are taken by default normal-type law except for the isotopic impurities abundance for which a uniform-type law is taken (no possible negative value).

The correlations are estimated from:

- provided variance-covariance matrices (e.g., neutron spectrum and nuclear cross sections)
- bayesian analysis of the physical phenomena: (e.g. correlations due to the calibration in energy of the activity measurement device)
- statistical analysis of the data when the population allows it (e.g. irradiation time histogram)

On the whole, an irradiation comprising ten dosimeters requires the analysis of more than 100 data inputs without taking into account the multigroup aspect of the neutron spectrum and cross sections; indeed, they comprise as much entry as energy groups. This important data set makes it possible to use the statistical methods described in the next paragraphs.

3.2 Uncertainties propagation

3.2.1 Variance composition formula

The standard uncertainty's propagation method rests on the variance's composition law which gives the standard deviation on the response $y = f(x_1, .., x_n)$ starting from uncertainties on the input variables $x_1, ..., x_n$.

If variables are statistically dependent, the variance's composition formula changes by integrating covariances [4]:

$$s[f(x_1,\cdots x_n)] = \left[\sum_i \left(\frac{\partial f}{\partial x_i}\right)^2 s^2(x_i) + 2\sum_{i=1}^{n}\sum_{j=i+1}^{n}\left(\frac{\partial f}{\partial x_i}\right)\left(\frac{\partial f}{\partial x_j}\right)\mathrm{cov}(x_i;x_j)\right]^{1/2} \quad (2)$$

Where s[…] denotes the standard deviation function.

This formula is proved to be correct if uncertainties on the x_i are "weak" (< 10%) and if the differential derivatives are not too high. This approach requires the knowledge of the literal analytical equation of the process in order to directly calculate their differential derivatives. The Latter could also be numerically calculated by finite differentiation methods but this would imply approximations and would have a high computation time cost.

In our process this method is only applied on activity measurement (stage 1). Indeed, a study realized on the other stages has shown that it is not possible to clarify the process literal equations needed for equation (2) application. This is mainly due to the treatment's non-linearity produced by evolution codes using iterative resolution methods.

3.2.2 Monte Carlo Methods

In order to circumvent the problems raised with paragraph 3.2.1 , a Monte Carlo simulation process has been implemented to evaluate output's uncertainties [5, 8].

For each input data, a sample of 1000 to 50000 simulations, according to the complexity of the treated case, has been randomly generated starting from the input data uncertainties laws stated on paragraph 3-a. The whole process (stages 2 to 4) has been applied to these input vectors to provide an equal number of output vectors containing the parameters of which one seeks uncertainty (fluence, flux, reaction rate,…). Then a statistical analysis of these output samples has been carried out:

- calculation of average values, standard deviations and other statistics,
- adjustment of a probabilistic distribution law,
- determination of the correlations between outputs.

This simulation method is applied on the determination of a fast neutron flux f(E>1MeV) using ACTIGE code and starting from the activity of an iron dosimeter. Table 2 illustrates the modelling of the input data uncertainties.

Table 2: Modelling of the input data uncertainties

	Activity	**Decay constant**	**Epithermic index**	**Ires ^{54}Fe**	**σ^{54}Fe (E>1Mev)**	**σth^{54}Fe (Thermal)**	**Φth**
Parameter	ACTM	DLMN54	RHO	SIFE54	SRFE54	STFE54	XFTH
Unity	Bq (x 10^{-3})	s^{-1} (x 10^{9})	No	barn	barn (x 10^{3})	barn	$n.cm^{-2}.s^{-1}$ (x 10^{-10})
Uncertainty modeling							
Type of law	Normal	Normal	x	Normal	Normal	Normal	x
Mean value	1	2,57	x	1,2	50	2,25	x
Standard deviation	2,55%	0,02%	x	16,68%	8,00%	8,00%	x
Generated sample							
Sample size	1000						
Mean value	0,9995	2,57	0,631	1,205	49,988	2,253	1,805
Standard deviation	2,63%	0,02%	11,21%	16,45%	8,28%	8,19%	3,74%

Table 3 gives uncertainty analysis results on $\phi_{(E>1MeV)}$ starting from the generated outputs samples.

Table 3: Uncertainties on $\phi_{(E>1MeV)}$ - Iron Dosimeter

Parameters	$\phi_{(E>1MeV)}$ $x10^{-10}$ $n.cm^{-2}.s^{-1}$
Mean value	3,820
Minimum	2,949
Maximum	5,246
Standard deviation	8,88%

3.2.3 Sensitivity analysis

From the output data population (3.2.2), the sensitivity analysis method allows the construction of a simplified linear model simulating the treatment process [5, 8]. The selected model explains at least 99,5% of the constructed sample's deviance and have good prediction capacities; with such a level of explanation threshold, the error induced by the model is considered to be negligible compared to the uncertainties sources.

