USE OF INTEGRAL SLAB EXPERIMENTS TO PROCESS NUCLEAR DATA ADJUSTMENTS : STUDY OF HAFNIUM NEUTRON CROSS-SECTIONS THROUGH DETERMINISTIC AND STOCHASTIC COMPUTATIONS

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ABSTRACT

The essential role of integral informations to validate computer codes data is well known by reactor physicists ; these experiments allowed in particular an extensive analysis of the major fuel nuclides cross-sections (see reference [1] for example). However, the recent progess of the methods (together with the performances of computers) gives the opportunity to exhibit from integral measurements (material buckling, critical masses, reactivity effects, spectrum indices) not only informations (variations) in terms of multigroup cross-sections but also in terms of the basic nuclear data (the resonance parameters).

In the present work integral slab experiments are exploited with the goal of reducing discrepancies between experimental results and calculations, and bring back new informations on particular nuclear data such as hafnium parameters. To solve the inverse problem, a statistical approach, based on the generalized least-squares method and perturbation theory, has been incorporated into our calculation system in order to deduce microscopic cross-section adjustments from observed integral measurements on this specific « mock-up » reactor.

This study established that the correlations beetween integral parameters and hafnium capture cross-sections enable specific variations in the resolved resonance range at the level of multigroup and punctual cross-sections recommended data (JEF-2.2 evaluation) to be highlighted. The use of deterministic methods together with Monte Carlo- type simulations enabled a depth analysis of the modelling approximations to be carried out. Furthermore, the sensitivity coefficient validation technique employed leads to a reliable assessment of the quality of the new basic nuclear data. In this instance, the adjustments proposed for certain isotope ^{177}Hf resonance parameters reduce, after error propagation, by 3 to 5 per cent the difference between experimental results (reactivity worths) and calculations. Beyond this particular application, the generalized qualification methodology integrated in our calculation system should enable other basic sizing parameters to be treated (chemical/geometric data or other unexplored nuclear data) to make technological requirements less stringent.

1. INTRODUCTION

Faced to major challenges in the field of nuclear power reactors (safety and innovative concepts implying more complex systems and guarantees of accuracy in the computations), the models employed to reproduce physical neutronic phenomena has been extensively improved during this last twenty years. In the same time, neutron cross-sections evaluations and the associated data included in the lattice codes are based on more and more sophisticated nuclear theories and technical experiments to cover a large range of reactor systems (fast to thermal neutron reactor).

Nevertheless, particular nuclear data are still not enough accurate and require the use of both differential and integral experiments (involving dedicated particles accelerators and in core reactor measurements) to improve their knowledge and reduce their corresponding uncertainties.

In this paper a statistical approach based on the least-squares method is proposed in order to prevent the effect of modelling error on the adjustment results. The new methodology is then applied to the case of integral slab experiment to study the current hafnium nuclear data coming from the european JEF-2.2 evaluation file.

2. ADJUSTMENT METHODOLOGY

In the usual statistical approaches applied to process nuclear data adjustments (mean-squares or equivalent bayesian methods) one of the more drastic hypothesis is the assumption of unbiaised models. This comes initially from the first order expansion of the integral data y_i considered as a functional of the nuclear parameters x_j :

$$y_i = f_i(x_1, x_2, ..., x_J) = f_i(x_1^0, x_2^0, ..., x_J^0) + \sum_{j=1}^{J} \left(\frac{\partial f_i}{\partial x_j} \right)_{x_j = x_j^0} .(x_j - x_j^0) + o\left((x_j - x_j^0)^2\right) \qquad (1)$$

where the modelization errors are supposed sufficiently weak in front of nuclear data errors to be neglected ($f_i(x_1^0, x_2^0, ..., x_J^0)$ is the integral parameter y_i^0 computed with the initial set $(x_1^0, x_2^0, ..., x_J^0)$).

By defining the relative variations :

$$\begin{aligned} \mathrm{x_j} &= (x_j - x_j^0)/x_j^0 \qquad (i = 1,2,...,I) \\ \mathrm{y_i} &= (y_i - y_i^0)/y_i^0 \qquad (j = 1,2,...,J) \end{aligned} \qquad (2)$$

and the correponding vectors :

$$\mathbf{z} = \begin{bmatrix} \mathbf{x} \\ \mathbf{y} \end{bmatrix} \text{ with } \mathrm{z_k = x_j} \ \ (\mathrm{k = 1,...J}) \ ; \ \mathrm{z_k = y_i} \ \ (\mathrm{k = J+1,..., J+I = K}) \qquad (3)$$

The relationship expressed by the equation (1) becomes :

$$\mathbf{Sz} = 0 \qquad (4)$$

with the sensitivity matrix coefficients :

$$s_{kl} = -\delta_{kl} \ \ (k = l) \ ; \ \ s_{kl} = \frac{x_l^0}{y_k^0} \left(\frac{\partial f_k}{\partial x_l} \right)_{x_l = x_l^0} \qquad \forall (k = J+1,...,K) \neq (l = J+1,...,K) \qquad (5)$$

The integral and differential experimental information can also being ranged in the vector :

$$\mathbf{z}^{\mathbf{exp}} = \begin{bmatrix} \mathbf{x}^{\mathbf{exp}} \\ \mathbf{y}^{\mathbf{exp}} \end{bmatrix} \tag{6}$$

using the same notations :

$$\begin{aligned} \mathrm{x}_{\mathrm{j}}^{\mathrm{exp}} &= \left(x_j^{\mathrm{exp}} - x_j^0\right) \Big/ x_j^0 \quad (i = 1,2,...,I) \\ \mathrm{y}_{\mathrm{i}}^{\mathrm{exp}} &= \left(y_i^{\mathrm{exp}} - y_i^0\right) \Big/ y_i^0 \quad (j = 1,2,...,J) \end{aligned} \tag{7}$$

It is then possible to find the inverse of system (1) by applying the basic statistical principles of the mean-squares method.

We first assume that $\mathbf{x}^{\mathbf{exp}}$ and $\mathbf{y}^{\mathbf{exp}}$ are random vectors normally distributed with the known covariance matrices :

$$\mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right) = \begin{bmatrix} \mathbf{D}\left(\mathbf{x}^{\mathbf{exp}}\right) & \\ & \mathbf{D}\left(\mathbf{y}^{\mathbf{exp}}\right) \end{bmatrix} \tag{8}$$

Therefore, the likelihood function is given by the expression of the gaussian distribution :

$$L\left(\mathbf{z}, \mathbf{z}^{\mathbf{exp}}\right) = (2\pi)^{-K/2} \left[\det \mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right)\right]^{-1/2} \exp\left[-\frac{1}{2}\left(\mathbf{z}^{\mathbf{exp}} - \mathbf{z}\right)^t \mathbf{D}^{-1}\left(\mathbf{z}^{\mathbf{exp}}\right)\left(\mathbf{z}^{\mathbf{exp}} - \mathbf{z}\right)\right] \tag{9}$$

and the best estimate of y results to be that vector for which this function take the maximum value (following the maximum likelihood principle).

This problem is now equivalent to the minimum condition :

$$\left(\mathbf{z}^{\mathbf{exp}} - \mathbf{z}\right)^t \mathbf{D}^{-1}\left(\mathbf{z}^{\mathbf{exp}}\right)\left(\mathbf{z}^{\mathbf{exp}} - \mathbf{z}\right) = \min \tag{10}$$

with the constraints (4).

