Generation of the $^{1}\text{H}$ in H$_{2}$O neutron thermal scattering law covariance matrix of the CAB model
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Abstract. The thermal scattering law (TSL) of $^1\text{H}$ in $\text{H}_2\text{O}$ describes the interaction of the neutron with the hydrogen bound to light water. No recommended procedure exists for computing covariances of TLSs available in the international evaluated nuclear data libraries. This work presents an analytic methodology to produce such a covariance matrix-associated to the water model developed at the Atomic Center of Bariloche (Centro Atomico Bariloche, CAB, Argentina). This model is called as CAB model, it calculates the TSL of hydrogen bound to light water from molecular dynamic simulations. The performance of the obtained covariance matrix has been quantified on integral calculations at “cold” reactor conditions between 20 and 80 °C. For UOX fuel, the uncertainty on the calculated reactivity ranges from ±71 to ±155 pcm. For MOX fuel, it ranges from ±110 to ±203 pcm.

1 Introduction

The calculation of a critical system is carried out by means of reactor physics simulation code that uses evaluated nuclear data. The evaluated nuclear data libraries contain reaction information necessary to quantify the neutronic parameters that describe the behavior of the system. In light water reactor calculations, neutrons are slowed down by the $^1\text{H}$ in $\text{H}_2\text{O}$ inelastic thermal scattering data, which are expressed in terms of thermal scattering law (TSL). The TSL describes the dynamics of the scattering target and gives information about the energy and angle of the scattered neutrons. To evaluate the safety margins, the uncertainties coming from the nuclear data have to be assessed. However, no covariance information for the TSL of $^1\text{H}$ in $\text{H}_2\text{O}$ is available in any nuclear data library.

Mathematical frameworks for producing covariance matrix for the TSL exist. In a previous work, a Monte-Carlo methodology was developed and applied to hexagonal graphite [1]. A different procedure based on an analytic method was also recently proposed [2]. It was applied to the TSL of $^1\text{H}$ in $\text{H}_2\text{O}$-associated to the JEFF-3.1.1 nuclear data library [3].

A new model for light water, namely the CAB model, was developed at the atomic center of Bariloche in Argentina [4]. The originality of this model relies on the use of molecular dynamic simulations for calculating the density of states of hydrogen in the water molecule. The objective of the present work is to produce a covariance matrix between the CAB model parameters and to test its performance on integral calculations between 20 and 80 °C.

2 Thermal inelastic neutron scattering

A description of the thermal scattering theory can be found in references [5,6]. In this section, an introductory background will be given to set the basis for the present work.

Working in the incoherent approximation, the total cross section of $\text{H}_2\text{O}$ as a function of the incident neutron energy $E_n$ is given by:

$$\sigma_{T}^{\text{H}_2\text{O}}(E_n) = 2\sigma_H^T(E_n) + \sigma_O^T(E_n),$$

(1)

where $\sigma_O^T$ is the total cross section of $^{16}\text{O}$ and $\sigma_H^T$ is the total cross section of $^1\text{H}$ which is given by:

$$\sigma_H^T(E_n) = \sigma_\nu(E_n) + \sigma_n(E_n).$$

(2)

For light isotopes, in the thermal energy range, the capture cross section $\sigma_\nu(E_n)$ can be approximated as:

$$\sigma_\nu(E_n) = \sigma_\nu^0 \sqrt{\frac{E_0}{E_n}},$$

(3)

where $\sigma_\nu^0$ is the capture cross section measured at the thermal neutron energy ($E_0 = 25.3$ meV).
In the low energy range, typically below 5 eV, the slowing down of neutrons in water is affected by the chemical bonds between the hydrogen and oxygen atoms. Such impact is taken into account in the neutronic calculations by using the double differential scattering cross section:

$$\sigma_n(E_n) = \int \frac{d^2\sigma_n}{d\theta dE} d\theta dE.$$  \hspace{1cm} (4)