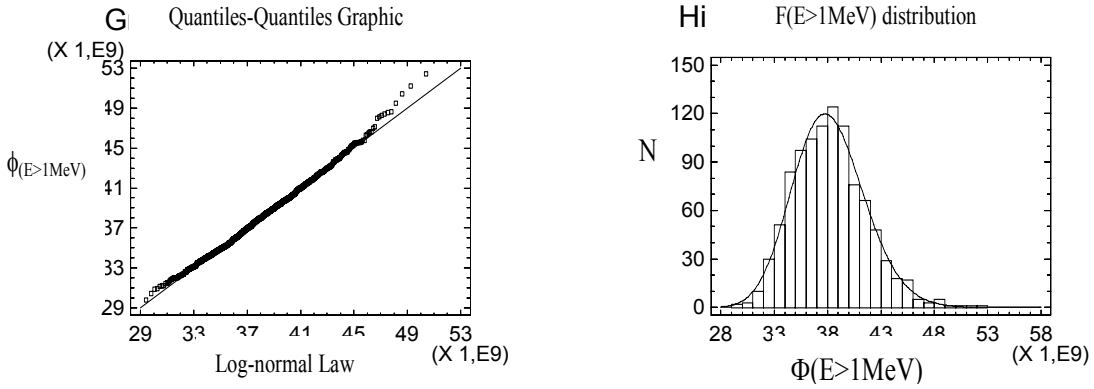
If the selected model does not contain interaction and no term above first order, the most influential parameters are classified after the estimate of the parameter's contribution for each one used in the built model. This contribution is defined by:

$$contribution(Y/X_i) = \hat{\beta}_i^2 \frac{\mathrm{var}(X_i)}{\mathrm{var}(\hat{Y})} \quad (3)$$

where var(X_i) is the variance of an input, var($\hat{Y}$) the variance of the estimated response using the simulated sample and $\hat{\beta}_i$ the coefficient of the built linear model.

In our case, various families of laws have been tested to adjust the $\phi_{(E>1MeV)}$ sample. This analysis shows that we can consider the random variable $\phi_{(E>1MeV)}$ distributed according to a lognormal law with at least 90% of confidence (Figure 1).

Figure 1: Distribution of uncertainty on $\phi_{(E>1MeV)}$ - Iron Dosimeter

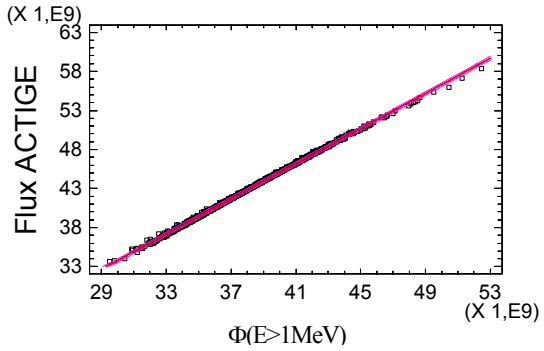


The simplest model explaining at least 99,5% of the sample's variance (99.91% in our case) with a good prediction capacity is the following model:

$$\Phi_{(E>1MeV)} = 10^{11} \times (3{,}8436 + 4{,}27141\ 10^{-4} \times ACTM - 116{,}083 \times SRFE54 + 779{,}85 \times SRFE54^2) \quad (4)$$

Figure 2 compares the 1000 couples ($\phi_{(E>1MeV)}$ estimated by ACTIGE; $\phi_{(E>1MeV)}$ estimated by the model). These points are well distributed on a line y = x which consolidates confidence in the established model.

Figure 2: Comparison of the computed and predicted values - Iron Dosimeter



The linear correlation coefficient between parameters SRFE54 and Φ(E>1MeV) derived from the sample analysis is worth -0,95; one thus has a very strong relation between these two variables. Because there is no term related to ACTM above the first order in the model, the contribution of the ACTM parameter is evaluated at 3 % (formula (3)). By complement with 1, the SRFE54 contribution is estimated at 97 %. Other inputs contributions are negligible. This is why uncertainty on Φ(E>1MeV) depends primarily on uncertainty on the 54Fe cross section.

This method is shown to be effective on a relatively linear treatment and without important correlation between input data. This characterizes stages 2 and 3 of our process.