As described in the reference [2-3], the application of the Lagrange's multipliers method implemented in the so called AMERE computer code, lead to the following expression of the a posteriori estimate :

$$\mathbf{z}^* = \left[\mathbf{1} - \mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right)\mathbf{S}^t \left[\mathbf{S}\mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right)\mathbf{S}^t\right]^{-1} \mathbf{S}\right] \mathbf{z}^{\mathbf{exp}} \tag{11}$$

and the corresponding covariance matrix :

$$\mathbf{D}\left(\mathbf{z}^*\right) = \mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right) - \mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right)\mathbf{S}^t \left[\mathbf{S}\mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right)\mathbf{S}^t\right]^{-1} \mathbf{S}\mathbf{D}\left(\mathbf{z}^{\mathbf{exp}}\right) \tag{12}$$

It can also be shown that the quantity :

$$\mathbf{z}^{*t}\ \mathbf{D}^{-1}\left(\mathbf{z}^*\right)\mathbf{z}^*$$

is a random variable distributed following a khi-square law.

In particular, for the J integral parameters, it is expected that χ^2_J is included in the theoritical margins :

$$1-\sqrt{2/J} \le \chi^2_J \le 1-\sqrt{2/J} \qquad (13)$$

This provides a convenient way to verify the gaussian assumption for the probability density of the integral and the microscopic parameter. Nevertheless, the khi-square test is not sufficient to detect the remaining systematic errors due to numerical approximations in the physical models.

Furthermore, the derived expression (11) proceed generally of an ill-posed and underdetermined problem because the number of integral measurements is too small with regard to the number of nuclear data involved. This means that the existence, the uniqueness and the continuity of the inverse problem are not guaranteed.

In fact, the use of a large set of integral expermental data (hundreds for example) in order to regularize the equation system (10) requires in the same time an accurate computation of the corresponding parameters, in order to respect our very first hypothesis.Unfortunately, the common practice in the resolution of the adjustment problems is to compute the overall integral parameters originated from various experiments (generally caracterized by different type of geometry, fuel components and spectra) using the same calculational scheme.

This is the starting point of our approach. To overcome these difficulties, we have first selected a reduced and synthetic set of integral and microscopic (the choose of absorber efficiencies as integral parameters allow us to focus on hafnium nuclear data). Then , by applying the mean-squares method on the basis of more and more refined models one can obtain reliable adjustments and highlight simultaniously the ability of each of them to restitute the correct variations.

If $\mathbf{y}_1$ denotes, for example, the reference calculation and $\mathbf{y}_2$, $\mathbf{y}_3$ the standard schemes (namely the punctual or the multigroup Monte-Carlo method and the discrete ordinates method implemented in the TRIPOLI4 (reference [4]), TRIMARAN2 (reference [5]) and APOLLO2 (reference [6]) neutron transport codes respectively) with the relative notation (2) one can easily quantify the effect of numerical approximation on the adjustment's results by making the simple difference between the a posteriori estimates $\mathbf{z}_1^*$ and $\mathbf{z}_2^*$ or $\mathbf{z}_3^*$.

Obviously this approach is only consistent if each computation is performed using the same set of nuclear data ($\mathbf{x} = \mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}_3$). In our particular case, both APOLLO2 and TRIPOLI4 codes uses librairies based the European JEF2.2 nuclear data file under investigation.

Important remark : The method of maximum likelihood, the Bayes scheme and the generalized least-squares method are the possible statistical approaches for performing nuclear data adjustment using integral measurements. They generally lead to similar expression of the a posteriori estimates and the related covariance matrices. In literrature these methods are also encountered (and should be more rigourously classified) as parts of the stastistical approaches employed to solve ill-posed inverse problems.
S. Tarantola, especially, has largely contributed to the improvement of statistical inversion in many applications (the regularisation of a discrete ill-posed problem can also be performed by adding deterministic criteria the search of quasisolutions as stated in the Tikhonov's reference [7]). In particular, he showed (reference [8]) by extension of the Bayes rules that the accuracy degree of the physical models could also be caracterized by a probability density straighforwardly introduced in the likelihood function. For example if the modelling errors are randomly distributed following the gaussian law, the likeliwood function may formally take the identical form (9) with an additional matrix $\mathbf{D}(\mathbf{z})$.

Because modelling and measurement uncertainties may be statistically summed, the same minimization routine is still operating except that the assumption of quasi-exact models is not remaining. For the future application we have planned to develop this generalized approach and valorized in the same time the numerical validation results ; in particular, when computations are performed using the Monte-Carlo method the error matrix is intrinsically gaussian and can be consequently added to the experimental dispersion the covariance matrix as it is done in section 5.

3. INTEGRAL AND NUCLEAR DATA ANALYSIS

After the integral experiment representative of the dedicated naval reactors were compiled and characterized during a first phase, we directed our selection towards a series of critical slab experiments in monodimensional geometry (i.e. which could be represented with a single dimension). Furthermore, the study of configurations with hafnium absorber seemed of the prime necessity due to the preponderant role of this material to control the core reactivity. In order to comply with our initial criterions we opted for the series of experiments implemented within the AZUR-Caramel program. In those experiments realized in the 70s-80s, a major part is dedicated to the validation of the calculation of the hafnium efficiency. Moreover, the reference configuration is formed by a lattice of parallel plates which can easily be simulated (because the fundamental mode is well established, the numerical approximation in the lattice codes lead to satisfactory results).

The AZUR-Caramel experiments, in particular, are part of the «tools» thanks to which the assembly calculations could have been validated. Nevertheless, we must note that these experiments were not initially intended for the validation of the basic data. As we can see on the figure 1, the reference core is formed of 35 parallel rows of 5 plates placed end to end. Each fuel plate is formed by 4 rows of 26 small UO2 plates which are inserted in a zircaloy alloy grid. Thus formed the lattice is made of a radial water moderator , i.e. a fuel and a clad which are repeated along the x-axis according to a constant pace. It is possible to study this lattice thanks to a mere «cell» calculation or a «multi-cell» calculation in the presence of absorber. In the course of this experiment two types of measurements were performed, the power distribution and the reactivity effects measurements. The main purpose was to define the integral efficiency of Hafnium slabs which composition is similar to those used for the real control cluster. The study of this absorber was conducted according to two parts. The first part was dealing with the substitution of the central row of fuel plates by a 5mm slab placed at successive heights in the assembly. The purpose of the second part was to measure the reactivity effect generated by the insert of slabs of various thickness, from 0.7mm to 5mm, which were totally inserted in the core.

As we shall see further on, this implies a specific treatment as far as the hafnium efficiency is concerned. In order to obtain the slabs efficiency, a equivalence method with the soluble boron has been used. The interesting aspect of this method is that it allows to study all sorts of configurations by adjusting each configuration to the critical value of the boron title (which is considered as a standard during calculations). The reactivity variations due to the slabs insert is therefore obtained by integrating the differential efficiency of the boron between two critical states. The purpose of this calculation is to reproduce at best the boron/hafnium equivalence. In the instance of the 5mm slab, we will have to ensure, for example, that the hafnium/boron calculations discrepancies does not exceed the 4.43% 3 sigma uncertainty attributed by experimentalists, for a global reactivity variation of -7010pcm (pcm = 10^{-5}).

Beside the uncertainty analysis of integral parameter, the assignment of standard deviation and correlation coefficients to each neutron cross-section is needed in order to obtain the full covariance matrix.