The double differential cross section expresses the probability that an incident neutron of energy $E_n$ will be scattered at a secondary energy $E$ and direction $\theta$. If $T$ is the temperature of the target and $k_B$ is the Boltzmann constant, the double differential scattering cross section for $^1$H in H$_2$O is calculated as [7]:

$$\frac{d^2\sigma_n}{d\theta dE} = \frac{\sigma_b}{4\pi k_B T} \sqrt{\frac{E}{E_n}} e^{-\frac{\Delta E}{k_B T}} S(\alpha \beta),$$ \hspace{1cm} (5)

where $\sigma_b$ is the bound scattering cross section of hydrogen and $S(\alpha, \beta)$ is the so-called thermal self-scattering function (or alternatively thermal scattering law), defined as a function of the dimensionless momentum transfer $\alpha$ and energy transfer $\beta$:

$$\alpha = \frac{E + E_n - 2\sqrt{EE_n \mu}}{A k_B T},$$ \hspace{1cm} (6)

$$\beta = \frac{E - E_n}{k_B T},$$ \hspace{1cm} (7)

where $\mu$ is the cosine of the scattering angle $\theta$ ($\mu = \cos(\theta)$) in the laboratory system and $A$ is the ratio of the scattering target to the neutron mass.

In practice, the calculation of the scattering law is performed with the LEAPR module of the NJOY processing system [8], in which the key parameter is the frequency spectrum $\rho(\beta)$ of $^1$H in H$_2$O. The frequency spectrum characterizes the excitations states of the material. In the CAB model, it is introduced in the LEAPR module as a decomposition of three partial spectra:

$$\rho(\beta) = \sum_{i=1}^{2} \omega_i \delta(\beta_i) + \omega_p \rho_p(\beta) + \omega_c \rho_c(\beta).$$ \hspace{1cm} (8)

The discrete oscillators are represented by $\delta(\beta_i)$ for $i = 1, 2$. They describe the intramolecular modes of vibration, where $\beta_i$ is the energy and $\omega_i$ the associated weight. The continuous frequency distribution $\rho_c(\beta)$ models the intermolecular modes. The weight corresponding to this partial spectrum is $\omega_c$. Finally, $\rho_p$ accounts for the translation of the molecule.

### 3 The CAB model

The frequency spectrum of $^1$H in H$_2$O of the CAB model [4] was calculated using the molecular dynamic simulation code GROMACS [9]. The water potential implemented in the code was the TIP4P/2005f potential [10].

| Parameter | Value |
|-----------|-------|
| $\sigma_0$ (nm) | 3.1644 |
| $\varepsilon_0$ (kJ/mol) | 0.7749 |
| $q_H$ (e$^-$) | 0.5564 |
| $q_M$ (e$^-$) | -1.1128 |
| $d_{OH}$ (nm) | 0.09419 |
| $D_{OH}$ (kJ/mol) | 432.581 |
| $\beta_{OH}$ (1/nm) | 22.87 |
| $\theta_{OH}$ (°) | 107.4 |
| $k_b$ (kJ/mol/rad$^2$) | 367.81 |
| $d_{OM}$ (nm) | 0.15555 |

### 3.1 The parameters of the CAB model

The parameters of the CAB model correspond to the TIP4P/2005f water potential. This potential is a flexible potential with four positions: two hydrogen atoms, one oxygen and one so-called M-site (dummy atom). The dummy atom is located over the angle bisector formed by the two hydrogens and the oxygen. Table 1 lists the TIP4P/2005f water potential parameters used in the CAB model.

The intermolecular interactions are represented by a Lennard-Jones potential $V_{LJ}$ between the oxygen atoms:

$$V_{LJ}(r_{ij}) = 4\varepsilon_0 \left[ \left( \frac{\sigma_0}{r_{ij}} \right)^{12} - \left( \frac{\sigma_0}{r_{ij}} \right)^{6} \right],$$ \hspace{1cm} (9)

and the Coulomb potential $V_c$ is given by:

$$V_c(r_{ij}) = k \frac{q_i q_j}{r_{ij}},$$ \hspace{1cm} (10)

where $\varepsilon_0$ is the depth of the potential well, $\sigma_0$ represents the distance where the potential is zero, $k$ is the Coulomb constant, $q_i$ is the electrical charge of the particle and $r_{ij}$ stands for the distance between two atoms.

