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To cite this version:

M. Tiphine, C. Carmouze, G. Nogueres, F. Cantargi, J.-I. Márquez Dámian. Effect and uncertainties of H in Ice thermal scattering laws on the neutron multiplication factor for PWR fuel criticality applications. ICNC 2019 - 11th International conference on Nuclear Criticality Safety, IRSN, Sep 2019, Paris, France. cea-02972089

HAL Id: cea-02972089
https://hal-cea.archives-ouvertes.fr/cea-02972089
Submitted on 20 Oct 2020

EFFECT AND UNCERTAINTIES OF H IN ICE THERMAL SCATTERING LAWS ON THE NEUTRON MULTIPLICATION FACTOR FOR PWR FUEL CRITICALITY APPLICATIONS

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ABSTRACT

In the context of the IAEA recommendation to ensure the transportation of fuel assemblies between 233 K and 311 K, thermal scattering laws of hydrogen in iced water have been produced with the LEAPR module of the NJOY code and included in the JEFF-3.3 nuclear data evaluation. Following this work, a benchmark was launched by the OECD/NEA Working Party on Nuclear Criticality-Safety subgroup-3 to evaluate the effect of the temperature on a PWR assembly criticality. This paper first focuses on the results obtained on this benchmark by CEA with the TRIPOLI-4® Monte-Carlo code. They show that, in terms of criticality-safety, computations made at 293 K are conservative and that the impact of density on the k_{eff} *is much stronger than the nature of the hydrogen bound or the adjustment of nuclear data to temperature. To go further, the uncertainties associated with the thermal scattering laws of hydrogen in iced water have been evaluated and propagated on one of the benchmark cases. The reference method to do so consists in a direct propagation of the LEAPR model parameters uncertainties. Another method, based on covariance matrix of the hydrogen in iced water scattering cross section, was also used in order to evaluate its relevance. The direct propagation leads to an uncertainty of 111 pcm. The uncertainty evaluated with the second method is lower by around 50 pcm. Whatever the method considered, those uncertainties remain acceptable in the criticality-safety context especially as the effect of the temperature on the* k_{eff} *and the impact of the hydrogen bound nature are both low regarding density effects.*

KEY WORDS

Thermal Scattering Laws, Criticality-Safety, Ice

1. INTRODUCTION

Although variations of a system temperature may affect both its physical and neutronic parameters, criticality-safety studies are often performed at 294 K, results obtained at this temperature being considered as conservative regarding standard reactor conditions. Following the IAEA recommendation [1] to ensure the transportation of fuel assemblies between 233 K and 311 K, the question of the temperature impact on criticality-safety evaluations has been raised.

In that context, thermal scattering laws of hydrogen in iced water (H in Ice) were produced and, following this work, a benchmark was launched by the OECD NEA Working Party on Nuclear Criticality Safety (WPNCS) subgroup-3 to evaluate the effect of temperature on the neutron multiplication factor for PWR fuel assemblies [2].

This paper focuses on the results obtained by CEA on the basis of this benchmark. To go further, the uncertainties associated to the thermal scattering laws of H in Ice have been evaluated and propagated on one of the benchmark cases. Two methods were used to do so, one consisting in a direct propagation of the LEAPR model parameters uncertainties and the other one based on covariance matrix of the scattering cross section of H in Ice.

2. CALCULATION TOOLS AND MODEL

a. Hydrogen scattering cross sections

The scattering of fast and thermal neutrons is illustrated on Figure 1. For fast neutrons, the kinetic energy prevails on the hydrogen binding energy, so that the hydrogen bound can be neglected in high energy range and $\sigma(H_2O) = 2\sigma(H) + \sigma(O)$. On the contrary, the kinetic energy of thermal neutron is equal or lower than the hydrogen binding energy and the hydrogen bound has to be taken into account.