It does not apply to stage 4 because of the use of a neutron spectrum unfolding method where correlations between input data become prevalent. Thus, we use a Sobol - Monte Carlo type methodology of analysis. Latter makes it possible to determine the contribution of input data uncertainties on the output data uncertainties without passing through a response model.

According to their respective distribution laws and correlations obtained in the paragraphs 3.1 and 3.2.2, one has randomly generated an input data matrix of the totality of the inputs and uncertain parameters used in the process of spectrum unfolding.

The Sobol indices theory is presented in reference [6]. Its application to the case of reactor dosimetry was carried out within the framework of the J. Jacques's thesis ([7],[8], [9]). One notes too the possible use of response models to calculate Sobol indices developed in [10].

The main idea of the Sobol approach for the sensitivity indices implementation is the decomposition of the function f(x) in sums of elementary functions:

$$f(x_1,\ldots,x_n) = f_0 + \sum_{i=1}^{n} f_i(x_i) + \sum_{i=1}^{n}\sum_{j=i+1}^{n} f_{ij}(x_i,x_j) + \ldots + f_{1,2,\ldots n}(x_1,\ldots,x_n) \quad (5)$$

such that the integral of all the sums are null and f_0 is a constant :

$$\int_0^1 f_{i_1,\ldots i_s}(x_{i_1},\ldots,x_{i_s})\,dx_{i_k} = 0 \quad if \quad 1 \le k \le s \quad (6)$$

The total variance D of $f(x)$ is defined by: $D = \int_{K^n} f^2(X)dX - f_0^2$ (7)

Since partial variances are implemented from (5):

$$D_{i_1,\ldots i_s} = \int_0^1 \ldots \int_0^1 f^2_{i_1,\ldots i_s}(x_1,\ldots,x_s)\,dx_1 \ldots dx_{ij} \quad \text{where } 1 \le i_1 \le \ldots \le i_s < n \text{ and } s=1,\ldots,n. \quad (8)$$

The sensitivity measurement is defined by: $S(i_1,\ldots,i_s) = \frac{D_{i_1,\ldots i_s}}{D}$ (9)

with the interesting property that the sum of all the sensitivity indices is worth 1.

Integrals (8) and (9) can be implemented as an MONTE-CARLO integral. Consequently, Monte Carlo estimates of f_0, D and D_i are given by the following formulas:

$$\hat{f}_0 = \frac{1}{N}\sum_{m=1}^{N} f(X_m) \quad (10)$$

$$\hat{D} = \frac{1}{N}\sum_{m=1}^{n} f^2(X_m) - \hat{f}_0^2 \quad (11)$$

$$\hat{D}_i = \frac{1}{N}\sum_{m=1}^{n} f\left(X^{(1)}_{\sim im}, x^{(1)}_{im}\right) f\left(X^{(2)}_{\sim im}, x^{(1)}_{im}\right) - \hat{f}_0^2 \quad (12)$$

where :
- N is the number of generated samples needed to obtain Monte Carlo estimates,
- X_m is a point resulting from sampling in K^n
- $X_{\sim im}=(x_{1m},\ldots,x_{(i-1)m},x_{(i+1)m},\ldots,x_{nm})$
- $^{(1)}$ and $^{(2)}$ mean that one treats 2 matrices of size [N x n] and independently randomly generated for X.

The total sensitivity index is given by: **ST(i)=S(i)+S(i,~i)=1-S(~i)** (13)

with S(~i) sum of the $S(i_1,\ldots,i_s)$ which does not include i index. In other words it is the total fractional variance complementary to the xi parameter.

The necessary integral to implement S(~i) is estimated by Monte Carlo methods :

$$\hat{D}_{\sim i} + \hat{f}_0^2 = \frac{1}{N}\sum_{m=1}^{n} f\left(X_{\sim im}^{(1)}, x_{im}^{(1)}\right) f\left(X_{\sim im}^{(1)}, x_{im}^{(2)}\right) \tag{14}$$

Thus the total contribution estimate of parameter i on output's total variation is given by:

$$\hat{ST}(i) = 1 - \frac{\hat{D}_{\sim i}}{D} \tag{15}$$

Practically the treatment is as follows:

1 One carries out the simulation of 2 samples of the same size N on the n independent entries. They are placed in the D1 and D2 matrices of size n x N.
2 One extracts the M1k and M2k matrices (dimensions k<N) from D1 and D2
3 Matrix M1k is processed giving an output matrix R1k
4 Column i of M1k matrix, $1 \le i \le n$, is then replaced by column i of the M2k matrix, giving a M1ki matrix
5 The matrix M1ki is processed giving an output matrix R1ki
6 One reiterates n times operations 4) and 5), i going from 1 to n.
7 One treats n+1 outputs matrices according to the equation (12) in order to determine the first order Sobol indices
8 Column i of M2k matrix, $1 \le i \le n$, is then replaced by column i of the M1k matrix, giving a M2ki matrix
9 The matrix M2ki is processed giving an output matrix R2ki
10 One reiterates n times operations 8) and 9), i going from 1 to n.
11 One treats n+1 outputs matrices according to the equation (15) in order to determine the total Sobol indices
12 One reiterates stages 2 to 7 until the convergence is reached for all the total Sobol indices.

This method has three notable characteristics which simplify its use:

- Input data must be statistically independent, if not, it is possible to gather them in a single input vector considered as macro-entry. In this case it will only be possible to deduce the sensitivity on this macro-data ([7],[8]).
- The results on the indices are additive allowing an incremental convergence if the size of data sample's matrix is sufficient.
- When a Sobol index is converged, i.e. its variation between two steps is lower than a chosen ε (1% for example), the process can continue without considering it: no column replacement and associated treatment for the associated input. This increases the convergence process speed after each converged index.

Specific MATLAB® software has been developed to apply this Sobol methodology to the following example.

Input data are the calculated neutron spectrum, the reaction rates of coupled dosimeters D1-D2 and D3-D4 (correlated by activity measurements) and the cross sections (E>1MeV) of the 4 used dosimeters. A set of 932500 simulations of input vectors has been built for the 7 statistically independent macro-entries; correlations between the energy groups of the spectrum being important, only the spectrum in entirety is regarded as a variable. There are actually several hundreds of data if one takes into account the multi-groups cross sections given in a 640 energy groups formalism. It should be noted that the internal macro-entries covariances were taken into account for random simulations. Table 4 (next page) synthesizes a part of the results.

Table 4: Results of Sobol treatment (total indices)

	932500 simulations				
Outputs	***Adjusted Reaction Rate***				***Flux (E>1MeV)***
Inputs	***D1***	***D2***	***D3***	***D4***	
Cross Section D1	**0,96**	0,01	0,00	0,00	**0,20**
Cross Section D2	0,00	**0,50**	0,00	0,00	0,00
Cross Section D3	0,00	0,03	**0,69**	0,01	**0,06**
Cross Section D4	0,00	0,02	0,02	**0,90**	**0,29**
30 Flux Group	**0,03**	**0,31**	**0,21**	**0,06**	**0,18**
Reaction rate D1-D2	0,00	0,04	0,02	0,01	**0,06**
Reaction rate D3-D4	0,00	0,03	0,04	0,01	**0,12**
Total	**1,00**	**0,93**	**0,99**	**0,98**	**0,92**

The convergence of the Sobol indices is correct on the whole of the data. A continuation of simulations would make it possible to improve Φ(E>1MeV) convergence but would not change the qualitative results analysis:

1) For each output, the sum of all the total indices is almost one, showing there is no significant non linear interactions (above the first order).
2) Dominating sources of uncertainties are in the decreasing order
   - Uncertainty on the D4 cross sections,
   - Uncertainty on the D1 cross sections,
   - Uncertainty on the neutron spectrum,
   - Uncertainty on the reaction rate D3-D4.

The analysis of the adjusted reaction rates shows two different behaviors according to the relative importances from uncertainties on the neutron spectrum and on the nuclear cross sections. It highlights the importance of a correct and physically realistic determination of the variance-covariance matrices associated with the input data.

3. Conclusion

Methods of uncertainties and sensitivity analysis applied at each stage of the neutron fluence evaluation process have been described in this paper in terms of advantages and limitations. Thus, one can set up an output data uncertainties evaluation method with a high confidence degree and which is easily applicable for each treated case.

The carried out study has also made it possible to undertake an in-depth analysis of the process which led to forecast some modifications in order to make the process more physically coherent: seek of a relevant comparison point for calculation/measurement, uncertainty decrease by suppressing non-physical stages which generate extra uncertainties.

It highlights moreover the need for having realistic variance-covariance matrices associated with input data. An important effort still remains to be realized on this variance-covariance matrices determination as well on the cross sections libraries as on the neutron computation codes.

At a methodological level, developments continue to allow the use of statistically correlated entries in a Sobol type treatment [9].

Lastly, the methodology principle presented in this paper is enough general to be easily transposable for other measurements data interpretation processes.

4. Acknowledgements

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