The intramolecular interactions are characterized by a Morse potential $V_M$. It accounts the stretching of the hydrogen–oxygen bond as follows:

$$V_M(r_{ij}) = D_{OH} \left[ 1 - e^{-\beta_{OH}(r_{ij} - d_{OH})} \right].$$ \hspace{1cm} (11)

For the bending mode, the harmonic angle potential $V_{HOH}$ is:

$$V_{HOH}(\theta_{ij}) = \frac{1}{2} k_b (\theta_{ij} - \theta_0)^2.$$ \hspace{1cm} (12)

In the above equations, $D_{OH}$ is the depth of the potential well, $\beta_{OH}$ is the steepness of the well, $d_{OH}$ is the equilibrium distance between the oxygen and the hydrogen, $k_b$ the strength constant and $\theta_0$ is the equilibrium angle between the hydrogens and oxygen.
3.2 Frequency spectrum of $^1$H in $\text{H}_2\text{O}$ used in the CAB model

In the CAB model, the translational mode is modeled with the Egelstaff-Schofield diffusion model [11]. The continuous frequency spectrum of $^1$H in $\text{H}_2\text{O}$ is then obtained by subtracting the Egelstaff-Schofield spectrum to the generalized frequency spectrum obtained from molecular dynamic simulations [4].

The continuous frequency spectrum of $^1$H in $\text{H}_2\text{O}$ as well as the discrete oscillators modeling the intramolecular modes at 294 K are shown in Figure 1. The continuous spectrum is dominated by the libration mode ($\approx 70$ meV). The structures of small amplitude observed at very low energy transfer ($\approx 5$ and $\approx 30$ meV) were observed experimentally [12] and are still visible even with a rigid model [13]. They should correspond to vibrational modes between the hydrogen and oxygen atoms of different water molecules.

The translational weight $v_t$, involved in the Egelstaff-Schofield diffusion model, was deduced from experimental measures of Novikov [14] of diffusion masses for light water at different temperatures. Table 2 summarizes the LEAPR parameters of the CAB model at 294 K and the weights corresponding to each vibration mode.

3.3 The average cosine of the scattering angle calculated with the CAB model in the laboratory system

The integration over the secondary energy $E$ of equation (5) gives the simple differential cross section (or angular distribution). The average for each incident neutron energy gives the average cosine $\bar{\mu}$ of the scattering angle:

$$
\bar{\mu}(E_n) = \frac{\int_0^\theta \cos \theta \sin \theta \left( \int_0^\infty \frac{d^2\sigma}{dE\,dE} \, dE \right) \, d\theta}{\int_0^\theta \sin \theta \left( \int_0^\infty \frac{d^2\sigma}{dE\,dE} \, dE \right) \, d\theta}.
$$

In Figure 2 it is compared the data measured by Beyster et al. [15] and the average cosine of the scattering angle calculated with the CAB model at 294 K. An overall good agreement is obtained between the calculated curve and the data.

3.4 The $\text{H}_2\text{O}$ total cross section calculated with the CAB model

The total cross section $\sigma_{\text{H}_2\text{O}}^\text{H}$ calculated with the CAB model at 294 K is shown in Figure 3. The theoretical curve is compared with a set of selected data measured at room temperature [16–19]. The CAB model correctly reproduces the measured values over the full energy range. Therefore, the generation of the covariance matrix will consists of determining the uncertainties of the CAB model parameters without changing their values.

4 Methodology for producing covariance matrices with the CONRAD code

The covariance matrix between the CAB model parameters was analytically calculated using the CONRAD (code for nuclear reaction analysis and data assimilation) code [20]. The methodology relies on a generalized least-square fitting algorithm and on the marginalization technique.

4.1 The generalized least-square method

The generalized least-square method implemented in the CONRAD code is designed to provide a set of best-estimate model parameters given a set of experimental data. It is based on the Bayes theorem [21], which states that the posterior information of a quantity is proportional to the prior, times a likelihood function, which yields the probability to obtain an experimental data set $\tilde{y}$ for a given model parameters $\tilde{x}$.