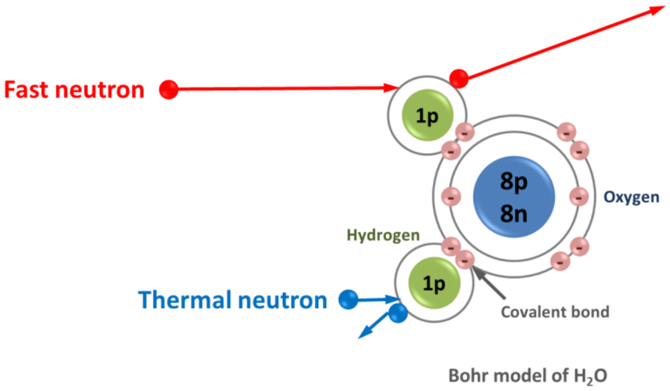

Figure 1. Scattering of neutrons

Figure 2 compares the scattering cross sections of H in Ice and H in H_2O along with the hydrogen scattering cross section in the free gas approximation.

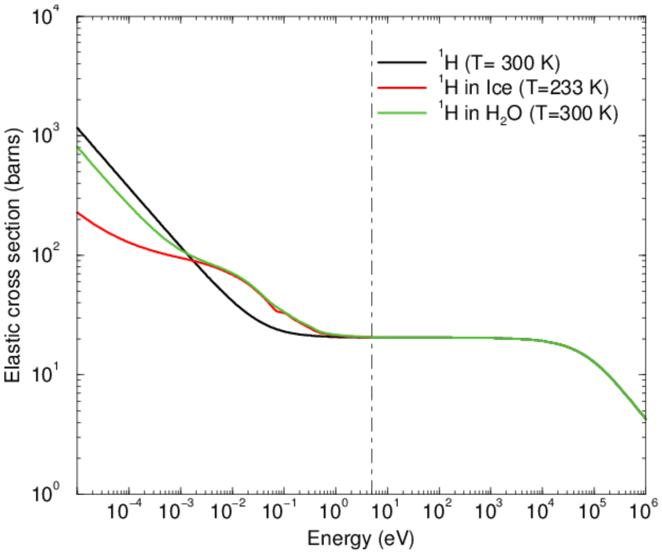

Figure 2. Scattering cross sections of H in Ice, H in H2O and H in the free gas approximation

This figure highlights the importance of the hydrogen bounds in the low energy range (below 4 eV). Indeed, the free hydrogen scattering cross section being lower than the two others between 10^{-3} and 1 eV, it is not necessarily conservative to use it for criticality-safety studies. For energies higher than 5 eV, the impact of the hydrogen bounds does not prevail and the free gas approximation can be used for computations involving H_2O.

b. Thermal Scattering Laws for H in Ice

In the low energy range (below 4 eV), the neutron scattering in iced water is affected by the hydrogen bounds. Thermal Scattering Laws (TSL also referred to as $S(\alpha,\beta)$) contain the dynamic and structural information on this hydrogen bound that are required to take into account the modifications on the energy and angular distribution of scattered neutrons. In this function, α represents the momentum transfer and β the energy transfer.

The TSL of H in Ice are calculated with the LEAPR module of the NJOY code [3], in which the key parameter is the frequency spectrum $\rho(\beta)$. The frequency spectrum of H in Ice can be decomposed into three components:

$$\rho(\beta) = w_c\rho_C(\beta) + w_1\delta(\beta_{E_1}) + w_2\delta(\beta_{E_2}) \tag{1}$$

The term $\rho_C(\beta)$ is a continuous distribution describing the rotational mode of the molecule and $\delta(\beta_{Ei})$ (for i=1,2) are two discrete oscillators used to define the intramolecular vibrations. w_c and w_i are the weights associated to each contribution. It is noteworthy that:

$$w_c + w_1 + w_2 = 1 \tag{2}$$

c. Definition of the LEAPR model parameters

The values of the LEAPR parameters at 233 K used for this study are reported in Table I.