The aim of the present study was to complete an already known error matrix (determined from the previous analyses, references [9-10], in a 15 macrogroup coarse energy mesh) by variances and covariances related to hafnium cross-sections.

For hafnium isotopes, a generalized treatement of nuclear data, based on the adjustment of experimental resonance parameter with respect to sophisticated nuclear models such as the Reich-Moore formalism is very untricated. In fact, we have first noticed after a compilation of the earlier measurements reports that informations on these data were uncomplete (the uncertainted analysis generally did not appear clearly) and divided into small portions of the energy ranges. In addition, any uncertainty data related to this element were included in the international evaluation file, that is the « covariance files » 30 to 33 in the standard ENDF/B format (in the JEF-2.2 File only 15 covariance file are presently stored).

To overcome this problem, the most convenient way was to assign uncertainties to the average hafnium cross-sections (capture cross-sections especially) by the staighforward comparison of international evaluations, nuclear models and measurement reports. The table 1 presents, the capture cross-sections at thermal energy and the resonance integrals derived from the present european (JEF-2.2), american (ENDF/BVI) and japonese (JENDL-3.2) datafile.

We shall remark that the different values of σ_γ^0 and I_γ are very close (and restrained in for the major 177, 178 and 179 isotopes in the 1 sigma uncertainty margin). In fact, those evaluations are based not only on the same Breit-Wigner multi-level model (in the resolved energy range) but also on the same set of differiential measurements (compiled in the 73's and 84's BNL, see references [11-12]). Thus, the deriving punctual cross-sections are almost identical. This implies in particular that the induced information brought back by the integral quantities in the adjustement issue could impact the overall evaluation.

Secondly, a deepened uncertainty analyze involving the basic nuclear data, the resonance parameters, is required. In order to deduce the uncertainty values in the 15 macrogroup enregy mesh we have to calculate the sensitivity coefficients relying the averaged neutron cross-sections to the resonance parameters, namely the energy, the neutron and the radiative width in the resolved resonance range. We first dealt with the major resonances, contained in the 1eV-20eV energy intervall for 177, 178 and 179 isotopes. Within this area, we can easily verify that the resonance width $\Gamma_{\alpha,\lambda}$ and the level spacing $D_{\lambda,\lambda'}$ are following the inequality :

$$\sqrt{\Gamma_{\alpha,\lambda}\Gamma_{\alpha,\lambda'}} \Big/ D_{\lambda,\lambda'} \leq 0{,}001 \tag{14}$$

for any resonance with spins J=3,4 or 5.

This inequality involves that overlapping between open channels of the compound nucleus is weak and the different resonances well separated. Although sensitivity coefficients were also computed using direct calculations (using the standard NJOY scheme, reference [13]), we can easily adopt the Breit-Wigner single level formalism and derive these coefficients in the resonance neighborhood, as :

$$s^j_{\bar{\sigma}_\gamma,\Gamma_n} \cong 1 - \frac{\Gamma_{nj}}{\Gamma_j} \text{ and } s^j_{\bar{\sigma}_\gamma,\Gamma_\gamma} \cong 1 - \frac{\Gamma_{\gamma j}}{\Gamma_j} \tag{15}$$

regarding the neutron and the radiative width.

The table 2 gives the example of the derived sensitivity coefficients for the ^{177}Hf isotope together with the measured nuclear parameters (references [14-16]) and the final uncertainties.

We can notize, in particular, that the average capture cross-sections are almost totally correlated with the neutron widths and the given 1σ standard deviation rises 20% for the 6.57 eV resonance.

To determine the uncertainties in other energy ranges we refered to the experimental values and the corresponding covariances related to the measurements of the punctual cross-sections (see references [17-18]). Beside the thermal and resolved resonance energy range we broadly used the error analysis performed by Kompe, reference [17], in the keV and the MeV region. Note finally that in addition to the diagonal components of the dispersion matrix (i.e. variances) we straighforwardly defined the non-diagonal covariances assuming that the dependant energy variances are weakly correlated (the study of the effect of the choose of correlation is not reported in this paper but it doesn't give rise to significatives differences in the adjustment results).

4. COMPUTATIONS AND SENSITIVITY STUDY (NON-LINEAR EFFECTS VALIDATION)

After the uncertainties and the main quantities (integral parameter and nuclear data) have been deduced from experiments, we must quantify the modelisation biaises. In particular, we must engage in a very accurate reference calculation in order to break ourselves of the hypothesis formulated at the time of the implementation of the deterministic methods.

In order to achieve these reference calculations, we used Monte-Carlo type simulations which allow to reach a precision level inferior to the experimental standard deviations. The main differences between the used codes, i.e. TRIMARAN 2 and TRIPOLI 4 relates to the treatment of the self-shielded cross-sections. Compared to TRIMARAN 2, the energy treatment which is achieved with TRIPOLI 4 is much more accurate insofar as the self-shielded cross-sections are not only averaged (over the172 groups energy mesh) but continuously described (with more than thousand dots). In both cases, the used self-shielded cross-sections sets are derived from the European evaluation JEF-2.2. The entry data are therefore totally coherent and allow a more precise comparison of deterministic and stochastic methods.

We have gathered, in the table 3, the experiment/calculation discrepancies resulting from the deterministic and stochastic simulations. The APOLLO2 calculations were achieved thanks to the discrete ordinates method which enables to solve the integro-differiential transport equation (see for example reference [19]). The calculation options were optimized in order to obtain well convergent eigenvalues. We studied, in particular, the anisotropy effects at the level of the scattering cross-sections and the flux, the self-shielding phenomenon and the leakage models' influence. Moreover, the global uncertainty relating to the TRIPOLI4 and TRIMARAN2 discrepancies results from the quadratic combination of the experimental accuracy to the standard discrepancies specific to the Monte Carlo method.

The table shows that the hafnium slabs efficiency is systematically over-estimated regardless of the used calculations. For a same cross-sections set (punctual or multi-group sections derived from the JEF-2.2 evaluation), we can observe very important experiment/calculation discrepancies which vary from 4% to 8% (depending on the slab's thickness) and often largely exceed the 1 sigma uncertainty. Consequently, it is now essential to tackle the problem relating to the qualification of nuclear data in order to take into account uncertainties linked to these parameters.

The first step of our work is to analyze the sensitivity of the values integral to the self-shielded cross-sections which appear in Boltzmann equation. This problem is formalized using the perturbation theory. The foundation idea is to look for a linear expression which would allow to rely the parameters variations δx with one or several integral variation δy where y represents the physical observable. In other words, we wish to obtain $s_{y,x}$ coefficients which would be proportional to the first derivative $(\partial y / \partial x)_{x^0}$.

In the case of small variations of parameters and after the resolution of the Boltzmann direct and adjoint equations of the direct and the adjoint flux, the perturbation theory in the first order approximation results in sensitivity coefficients which are independent of the type of perturbations and which can be calculated once for all from the direct and adjoint flux (Ψ and Ψ^*). In our study we will essentially cover the caculation of the sensitivity coefficients associated to reactivity worths which are derived from the standard perturbation theory.