![Fig. 1. Continuous frequency spectrum of $^1$H in $\text{H}_2\text{O}$ and internal vibration modes ($E_1 = 205$ meV and $E_2 = 415$ meV) of the CAB model as a function of the excitation energy at $T = 294$ K.](image-url)
In the CONRAD code, the procedure consists of resolving iteratively by the Newton–Raphson method the following sets of equations for the model parameter $\vec{x}$ and covariance matrix $M_x$ [22]:

$$
\vec{x}^i = \vec{x}^{i-1} M_x^{-1} (G_x^{-1})^T (M_y)^{-1} (\vec{y} - \vec{t}^{i-1}), \quad (14)
$$

$$
(M_x^i)^{-1} = (M_x^{i-1})^{-1} (G_x^{i-1})^T (M_y)^{-1} G_x^{i-1}, \quad (15)
$$

where $M_y$ is the experimental covariance matrix and $\vec{t}$ is the theoretical model. The matrix $G_x$ is the derivative matrix of the theoretical model with respect to the parameters $\vec{x}$:

Fig. 2. Average cosine of the scattering angle calculated with the CAB model compared with experimental data at 294 K [15].

Fig. 3. Total cross section calculated with the CAB model at 294 K compared with experimental data [16–19].
4.2 The marginalization technique

The marginalization technique was designed to take into account the uncertainties of systematic origin in the nuclear data evaluating process. Such type of uncertainties usually introduce strong correlations between the experimental values.

These parameters, called nuisance parameters, correspond to the aspect of physical realities whose properties are not of particular interest as such but are fundamental for assessing reliable model parameters [23].

If \( \bar{\theta} = (\theta_1, \ldots, \theta_m) \) is the nuisance parameter vector and \( M_0 \) stands for the covariance matrix, then the posterior covariance matrix after the marginalization \( M_{x|marg} \) is obtained as [24]:

\[
M_{x|marg} = M_x + (G^T_x G_x)^{-1} G^T_x G_\theta M_0 G^T_x G_x (G^T_x G_x)^{-1},
\]

where the matrix \( G_\theta \) is the derivative matrix of the theoretical model with respect to the nuisance parameters vector:

\[
G_\theta = \left( \begin{array}{cccc}
\frac{\partial \theta_1}{\partial \theta_1} & \cdots & \frac{\partial \theta_1}{\partial \theta_m} \\
\vdots & \ddots & \vdots \\
\frac{\partial \theta_k}{\partial \theta_1} & \cdots & \frac{\partial \theta_k}{\partial \theta_m}
\end{array} \right).
\]

If we define the extended model parameter vector as

\[
\bar{x} = (x, \bar{\theta}),
\]

then the full covariance matrix \( \Sigma \) between \( (x, \bar{\theta}) \) is expressed as:

\[
\Sigma = \begin{pmatrix}
M_{x|marg} & M_{x,\theta} \\
M_{x,\theta}^T & M_{\theta}
\end{pmatrix}.
\]

The cross-covariance term \( M_{x,\theta} \) is calculated by introducing “variance penalty” terms [25]. The “variance penalty” is a measure of the contribution of the uncertainty of the nuisance variables to the variance of the calculated quantity \( t \). The cross-covariance term is:

\[
M_{x,\theta} = - (G^T_x G_x)^{-1} G^T_x G_\theta M_\theta.
\]

The following section explains how the generalized least-square method and the marginalization technique were applied to calculate the covariance matrix between the CAB model parameters.
Finally, the nuisance parameter vector is:

\[ \tilde{\theta} = (n, N, B, T, \omega_1, \sigma^H_0). \]  

(24)

Table 3 summarizes the nuisance parameters with the uncertainties adopted for each experimental data set.

6 Results

The covariance matrix \( \Sigma \) between the model parameters was determined with the CONRAD code by using a two-step calculation scheme. The generalized least-square method provides the covariance matrix between the CAB model parameters \( M_x \) (Eq. (15)). Afterwards, these results are used in the marginalization technique to calculate the posterior covariance matrix \( M^\text{post} \) (Eq. (17)).

At the beginning of the fitting procedure, it is assumed that the CAB model parameters are uncorrelated and have relative prior uncertainties of 1\%. The posterior uncertainties reported in Table 4 are rather low. They lie below the prior uncertainties. The correlation matrix shows weak correlations between the parameters.

After the marginalization of the nuisance parameters, stronger correlations between the model parameters are calculated. Table 5 summarizes the relative uncertainties of the CAB model parameters and their correlations. Compared with the results after the fit, it can be seen that more realistic uncertainties are achieved.