Table I. LEAPR parameters for H in Ice at 233 K

Parameters		T=233 K
Energy interval (meV)	δ	1.0
First oscillator energy (meV)	E_1	205.0
Second oscillator energy (meV)	E_2	391.0
Continuous spectrum weight	w_c	0.5
First oscillator weight	w_1	0.1667
Second oscillator weight	w_2	0.3333
H free-atom scattering cross section (barn)	σ_s	20.44

The parameter δ is the energy interval on which the phonon distribution is reconstructed, $E_i$are the energies of the discrete oscillators and, w_c and w_i are the weights associated to each spectrum contribution (see Eqs. 1-2). Their values come from the JEFF-3.3 evaluation for H in Ice provided by the nuclear data group of Centro Atómico Bariloche. Finally, σ_s, the free scattering cross section of hydrogen evaluated at 0 K, is a fixed parameter whose value has been reviewed in previous works [4].

d. Description of the WPNCS subgroup-3 benchmark

To understand the k_{eff} variation associated with temperature dependent criticality calculations for PWR assembly, a benchmark has been proposed by the WPNCS subgroup-3. It focuses mainly on temperatures below 293 K as the PWR k_{eff} evolution for higher temperatures is well known. It considers two cases representative of storage configurations that can be encountered in criticality-safety studies: a 17x17 PWR-type fuel assembly surrounded by a 1 m thick water reflector and an infinite array of PWR fuel assemblies (Figure 3).

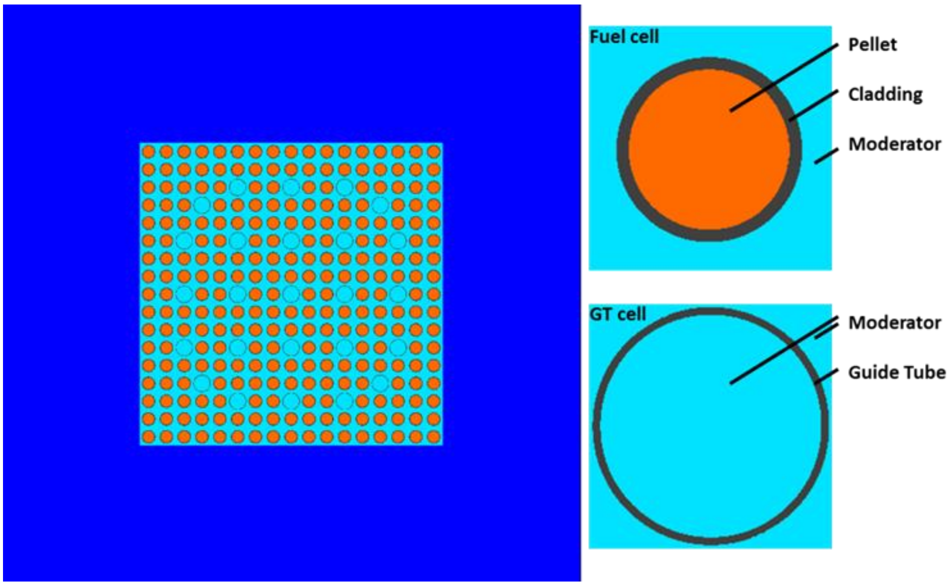

Figure 3. TRIPOLI-4® modeling of the benchmark PWR assembly

For both cases, guide tubes (GT) cells are modelled and the UO2 fresh fuel enrichment is 4.5 wt%. Along with the fresh fuel, two used fuel cases are also considered, 45 GWd/t as a representative burnup and 30 GWd/t as an intermediate case. While the reflector is kept at room temperature (293 K), the neutron multiplication factor (k_{eff}) is calculated at a fuel, moderator and cladding tube temperature of 233 K, 253 K, 293 K, 333 K and 588 K. For each temperature, the water density in moderator is adjusted (see Table II). The hydrogen bound data depends on the moderator temperature: H in Ice at 233 K and 253 K and H in liquid water at 293 K and above.

Table II. Specification for the water density in moderator

Material	Ice 1	Ice 2	Water - room temperature	Water - elevated temperature	Water - elevated temperature and pressure
Temperature (K)	233	253	293	333	588
Density (g.cm^{-3})	0.9228	0.9208	0.9980	0.9830	0.6940
Hydrogen bound	ice	ice	liquid	liquid	Liquid

The CEA calculations are performed with the continuous energy Monte-Carlo code TRIPOLI-4® [5], which is part of the criticality-safety calculation package CRISTAL V2 [6]. The JEFF-3.3 nuclear data evaluation [7] is used and data are processed with GALILEE V0-3.2 [8], based on NJOY [3], for cross section computations and CALENDF [9] to generate probability tables.