While noting that the considered perturbations, i.e. the multigroup self-shielded cross-sections, affect similarly the initial state and the state which is obtained after modification of the boron density or slab insert, we deduced the sensitivity coefficients associated to the reactivity variations simply by making the difference between perturbed and the reference situations. This yields to the usual expressions of the equivalent perturbation theory :

$$s_{y,x} \equiv s^{g}_{\Delta\rho,\bar{\sigma}_{i,r}} = \frac{\left(s^{g}_{\rho,\bar{\sigma}_{i,r}}\right)_{pert} - \left(s^{g}_{\rho,\bar{\sigma}_{i,r}}\right)_{ref}}{\Delta\rho} \text{ with } s^{g}_{\rho,\bar{\sigma}_{i,r}} \propto \left\langle \Psi^*, \delta B(\bar{\sigma}_{i,r})\Psi \right\rangle \tag{16}$$

where $\langle ...,... \rangle$ denotes the integration over the whole space of the modified Botzmann operator δB implying the multigroup cross-sections $\bar{\sigma}_{i,r}$ for the isotope i and the reaction r.

These coefficients are useful to identify the main nuclear phenomenon and reduce in the same time the number of basic data to be dealt with in the adjustment problem. In order to select the synthetic parameters, the applied method consists in using the total sensitivities (i.e. the sensitivity coefficients summed over the energy group and reactions) for all isotopes appearing in the compositions. As a matter of fact, since the perturbation of a partial cross-section usually generates same sign reactivity effects (capture, fission etc…) we can a priori exclude compensatory phenomenon between negative and positive values. This enables, for instance, to eliminate the isotopes for which the total sensitivity is less than 1pcm/%, which corresponds, for a 100% uncertainty, to an error on the eigenvalue equal to the calculation accuracy, i.e. 1pcm. In the case of slab geometry the sensitivities of the elements {Hf, ^{238}U, ^{235}U, Zr, ^{27}Al, ^{16}O, H_2O} inducing {capture, scattering or fission} reaction either in the moderator or as fissile/structure materials can be regrouped in a unique synthetic set.

Furthermore, it is necessary to look for an intermediary energy mesh between the 172 group energy mesh and the 1 gorup energy mesh. In order to include the major energy domains, we decided to adopt a 15 macro-groups energy mesh according with previous studies (see reference [9]). However, we made sure that the main nuclear phenomenon was taken into account in the different energy intervals and for the main studied nuclides. While using this energy mesh , the sensitivities in each macro-group were obtained by adding coefficients to the 172 group energy mesh.

In conjunction with the k_{eff} study and by analyzing the reactivity variations, we noticed that the sensitivities (correlation) between the absorbers efficiencies and the hafnium capture cross-sections were particularly high in the thermal range and in the resolved resonance range. As shown in table 4 the isotopes which contribute mainly to the total sensitivities for these integral parameters are the isotopes ^{177}Hf, ^{178}Hf and ^{179}Hf.

By using the definition (x) to express sensitivity coefficients we have still assumed that there is a simple linear relation between integral data and the basic parameters. Although Boltzmann equation involves linear mathematical operators, cross-sections normalizations performed during the spatial and energetic discretizations phases might possibly generate non linear effects and relationships.

A change in the nuclear data (such as resonance parameters) might affect the fine details of the neutron spectrum in the assembly under consideration to such an extent as to lead to non-negligibles changes in group constants (basically the sensitivity coefficients computed over the 172 group mesh). It recently was found such Spectra Fine-Structure Effects (called SFSE in the paper [20]) are of particular concern in the resonance range of U238 cross-section in thermal neutron reactor.

In order to examine the SFSE on our cross-section sensitivity analysis we first compared the sensitivity coefficients derived from direct calculations with those obtained initially from the application of the standard perturbation theory. With this direct method,we can thus compare the partial derivatives to the average reactivity slopes for a given parameter variation. In additon and the exact perturbation method implemented in APOLLO2 with the first order approximation enabled us to work out the (first order) linear terms and the (higher order) non-linear terms, we could validate, in certain instances, the calculated sensitivity.

The table 4 summarizes the resulting sensitivity coefficients for hafnium cross-sections and the reactivity variations.It appears that non linear effects involved in the computation of macroscopic cross-sections (due to homogeneization ore even leakage models) do not reach 5% for the major absorber isotopes and are sifficiently small to be neglected.

To complete this validation, we have also considered the energetic treatment of cross-sections and especially the effect of self-shielding calculation in the process of sensitivity coefficients. Until now, sensitivity coefficients for reactivity effectss was referring to self-shielded cross-sections. To take into account the group average weighting we shall define the following f-factor as the ratio between the self-shielded and the infinite dilute cross-section :

$$\tilde{\sigma}_{i,r} = \left(\overline{\sigma}_{i,r}\right)_{\infty} f_{i,r} \text{ in the group g} \tag{17}$$

The change in the g constant $\tilde{\sigma}_{i,r}$, due to any given nuclear data is :

$$\delta\tilde{\sigma}_{i,r} \approx f_{i,r}\,\delta\left(\overline{\sigma}_{i,r}\right)_{\infty} + \left(\overline{\sigma}_{i,r}\right)_{\infty}\delta f_{i,r} \tag{18}$$

If $\left(\overline{\sigma}_{i,r}\right)_{\infty}\delta f_{i,r} << f_{i,r}\,\delta\left(\overline{\sigma}_{i,r}\right)_{\infty}$ (i.e. if SFSE are negligible), the relation between a change in the infinite dilution cross-section (due to the nuclear data variation) and the change in the group constant is simply linear. If however the $\left(\overline{\sigma}_{i,r}\right)_{\infty}\delta f_{i,r}$ term is not negligible, then the relation between a change in a nuclear data and a change in the multigroup cross-sections becomes more untricated.

To determine these non-linear contributions, in the case of the hafnium cross-sections, we have alternatively included and excluded the elementary isotopes (177, 178, 179) during the self-shielding calculations (see reference [21] for the detailled formalism). This allowed us by the comparison of self-shielded and infinite dilute cross-sections to exhibit the global sensitvity coefficients :

$$\left(\delta\rho \Big/ \frac{\delta\tilde{\sigma}_{i,r}}{\tilde{\sigma}_{i,r}}\right)^{-1} \approx \left(\delta\rho \Big/ \frac{\delta\left(\overline{\sigma}_{i,r}\right)_{\infty}}{\left(\overline{\sigma}_{i,r}\right)_{\infty}}\right)^{-1} + \left(\delta\rho \Big/ \frac{\delta f_{i,r}}{f_{i,r}}\right)^{-1} \tag{19}$$

On the figure 2 and 3, we present the sensitivity profile and the sensitivity coefficients condensed in the 15 macrogroup energy mesh for the ^{177}Hf capture cross-sections. In the table 5 we have reported the global sensitivity deriving from the summation of the multigroup sensitivity coefficients over the whole energy range in the case of the 5mm hafnium plate. We shall remark first that figures 2 and 3 stress the high sensitivity peaks which correspond precisely to the of the 177 isotope between 1eV and 20eV.

In addtition, we see in table 5 that the self-shielding effect on the global sensitivities is relatively weak for the main absorbers. The relative variations between the sensitivities related to infinite dilute and self-shielded cross-sections are less than 6%. The major contribution of the f-factor appear for the high resonance of ^{178}Hf at 7.8 eV but do not exceed 6pcm/% in the macrogroup 12 (see the figure 4).