The uncertainties range between 2 and 6\%, excepted for the parameter \( \varepsilon_0 \), which is involved in the expression of the Lennard–Jones potential between the oxygens. Its relative uncertainty reaches 14.6\%. Such result indicates that the calculated uncertainties on the CAB model parameters must be taken with care. If the parameters of the water potential remain within such 1\% uncertainties, then the forces between the atoms originated by the potentials would be severely modified. These perturbations would probably introduce unphysical changes at the level of the water molecule. Therefore, we have to keep in mind that the present results are only dedicated to generate usable uncertainties in applied neutronic field.

7 Uncertainties propagation of the CAB model parameters

7.1 Covariance matrix of the thermal scattering function

The thermal scattering function contains a very large number of values. To solve this difficulty, the \( S(\alpha, \beta) \) values were averaged in 37 momentum transfer intervals. The average scattering function \( \bar{S}_{ij}(\alpha_{ij}, \beta_0) \), for a given energy transfer \( \beta_0 \), is obtained as follows:

\[
\bar{S}_{ij}(\alpha_{ij}, \beta_0) = \frac{\int_{\alpha_{ij}}^{\infty} S(\alpha, \beta_0) d\alpha}{\int_{\alpha_{ij}}^{\infty} d\alpha}.
\]

(25)

Figure 5 shows the symmetric forms of \( S(\alpha, \beta_0) \) and \( \bar{S}(\alpha, \beta_0) \) as a function of the momentum transfer for \( \beta_0 = 1.0 \) calculated at 294 K. Figure 6 shows the relative uncertainties and the correlation matrix of the multigroup scattering function for two energy transfers (\( \beta_0 = 1.0 \) and 10.0). They were obtained from the propagation of the CAB model parameter uncertainties reported in Table 5.
In both cases the relative uncertainties on the $S_{ij}(\alpha_{ij}, \beta_0)$ function range between 10% in the peak of the distribution up to approximately 30% in the wings.

### 7.2 Covariance matrix of the $^1$H in H$_2$O scattering cross section

The left-hand plot of Figure 7 shows the relative uncertainties and the correlation matrix of the $^1$H in H$_2$O scattering cross section after the uncertainty propagation of the CAB model parameters at 294 K. Figure 8 compares the theoretical curve with the experimental data introduced in the CONRAD calculations.

Uncertainties and correlations reported in Table 5 provide realistic uncertainties on the scattering cross section. At the thermal neutron energy (25.3 meV), the relative uncertainty reaches approximately 3.3%. Beyond 1 eV, the uncertainty, mainly driven by the relative uncertainty of the bound scattering cross section of hydrogen, is close to 0.9%.

The spurious structures seen between 1 and 5 eV might be originated from the transition to the short collision time approximation used in LEAPR to calculate the TSL.

### 7.3 Covariance matrix of the average cosine $\bar{\alpha}$ of the scattering angle

The right-hand plot of Figure 7 shows the relative uncertainties and the correlation matrix of the average cosine of the scattering angle at 294 K. At the thermal energy, the relative uncertainty is approximately 12%.

### Table 3. Uncertainties on the nuisance parameters (sample area density, normalization factor, background correction, temperature) for each experimental data introduced in the CONRAD calculations.

| Parameter | Zaitsev et al. [16] | Heinloth [17] | Herdade [18] | Dritsa [19] | Beyster et al. [15] |
|-----------|---------------------|---------------|---------------|-------------|---------------------|
| $n(\alpha)/b$ | – | 0.00335 ± 0.00008 | 0.00834 ± 0.00025 | 0.02438 ± 0.00007 | – |
| $N$ | 1.0 ± 0.045 | 1.0 ± 0.01 | 1.0 ± 0.01 | 1.0 ± 0.01 | 1.0 ± 0.05 |
| $B$ | – | ±0.001 | ±0.001 | ±0.001 | ±0.005 |
| $T(K)$ | 294 ± 5 | 294 ± 5 | 294 ± 5 | 294 ± 5 | 294 ± 5 |