3. EFFECT OF TEMPERATURE ON THE NEUTRON MULTIPLICATION FACTOR FOR PWR FUEL CRITICALITY APPLICATIONS

The results obtained by CEA on the WPNCS subgroup-3 benchmark are represented on Figure 4. Values of k_{eff} evaluated at 293 K (d=0.998 g.cm^{-3}) are given in Table III. For each burn-up, at a temperature T:

$$\Delta k_{eff}(T) = (k_{eff}(T) - k_{eff}^{293K})/k_{eff}^{293K} \tag{3}$$

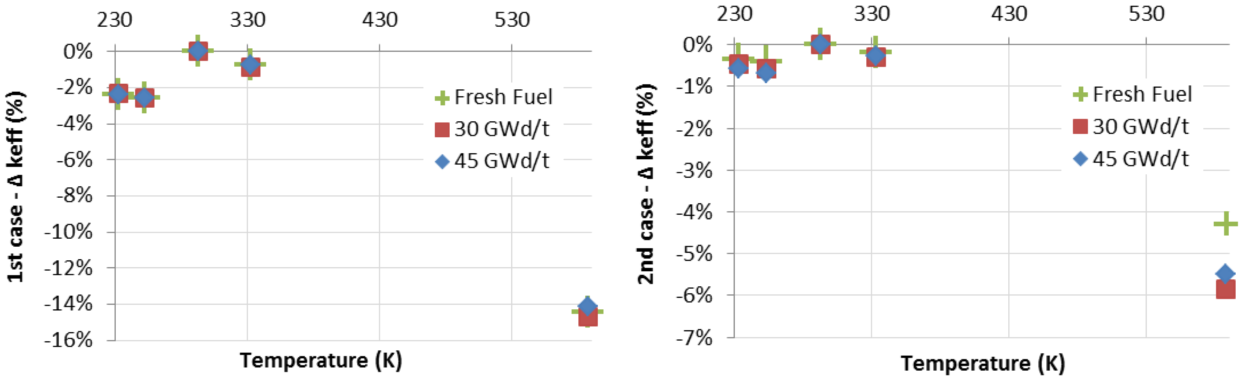

Figure 4. Impact of temperature on the k_{eff} for cases 1 (single unit, left) and 2 (infinite array, right) with moderator density adjusted to the temperature (1σ = 0.02%)

Table III. Benchmark results: k_{eff} ±1σ (standard Monte-Carlo deviation) at T=293 K d=0.9980 g.cm^{-3}

Burn-up	Case 1 - Single Unit	Case 2 - Infinite Array
0 GWd/t	0.93056 ± 0.015%	1.48734 ± 0.012%
30 GWd/t	0.77577 ± 0.018%	1.23261 ± 0.013%
45 GWd/t	0.70653 ± 0.020%	1.12461 ± 0.013%

When the moderator density is adjusted to its change in temperature, temperatures of 253 K and 233 K lead to a decrease of the k_{eff} of around -2.5% in the single unit case and -0.5% in the infinite array case in comparison with the 293 K computations. It is noteworthy that this decrease of the k_{eff} indicates that the 293 K case remains conservative in terms of criticality-safety.

To go further, the effects of water density and of the adjustment of nuclear data to temperature are represented separately on Figure 5 and Figure 6 for the single unit case. On Figure 5, the temperature is fixed and the moderator density varies from 0.6940 g.cm^{-3} (density considered at 588 K in the benchmark) to 0.9980 g.cm^{-3} (density considered at 293 K in the benchmark). On Figure 6, the density is fixed and temperature varies from 233 K to 588 K.

When the temperature is kept constant (Figure 5), in comparison with the standard density case (0.998 g.cm^{3}), computations with the density of ice leads to a decrease of the k_{eff} of about -2.9%. With a constant density (Figure 6), compared to results obtained at 293 K, computations at 253 K and 233 K leads to a decrease of the k_{eff} of about 0.4%. Thus, the effect of density appears to prevail on the effects of the adjustement of nuclear data to temperature.