As shown on the figure 2 of the ^{177}Hf sensivity profile, the SFSE contribution oscillates and takes values close to zero. Furthermore we have noticed that the changes of thickness (5mm to 0,7mm) between the different hafnium plate give rise to non significant variations of the self-shielding effect. Regarding the 177 isotope the relative changes of the 15 macrogroup sensitivity coefficients rise 4 pcm/% (macrogroup 13 et 12) and for the 178 isotope is less than 6 pcm/% which is reached for the thicker plate.

Hence, this validation of the sensivity coefficients enabled us to use straighforwordly linear expression (first-order approximation of the perturbation theory) and allow the derivation of the overall sensitivity matrix.

5. POSTERIOR CROSS-SECTIONS AND C/E DISCREPANCIES

As shown on the figure 5, we have decided to solve the adjustment problem using the AMERE least-squares formalism follwing, according to two successive phases. During the first phase we have mainly tried to highlight the variations of the average cross-section (in a 15 macro-groups condensed energy mesh). Using the multigroup cross-sections as integral information and the nuclear (resonance) parameter as basic data we have assessed, from the previously observed variations, the corresponding adjustments. Moreover, in order to take into account the residual modelization biases and the initial hypothesis we have simultaneously applied the Lagrange's multipliers method to the experience/calculation discrepancies initiated from the APOLLO 2, TRIMARAN 2 and TRIPOLI 4 computations.

In each case, the used data rely on the same set of nuclear data (neutron cross-sections deriving from JEF2-2 evaluation). Uncertainties of these data therefore affect identically the calculation results. On the other hand, depending on the chosen modelization the calculated values do not exactly coincide and may lead to different adjustment results. Consequently, we must make sure that each series of experience/calculation discrepancies taken into account leads to consistent results over all selected parameters.

Results deriving from the first adjustment phase together with the contribution of each considered parameter appear in table 6. These values actually correspond for the APOLLO2 computation to the decomposition of the variations of integral parameters studied which is obtained by summing the cross-sections adjustments over the reactions and the energy groups. We observe that the cross-sections implied in the reducing of the experience/calculation discrepancies are mainly hafnium capture cross-sections. As far as the reactivity widths are concerned and in the case of the 7 mm plate, the hafnium adjustment cross-sections effect reaches up to 5% and implies almost exclusively the capture of the 177 isotope. In addition, we observe from the figure 5 that the 15 macro-groups cross-sections adjustments are consistent with all used calculation methods and all the isotopes. It also appears clearly that the reducing of the 177 isotope capture cross-sections in the main resonance range is included into the 15/20% interval. Furthermore, the statistical khi-square test shows that the gaussian distribution assumption is complied with regardless of the used calculation methods (the a posterior khi-square value is less than 1).

The above table 7 indicates in our case that the posterior chi-square theoretical margins are complied whether the APOLLO2 or the TRIMARAN 2 and TRIPOLI 4 type of calculations are used. The chi-square values being all less than 1 after adjustment , the normal law hypothesis used together with the maximum of probability principle are therefore totally valid.

In addition, when we detail the experience/calculation discrepancies before and after adjustment according to the integral parameters studied, we observe over whole a clear improvement of the values deriving from our first interpretation (prior discrepancies).

As stated in table 8, the main effect of the adjustment affects the reducing of the discrepancies when they relate to the integral efficiency of the absorber plates (the reduction being essentially obtained by the decrease of the average ^{177}Hf(n,γ) capture cross-section). As for the multigroup cross-sections these adjustments are almost independent from the models (all the posterior chi-square values being registered in the theoritical margins). We can notice that the effect of the adjustment on the TRIPOLI4 type of calculations (the reference calculation performed according to a continuous-energy description of the cross-sections) bears as much significance. In the case of thin hafnium slab the reducing of (E-C)/C discrepancies can reach up to 5.25% (case of the 0.7mm slab). These results are particularly interesting insofar as the discrepancies observed before the implementation of the method between the bore and hafnium calculation (due to hafinum parameters) almost disappears after the self-shielded cross-sections are modified.

The purpose of the second phase thus consists in studying thoroughly the adjustments highlighted at the level of the average cross-sections in order to translate them afterwards in terms of nuclear data. In fact, if we use the average self-shielded cross-sections which depend now on resonance parameters as integral data**,** the adjustment issue is therefore totally similar to the one dealt with during the first phase. By using those results together with the sensitivity coefficients $s^{j}_{\bar{\sigma}_\gamma,\Gamma_n}$ and $s^{j}_{\bar{\sigma}_\gamma,\Gamma_\gamma}$ derived from the single level Breit-wigner model, we can therefore apply the Lagrange multipliers method (i.e. reprocess the AMERE code) and look for the corrections to be applied to the resonance parameters E_{0j}, Γ_{nj} and $\Gamma_{\gamma j}$

We have gathered in table 9 the adjustment of the resonance parameters of the 177 isotope which have been obtained (in the main resonance area) from the TRIPOLI 4 type of calculations. We observe an important decrease in the neutron width and consequently of the total width. In particular, the correction of the 6.57 eV neutron width (with spin j = 4) which is worth –5% contributes for more than 37% to the reducing of discrepancy on the capture cross-section between 4 eV and 22.6 eV. In this macro group the –15% variation which we had identified during the first phase is almost absorbed after changing this nuclear parameter, the residual discrepancy being within the 2% range.

At the end of the full adjustment process, we thus reach quite fine corrections of the parameters of the ^{177}Hf resonances. As far as the JEF 2-2 evaluation is concerned the neutron widths must globally be reduced in the 1eV-20eV area (as reported in table 9). The radiative widths and the resonance energy peaks are not really affected by the adjustment. The energy resolution is generally quite good and the low sensitivity of the capture cross-sections to the parameters $\Gamma_{\gamma j}$ (see table 2) does not generate noticeable changes of the average values (around 65 meV). It is therefore interesting to evaluate at this point, the impact of the adjustments on the form and structure of the major resonances.

In figure 7 we have represented especially the impact of the adjustment in the neighborhood of the 6.57eV resonance of the ^{177}Hf. We can observe that the reducing of the neutron width compared with the initial JEF-2.2 evaluation affects in particular the resonance peak, i.e. the maximum contribution of the resonance term which behaves like $g_j\Gamma_{nj}/\Gamma_j$ (g_j is the so called westcott factor). In this particular case, we must underline the fact that the information deriving from the integral experiments and the one deriving from the differential experiments is complementary. In fact, beyond the first hafnium resonance, experimentalists (references [14-16]) deduced, in conclusion of the area and shape analysis of the transmission data standard deviation related to the neutron widths comprised between 5% and 20% (at 6.57eV). We can therefore consider that taking into account plates experiments compliments the knowledge acquired so far on these nuclear data.

6. CONCLUSION

The essential role of integral experiments for data validation has been recognized for a long time. However, recent progress in the computational methods (statistical/deterministic) has allowed to obtain from integral experiments additionnal information for the cross-sections evaluation, not only in terms of possible data adjustment at the level of multigroup value, but also in terms of basic nuclear parameters.

This paper fulfills, notably, how specific integral measurements (the reactivity woths deduced from slab experminents) enable us to focus on particular nuclear data (hafnium resonance parameters) and leave the possibility to reduce their corresponding uncertainties by an adapted adjustment method. In the framework of the standard mean-squares adjustment method, it is shown that the use of deterministic methods together with Monte Carlo- type simulations allows a depth analysis of the modelling approximations to be carried out. Furthermore, the sensitivity coefficient validation technique employed and the study of spectra fine structure effects particularly leads to a reliable assessment of the quality of the new basic nuclear data.