### Table 4. Relative uncertainties and correlation matrix between the CAB model parameters after the fitting procedure.

| Parameter | Value | Relative uncertainties | Correlation matrix |
|-----------|-------|------------------------|--------------------|
| $\sigma_0$ (kJ/mol) | 0.31644 | 0.6% | 100 –17 25 63 –31 –14 19 –25 19 |
| $\varepsilon_0$ (nm) | 0.7749 | 0.8% | 100 –33 –3 –10 –16 1 –26 –15 |
| $q_H$ (e$^-$) | 0.5564 | 0.7% | 100 –51 –18 –13 12 –25 15 |
| $d_{OH}$ (nm) | 0.09419 | 0.7% | 100 5 –14 –16 –5 5 |
| $D_{OH}$ (kJ/mol) | 432.581 | 0.8% | 100 –31 15 –22 –2 |
| $\beta_{OH}$ (1/nm) | 22.87 | 0.7% | 100 –7 –24 4 |
| $\theta_{OH}$ (°) | 107.4 | 0.9% | 100 –4 –9 |
| $k_o$ (kJ/mol/rad$^2$) | 367.81 | 0.8% | |
| $d_{OM}$ (nm) | 0.13288 | 0.7% | |

### Table 5. Relative uncertainties and correlation matrix between the CAB model parameters after the marginalization.

| Parameter | Value | Relative uncertainties | Correlation matrix |
|-----------|-------|------------------------|--------------------|
| $\sigma_0$ (nm) | 0.31644 | 2.3% | 100 –77 93 69 33 –18 –64 83 –14 |
| $\varepsilon_0$ (kJ/mol) | 0.7749 | 14.6% | 100 –71 –98 –85 53 97 –54 –32 |
| $q_H$ (e$^-$) | 0.5564 | 3.2% | 100 59 28 –2 –60 81 –18 |
| $d_{OH}$ (nm) | 0.09419 | 6.3% | 100 89 –63 –96 44 38 |
| $D_{OH}$ (kJ/mol) | 432.581 | 6.2% | 100 –63 –88 6 57 |
| $\beta_{OH}$ (1/nm) | 22.87 | 4.2% | 100 51 –11 –28 |
| $\theta_{OH}$ (°) | 107.4 | 6.4% | 100 –45 –9 |
| $k_o$ (kJ/mol/rad$^2$) | 367.81 | 3.8% | |
| $d_{OM}$ (nm) | 0.13288 | 2.7% | |

In both cases the relative uncertainties on the $S_{ij}(\alpha_{ij}, \beta_0)$ function range between 10% in the peak of the distribution up to approximately 30% in the wings.
The bottom plot of Figure 8 compares the calculated \( \bar{m} \) with the data used in the CONRAD analysis. The obtained uncertainties bands overlap the data over the full energy range.

7.4 Propagation to integral calculations

One of the main goals of the present work is to quantify the uncertainty due to the TSL of \(^1\text{H}\) in \(\text{H}_2\text{O}\) in integral calculations. The performances of our covariance matrix between the CAB model parameters was investigated on the MISTRAL-1 and MISTRAL-2 configurations carried out in the EOLE critical facility of CEA Cadarache (France).

7.4.1 The MISTRAL experimental program

A detailed description of the experiments can be found in reference [28]. The reactivity excess was measured at “cold” reactor conditions, from 10 to 80°C.

The MISTRAL-1 configuration is an UO\(_2\) core (3.7\% enriched in \(^{235}\text{U}\)), while the MISTRAL-2 configuration is a MOX core (7.0\% enriched in Am-PuO\(_2\)). Examples of radial cross section of the cores are shown in Figure 9. In the first case the criticality is reached by adjusting the boron concentration in the moderator. In the second case, the critical size of the core was adequately modified (8.7\% fuel pins enriched in Am-PuO\(_2\)).

7.4.2 Propagation of the CAB model uncertainties to the MISTRAL calculations

The Monte-Carlo code TRIPOLI\(^{\circledR}\) [29] was used to calculate the reactivity of MISTRAL-1 and -2, as a function of the temperature [30].