Furthermore, the impact of the nature of the hydrogen bound can be roughly evaluated from Figure 6, considering that at 293 K and above (where the liquid water hydrogen bound is considered) the effect of temperature on the k_{eff} is linear. This makes the estimation of a k_{eff} at 233 K with a liquid water hydrogen bound possible. For each density considered the difference between the k_{eff} estimated with H in H_2O and the one calculated with H in Ice is always lower that 0.1%. The impact of the nature of the hydrogen bound appears to be much less important than the impact of density or temperature. This prevalence of density effect over temperature and nuclear data effects is consistent with previous works on the H in Ice TSL impacts on PWR assembly [10].

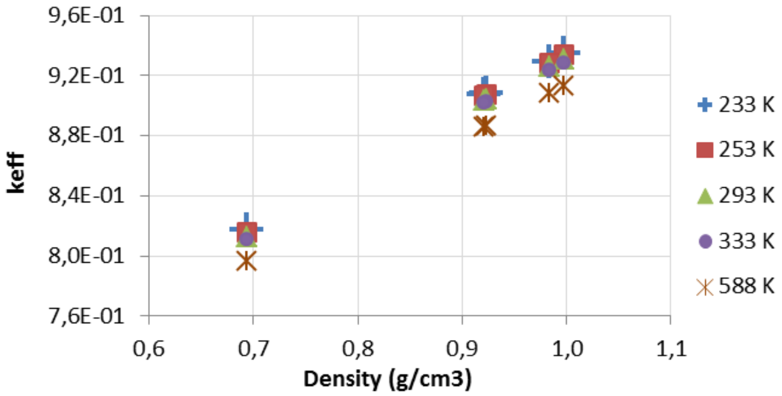

Figure 5. Impact of density on k_{eff} for the single unit case

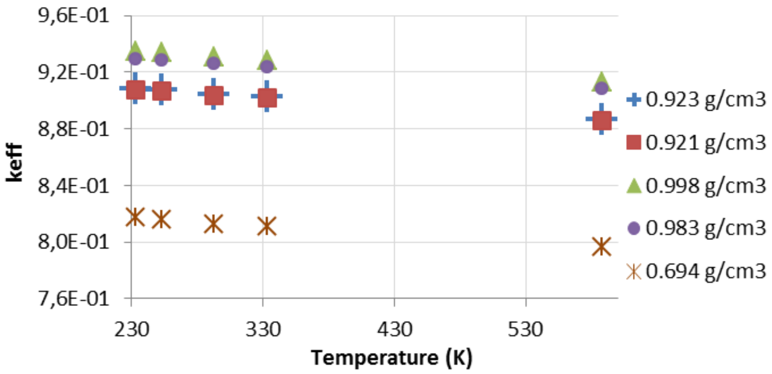

Figure 6. Impact of temperature on k_{eff} for the single unit case

4. PRODUCTION AND PROPAGATION OF H IN ICE TSL UNCERTAINTIES

The propagation of the uncertainties of H in Ice TSL is conducted on one case of the WPNCS subgroup-3 benchmark: a fresh PWR assembly surrounded by a 1m water reflector with fuel, guide tubes and moderator at 233 K.

To compare their relevance, two methods are used to do so. The first one consists in a direct propagation of the LEAPR parameters uncertainties. The other one uses uncertainty/sensitivity analyses and requires the covariance matrix of the scattering cross section of H in Ice. Although the first method is considered as the reference one, the second one is easier to execute as long as the covariance data are available.

a. Direct propagation of the LEAPR parameters uncertainties

This method is considered here as the reference method and requires a high expertise level to define the perturbations to apply on the LEAPR model parameters.

Definition of the LEAPR parameters uncertainties

The LEAPR parameters that are perturbed in this study, along with their uncertainty, are summarized in Table IV. The uncertainties on the LEAPR parameters of interest for this work have been determined using an approach previously used for light water [11]. Each variance has been optimized thanks to transmission data of ice measured at the LINAC facility of Bariloche. The present uncertainties are still preliminary and should be improved. The free hydrogen scattering cross section σ_s is a fixed model parameter whose value and uncertainty have been reviewed in previous works [4].

For simplicity, the uncertainty on the phonon spectrum is taken into account with a scaling factor Δ. This factor is applied to the vibration energy grid e_k used to reconstruct the phonon distribution:

$$e_k = \Delta k \delta \tag{4}$$

where δ is the energy interval given in the LEAPR input file (see Table I). The uncertainty on the scaling factor is evaluated so that the theoretical calculations are in reasonable agreement with the experimental values.