In this instance, it has been established that the correlations beetween integral data and hafnium capture parameters induce significative variations of multigroup and punctual cross-sections within the resolved resonance range (both deriving from the actual JEF-2.2 evaluation) to be highlighted. The adjustments proposed for certain ^{177}Hf resonance parameters (neutron width between 1 eV and 20 eV) reduce, after error propagation, by 3 to 5 per cent the difference between experimental results (reactivity effects) and calculations (for the detailled results see also the reference [22]).

Beyond this application, we have planned to generalize the statistical adjustment approach in order to treat formally other inverse problem and basic sizing parameters (chemical/geometric data or other unexplored nuclear data) to make technological requirements less stringent. In particular, the numerical validation results of the physical models should be advantagiously introduce in an additional error matrix while the likelihood function and the minimization routine remain the same work.

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Table 1 : Hf thermal capture cross-sections and resonance integrals quoted in the international evaluation files

Evaluation	Isotope	σ_γ^0(barns)	I_γ (barns)
JEF-2.2	^{174}Hf	403,8	320,3
ENDF/B-VI	^{174}Hf	564,9	355,1
JENDL-3.2	^{174}Hf	561,5	361,8
JEF-2.2	^{176}Hf	14,07	612,8
ENDF/B-VI	^{176}Hf	13,84	400,2
JENDL-3.2	^{176}Hf	23,50	892,7
JEF-2.2	^{177}Hf	376,2	7232
ENDF/B-VI	^{177}Hf	375,5	7221
JENDL-3.2	^{177}Hf	373,7	7209
JEF-2.2	^{178}Hf	78,48	1922
ENDF/B-VI	^{178}Hf	84,49	1915
JENDL-3.2	^{178}Hf	84,07	1914
JEF-2.2	^{179}Hf	39,15	543,1
ENDF/B-VI	^{179}Hf	43,82	548,6
JENDL-3.2	^{179}Hf	42,81	521,6
JEF-2.2	^{180}Hf	13,08	35,44
ENDF/B-VI	^{180}Hf	13,08	34,28
JENDL-3.2	^{180}Hf	13,00	33,85

Table 2 : ^{177}Hf capture cross-sections uncertainties deriving from the nuclear data measurements in the 1eV-20eV resolved resonance range

E_{0j} (eV)	$\delta E_{0j}/E_{0j}$ (%)	Γ_{nj} (meV)	$\delta\Gamma_{nj}/\Gamma_{nj}$ (%)	$\Gamma_{\gamma j}$ (meV)	$\delta\Gamma_{\gamma j}/\Gamma_{\gamma j}$ (%)	$s_{\bar{\sigma}_{\gamma j},\Gamma_{nj}}$	$\delta\bar{\sigma}_{\gamma j}/\bar{\sigma}_{\gamma j}$ (%)
1,096	0,2	2,22	1,6	62,83	1,5	0,97	1,9
2,384	0,2	8	3,4	61	1,6	0,88	3,6
5,89	0,3	5,35	5,9	65,47	-	0,92	6,1
6,57	**0,3**	**8,05**	**20**	**64,96**	**-**	**0,89**	**18,4**
8,86	0,2	5,71	10,3	64,97	-	0,92	9,9
10,94	0,3	0,5	8,5	75,52	-	0,99	9
13,69	0,2	0,54	8,5	64,82	-	0,99	8,9
13,97	0,1	3,06	9,7	74,56	19	0,96	9,8
22,01	0,2	1,57	5,3	67,34	-	97,73	5,7
22,31	0,1	0,76	13,7	66,5	-	98,87	13,8

JEF-2.2 references [14-16] : Liou (1975), Fuketa (1965), Rhor (1972/1975)

Table 3 : Prior discrepancies between the experimental and the computed integral parameters (reactivity worths)

Reactivity worths	Exp. (pcm)	Std dev. (1σ)	(E-C)/C Apollo2	(E-C)/C Trimaran2	Std dev. (MC/Exp)	(E-C)/C Tripoli4	Std dev. (MC/Exp)
$\Delta\rho^{hafnium}_{5mm}$	-7001	4,43 %	-5,55 %	-4,35 %	4,83 %	-3,57 %	4,64 %
$\Delta\rho^{hafnium}_{2.8mm}$	-6072	4,21 %	-6,80 %	-6,83 %	4,73 %	-7,71 %	4,48 %
$\Delta\rho^{hafnium}_{2.8mm+eau}$	-6861	4,44 %	-2,64 %	-1,93 %	4,87 %	-2,52 %	4,66 %
$\Delta\rho^{hafnium}_{1.4mm}$	-5030	4,49 %	-6,66 %	-5,02 %	5,22 %	-6,45 %	4,86 %
$\Delta\rho^{hafnium}_{0.7mm}$	-3916	4,50 %	-7,31 %	-6,92 %	5,61 %	-7,95 %	5,07 %

Table 4 : Study of non-linear effects during the computation of the Hf sensitivity coefficients relying the reactivity worths and the multigroup capture cross-sections (unit : pcm/%)

Isotopes	Premier ordre	Ordres sup.	Total	directe
^{174}Hf	1	0	1	2
^{176}Hf	8	0	8	7
^{177}Hf	158	**1**	159	148
^{178}Hf	36	0	36	35
^{179}Hf	40	0	40	41
^{180}Hf	7	0	7	6

Table 5 : Self-shielding effect on the computation of the total Hf sensitivities (unit : pcm/%)

Isotopes	Self-shielded cross-sections	Infinite dilute cross-sections
^{174}Hf	1	1
^{176}Hf	8	8
^{177}Hf	158	149
^{178}Hf	36	35
^{179}Hf	40	42
^{180}Hf	7	12

Table 6 : Contribution of each considered cross-sections to the first ajustment phase results (APOLLO2 computation)

Reactivity worths	prior (E-C)/C	^{238}U (pcm)	^{235}U (pcm)	Zr_{nat} (pcm)	^{16}O (pcm)	^{10}B (pcm)	H_2O (pcm)	^{174}Hf (pcm)	^{176}Hf (pcm)	^{177}Hf (pcm)	^{178}Hf (pcm)	^{179}Hf (pcm)	^{180}Hf (pcm)	posterior (E-C*)/C*
$\Delta\rho^{hafnium}_{5mm}$	-5,55 %	16	-12	-21	-2	8	-281	0	1	2253	29	40	1	**-3,51 %**
$\Delta\rho^{hafnium}_{2.8mm}$	-6,80 %	16	-13	-22	-2	9	-225	0	1	3003	33	42	1	**-3,96 %**
$\Delta\rho^{hafnium}_{2.8mm+eau}$	-2,64 %	15	-12	-21	-2	8	-758	0	1	2861	31	40	1	**-0,47 %**
$\Delta\rho^{hafnium}_{1.4mm}$	-6,66 %	16	-14	-23	-2	12	-749	0	1	4100	41	42	2	**-3,24 %**
$\Delta\rho^{hafnium}_{0.7mm}$	-7,31 %	17	-15	-24	-2	15	-774	0	1	5375	52	43	3	**-2,62 %**

Table 7 : Khi-square test of the adjustment results (reference and standard computations)