When the TSL of the CAB model is introduced in the JEFF-3.1.1 library [31], the differences \(\Delta \rho\) between the calculated and experimental reactivities for MISTRAL-1 (UOX core) at 20 and 80°C are close to 300 pcm:

\[
\Delta \rho(20^\circ\text{C}) = 283 \pm 71 \text{ pcm},
\]
\[
\Delta \rho(80^\circ\text{C}) = 286 \pm 155 \text{ pcm}.
\]

For MISTRAL-2 (MOX core), they reaches 900 pcm:

\[
\Delta \rho(20^\circ\text{C}) = 900 \pm 110 \text{ pcm},
\]
\[
\Delta \rho(80^\circ\text{C}) = 869 \pm 203 \text{ pcm}.
\]

The large discrepancies observed for the MOX core are due to the contribution of the \(^{241}\text{Am}\) capture cross section, which is significantly underestimated in the JEFF-3.1.1 library.

The quoted uncertainties account for the statistical uncertainties due to the Monte-Carlo simulations (±25 pcm) and the uncertainty due to the TSL of \(^1\text{H}\) in \(\text{H}_2\text{O}\) (Tab. 5). The latter contribution was determined by a direct perturbation of the CAB model parameters.
Fig. 6. Relative uncertainties and correlation matrix of the $\mathcal{S}(\alpha, \beta_0)$ functions for $\beta = 1.0$ (left-hand plot) and $\beta = 10.0$ (right-hand plot) calculated with the CAB model at 294 K with the uncertainties reported in Table 5.

Fig. 7. Relative uncertainties and correlation matrix of the $^1\text{H}$ in H$_2$O scattering cross section (left-hand plot) and of the average cosine $\mu$ of the scattering angle (right-hand plot) calculated with the CAB model at 294 K with the uncertainties reported in Table 5.
At room temperature, the low uncertainty of 71 pcm indicates that the uncertainty on the TSL of light water coming from the CAB model could become a negligible contribution in many UOX configurations. This assumption is confirmed by the results reported in Table 6. For a standard UOX cell, it appears that the uncertainty on the capture cross section of hydrogen (±150 pcm) is even more important than the contribution due to the scattering process.

However, the present results confirms the higher sensitivity of the MOX cores to the TSL of light water. This trend is due to the large resonances in the cross sections of the Pu isotopes. In that case, the uncertainty of 110 pcm obtained at room temperature is no longer negligible. This is also confirmed in Table 7 by comparing the various contributions to the final uncertainty on the reactivity calculated for a MOX cell.

**Fig. 8.** Comparison of the theoretical scattering cross section (top plot) and of the average cosine $\langle \cos \theta \rangle$ of the scattering angle (bottom plot) with the experimental data introduced in the CONRAD calculations.
8 Conclusions

The present work presents the methodology for generating the covariance matrix between the CAB model parameters, which describes the neutron scattering with the hydrogen bounded to the light water molecule. The covariance matrix has been calculated by using the generalized least-square and marginalization algorithms implemented in the CONRAD code.

The obtained uncertainties were propagated to produce covariance matrices for the thermal scattering function. A multigroup treatment on the momentum transfer was adopted to handle the large amount of data contained in the $S(\alpha, \beta)$ function.

Covariance matrices for the $^1H$ in $H_2O$ scattering cross section and for the average cosine of the scattering angle were also produced. The calculated uncertainty bands in both cases overlap the experimental data selected for the

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**Table 6.** Example of uncertainties on the reactivity (UOX configuration at room temperature) in pcm due to the nuclear data. The contribution of $^1H$ in $H_2O$ comes from the present work. The other contributions were calculated with the covariance data base COMAC [32] developed at the CEA of Cadarache.

| Isotopes   | (n,f) | Capture | (n,n) | (n,n') | (n,xn) | $v_{tot}$ | $X_{fast}$ | $X_{th}$ | Total |
|------------|-------|---------|-------|--------|--------|-----------|------------|----------|-------|
| $^1H$ in $H_2O$ | 150   | 71      |       |        |        |           |            |          | 166   |
| $^{10}B$   | 26    |         |       |        |        |           |            |          | 26    |
| $^{16}O$   | 97    | 14      | 2     |        |        |           |            |          | 98    |
| $^{90}Zr$  | 11    | 72      | 4     |        |        |           |            |          | 72    |
| $^{91}Zr$  | 27    | 30      | 2     |        |        |           |            |          | 40    |
| $^{92}Zr$  | 27    | 20      | 2     |        |        |           |            |          | 33    |
| $^{94}Zr$  | 2     | 8       | 2     |        |        |           |            |          | 8     |
| $^{96}Zr$  | 2     | 6       |       |        |        |           |            |          | 6     |
| $^{234}U$  | 1     | 6       | 2     |        |        |           |            |          | 6     |
| $^{235}U$  | 104   | 174     | 13    |        |        |           | 276        | 142      | 371   |
| $^{236}U$  | 1     |         |       |        |        |           |            |          | 1     |
| $^{238}U$  | 29    | 165     | 83    | 38     | 18     | 32        | 9          | 142      | 195   |
| Total      | 108   | 303     | 137   | 39     | 18     | 277       | 9          | 142      | 470   |