Table IV. LEAPR parameters for H in Ice at 233 K

Parameters		Values	Uncertainty
Scaling factor	Δ	1.000	10%
First oscillator energy (meV)	E_1	205.0	2.9%
Second oscillator energy (meV)	E_2	391.0	10.2%
Continuous spectrum weight	w_c	0.5	0.5%
H free scattering cross section (barn)	σ_s	20.44	0.2%

Direct propagation of the LEAPR parameters uncertainties

The direct perturbation of the LEAPR parameters uncertainties makes possible to investigate the sensitivity of the calculated k_{eff} to each parameter. Results are summarized in Table V.

Table V. Uncertainties Δk_{eff} for each LEAPR parameter for a convergence criterion of 1 pcm

LEAPR parameter	Δk_{eff} (pcm)
$\Delta + w_c$	-111
E_1	-5
E_2	-3
σ_s	+5

In the present work, the LEAPR parameters are assumed independent. It is then considered that all the contributions to the calculated k_{eff} are also independent. The total uncertainty is thus evaluated with a quadratic sum of all the individual contributions. All parameters being considered, the final uncertainty on the calculated k_{eff} is of 111 pcm.

The decomposition of each contribution shows that the scaling factor Δ and the continuous spectrum weight are the most sensitive parameters. The intra-molecular vibrations (E_1 and E_2) and the free hydrogen scattering cross section have negligible contributions to the k_{eff} uncertainty.

b. Propagation of H in Ice scattering cross section covariances

The uncertainties of H in Ice scattering cross sections are propagated through the criticality calculations of the PWR assembly configuration by using the sandwich formula:

$$\varepsilon^2 = {}^{t}S D_{\sigma} S \tag{5}$$

S is the sensitivity coefficients vector of the criticality configuration k_{eff} to the uncertain parameters, H in Ice scattering cross section in this case ;

${}^{t}S$ is the transpose of S ;

D_{σ} is the H in Ice scattering cross section covariance matrix. The diagonal elements are the square of the relative cross section uncertainties. The non-diagonal elements are the product of the cross section uncertainties by the cross section correlations between the energy groups (Figure 7). The covariance matrix is condensed on a 26 energy groups structure as well as the k_{eff} sensitivity coefficients vector.

The description and calculation of those parameters are described below.

Production of correlations for the covariance matrix for H in Ice scattering cross section

The analytical method applied to generate the correlation matrix for the H in Ice scattering cross section relies on the CONRAD code [12], in which mathematical algorithms were implemented to account for uncertainties of various origins.

Figure 7 shows the relative uncertainties and the correlation matrix of H in Ice scattering cross section after the uncertainty propagation of the LEAPR model parameters at 233 K. The relative uncertainty below 10 meV is underestimated. Such an issue will be solved by introducing a defect model parameter [11].

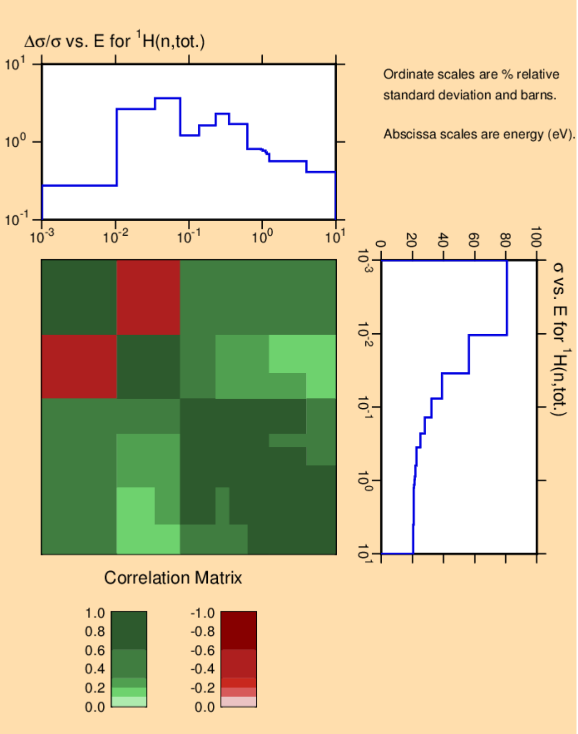

Figure 7. Relative uncertainties and correlation matrix for the neutron scattering cross section of H in Ice at 233 K (represented with 26 energy groups)