(E-C)/C	Theoritical margin	$(\chi^2_1)_{prior}$	$(\chi^2_1)_{posterior}$
APOLLO2	0,63485 / 1,3651	3,44723	0,88865
TRIMARAN2	0,63485 / 1,3651	1,37942	0,96069
TRIPOLI4	0,63485 / 1,3651	1,51334	0,81499

Table 8 : Experimental and computed reactivity effects discrepancies derived from the reference transport computation before and after the adjustment process (with the corresponding reduced uncertainty)

Integral parameter	(E-C)/C TRIPOLI4	(E-C*)/C* TRIPOLI4	Reduced uncertainty (2σ)
$\Delta\rho^{hafnium}_{5mm}$	**-3.57 %**	**-0.83 %**	4.64 %
$\Delta\rho^{hafnium}_{2.8mm}$	**-7.71 %**	**-4.40 %**	4.48 %
$\Delta\rho^{hafnium}_{2.8mm+eau}$	**-2.52 %**	**0.68 %**	4.66 %
$\Delta\rho^{hafnium}_{1.4mm}$	**-6.45 %**	**-2.20 %**	4.86 %
$\Delta\rho^{hafnium}_{0.7mm}$	**-7.95 %**	**-2.70 %**	5.07 %

Table 9 : Improvements in the knowledge of ^{177}Hf nuclear parameters (file JEF-2.2)

Resonance	Γ_{nj} Variation (%)
1.098 eV	-0.30
2.388 eV	-1.29
5.89 eV	-0.45
6.6 eV	**-4.97**
8.88 eV	-1.36
10.95 eV	-1.00
13.67 eV	-1.01
13.96 eV	-1.27
21.97 eV	-0.39
22.26 eV	-2.59

Figure 1 : Integral slab experiments (reference assembly of the AZUR program)

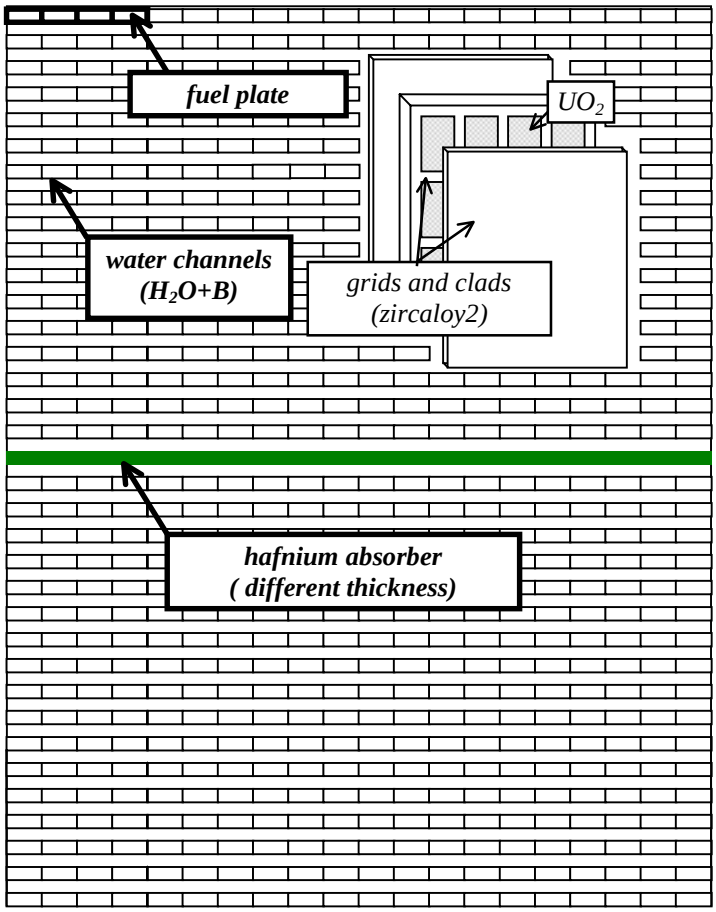


Figure 2 : Sensitivity profile of reactivity worth (5mm absorber plate) to the ^{177}Hf capture cross-sections

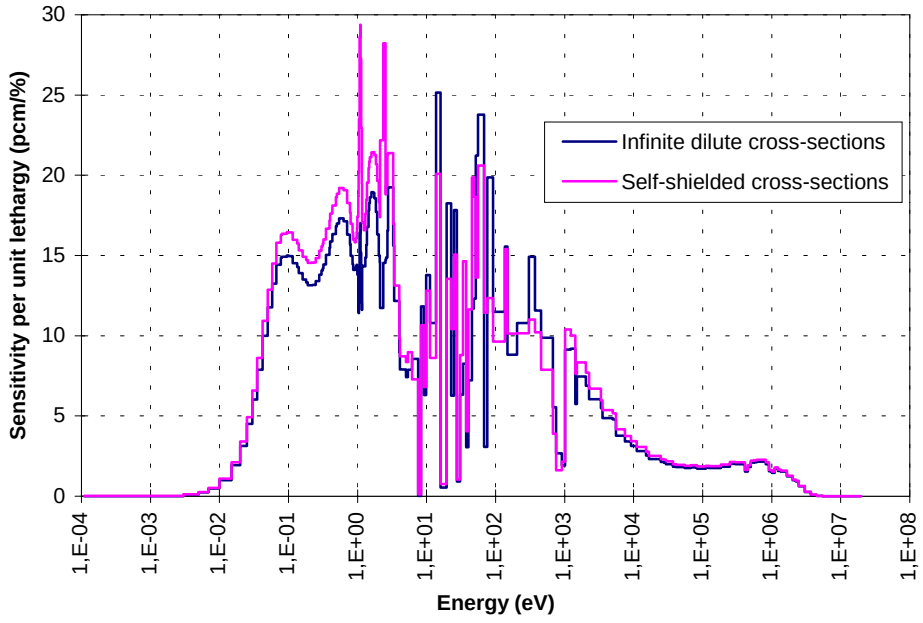


Figure 3 : 15 macrogroup sensitivity of reactivity worth (5mm absorber plate) to the ^{177}Hf capture cross-sections

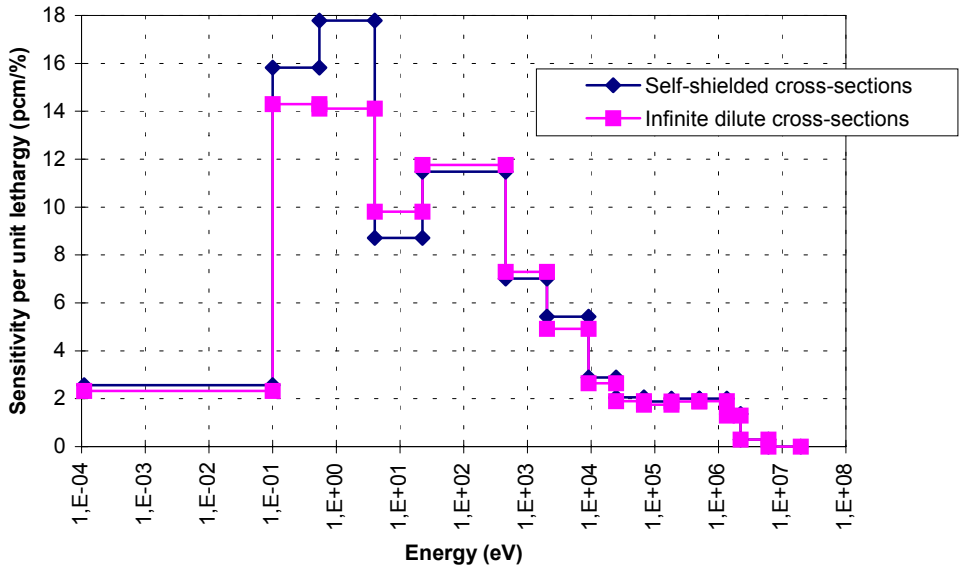


Figure 4 : Sensitivity profile of reactivity worth (5mm absorber plate) to the ^{178}Hf capture cross-sections