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Fig. 9. Radial cross sections of the MISTRAL-1 core (left-hand plot) and the MISTRAL-2 core (right-hand plot) at $T=20^\circ C$. 

![Radial cross sections](image-url)
The contribution of the uncertainty due to the $^1$H in $\text{H}_2\text{O}$ thermal scattering data was then evaluated on the MISTRAL-1 (UOX) and MISTRAL-2 (MOX) integral experiments carried out in the EOLE facility of CEA Cadarache. The calculated uncertainty at 20°C reaches ±71 pcm for the MISTRAL-1 core. At 80°C, the uncertainty is almost twice with respect to room temperature. The same trend was found for the MISTRAL-2 configuration, where the uncertainty on the reactivity is ±110 pcm at 20°C. The present results highlight the quality of the CAB model for calculating the TSL of light water at room temperature. For UOX configurations, we can expect a negligible contribution on the final uncertainty in nuclear criticality and safety studies.

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**Author contribution statement**

Parameters of the Molecular Dynamic simulation were established by J.I Marquez Damian with the GROMACS code. The determination of the covariance matrix between the GROMACS parameters and the propagation of the uncertainties were performed by J.P. Scotta and G. Noguere by using the CONRAD code.

### Table 7. Example of uncertainties on the reactivity (MOX configuration at room temperature) in pcm due to the nuclear data. The contribution of $^1$H in $\text{H}_2\text{O}$ comes from the present work. The other contributions were calculated with the covariance data base COMAC [32] developed at the CEA of Cadarache.

| Isotopes     | (n,f) | Capture | (n,n) | (n,n') | (n,xn) | $\nu_{tot}$ | $\chi_{fast}$ | $\chi_{th}$ | Total |
|--------------|-------|---------|-------|--------|--------|-------------|--------------|------------|-------|
| $^1$H in $\text{H}_2\text{O}$ | 46    | 110     |       |        |        |             |              |            | 119   |
| $^{10}$B     |       |         |       |        |        |             |              |            | 8     |
| $^{16}$O     | 114   | 24      | 4     |        |        |             |              |            | 117   |
| $^{90}$Zr    | 11    | 24      | 7     |        |        |             |              |            | 27    |
| $^{91}$Zr    | 13    | 16      | 4     |        |        |             |              |            | 21    |
| $^{92}$Zr    | 8     | 22      | 4     |        |        |             |              |            | 24    |
| $^{94}$Zr    | 2     | 59      | 3     |        |        |             |              |            | 59    |
| $^{96}$Zr    | 2     | 13      | 1     |        |        |             |              |            | 14    |
| $^{235}$U    | 2     | 6       | 3     | 1      |        |             |              |            | 9     |
| $^{238}$U    | 114   | 88      | 80    | -60    | 25     | 35          | 12           | 4          | 160   |
| $^{239}$Pu   | 1     | 70      | -20   | 1      |        |             |              |            | 67    |
| $^{240}$Pu   | 278   | 371     | 26    | 5      |        |             |              |            | 484   |
| $^{241}$Pu   | 42    | 178     | -16   | -5     | 1      | 2           | 9            |            | 182   |
| $^{242}$Pu   | 108   | 96      | 8     |        |        | 88          | 58           |            | 179   |
| $^{241}$Am   | 3     | 131     | 10    | 2      |        | 2           | 1            |            | 131   |
| Total        | 322   | 475     | 156   | -59    | 25     | 111         | 60           | 126        | 619   |

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