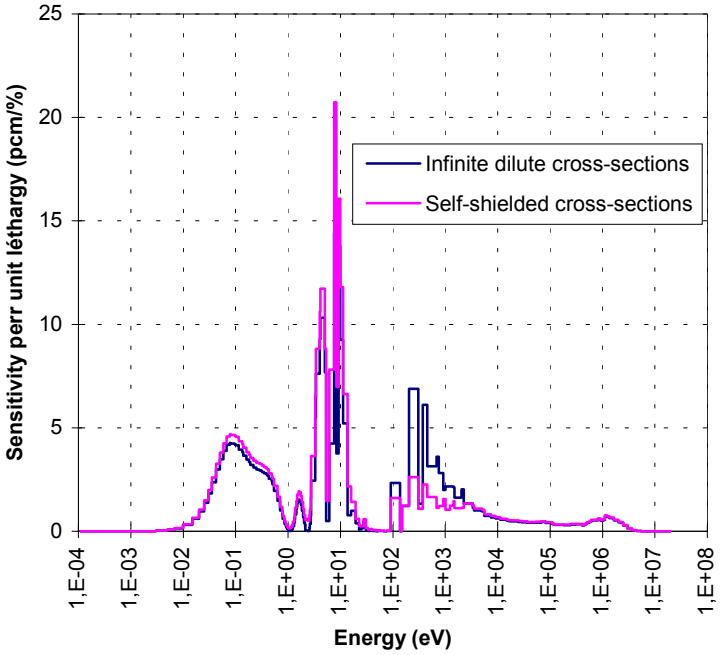


Figure 5 : Two-phases adjustment scheme

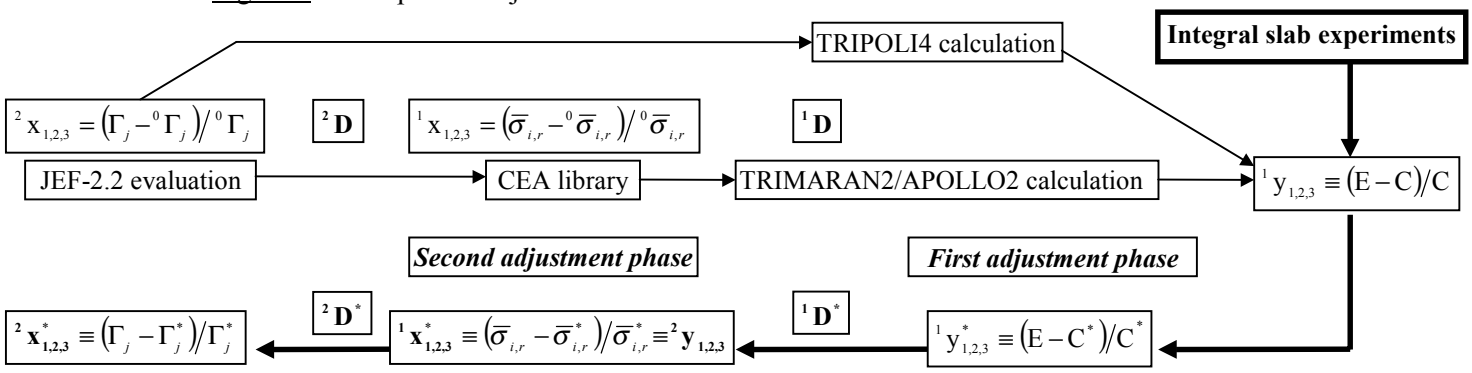


Figure 6 : Resulting adjustments vs several C/E - capture 177Hf

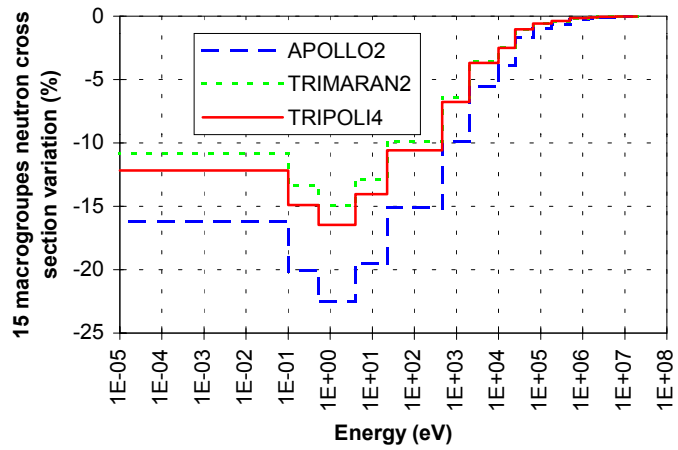
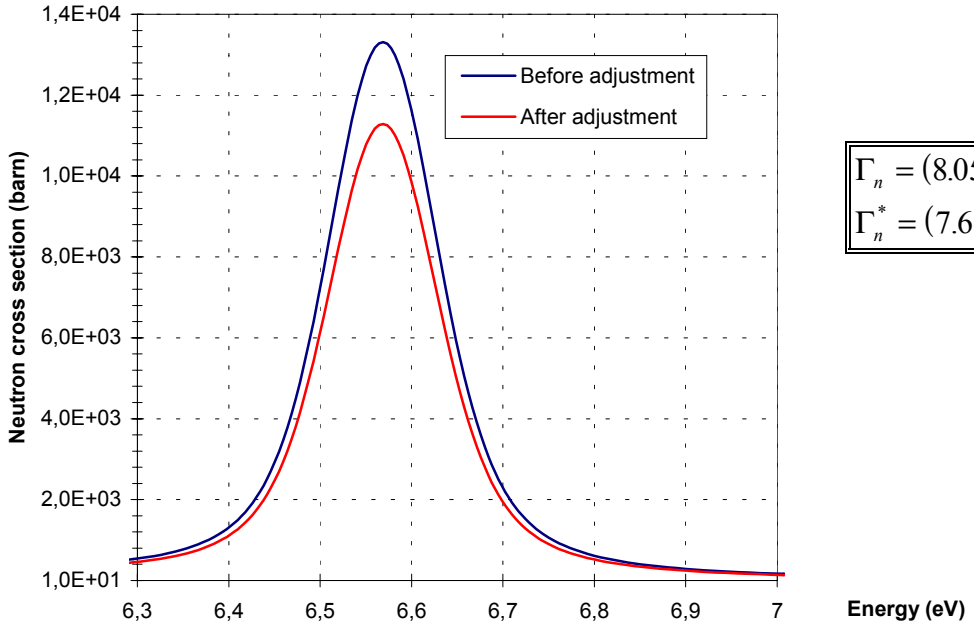


Figure 6 : Recommended adjustment for a particular resonance of Hf^{177} isotope



$$\Gamma_n = (8.05 \pm 1.61) meV$$
$$\Gamma_n^{*} = (7.65 \pm 1.45) meV$$