Propagation of the H in Ice scattering cross section uncertainties on the k_{eff}

The k_{eff} sensitivity to the H in Ice scattering cross section is evaluated with the iteration fission probability (IFP) method available in the TRIPOLI-4.11 Monte-Carlo code. Briefly, the IFP is a method used to compute the adjoint neutron flux based on the importance of a neutron regarding the average amount of its descendants [13]. The k_{eff} sensitivity profile is shown on Figure 8. It highlights the limited impact of the scattering cross section of H in Ice on the reactivity of the PWR assembly configuration, with a maximal value of 17 pcm/% around 4 eV.

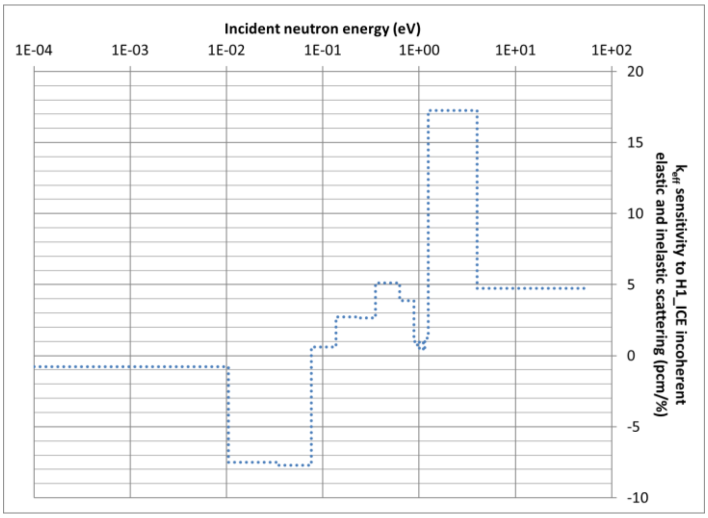

Figure 8. k_{eff} sensitivity profile to the H in Ice scattering cross sections

The limited effect of the H in Ice scattering cross section combined with relatively small uncertainties and values leads to a small propagated uncertainty of the k_{eff} of around 47 pcm.

c. Discussion of the results

Two methods have been used to evaluate and propagate the uncertainty associated to the TSL of H in Ice. The direct propagation of the LEAPR model parameters uncertainties, considered as the reference method, leads to an uncertainty of 111 pcm whereas the method using covariance matrix of the scattering cross section of H in Ice leads to an uncertainty of around 47 pcm. This seems to indicate that the direct propagation of the LEAPR model parameters uncertainties is the only reliable method available. The other method, despite being easier to implement, can only be used to provide orders of magnitude.

Whatever the method considered, those uncertainties remain low in the criticality-safety context. This conclusion is enhanced by the low variation of the k_{eff} with temperature and the low impact of the hydrogen bound nature shown in paragraph 3. Once again, it should be noted that the k_{eff} evaluated at 293 K is conservative for criticality-safety studies.

5. CONCLUSION

This paper follows the IAEA recommendation to ensure the transportation of fuel assemblies between 233 K and 311 K, and the addition of thermal scattering laws of H in Ice in JEFF-3.3. It is also part of a benchmark proposed by the OECD WPNCS subgroup-3.

Two cases representative of fuel storage configurations encountered in the criticality-safety context were studied at different temperatures in order to evaluate the effect of low temperatures on the PWR assembly k_{eff}. The first configuration considers a single assembly surrounded by a 1 m thick water reflector and the other an infinite array of assemblies. At 233 K and 253 K, the H in Ice bound was taken into account, whereas the H in H_2O bound was considered at 293 K and above. For each temperature, the density of the water in the moderator was adjusted. The study of the k_{eff} evolution with changes in density and in the nuclear data temperature shows that the density effect prevails on the temperature effect and on the nature of the hydrogen bounds. It also highlights that the results obtained at 293 K are conservative in a criticality-safety context as the k_{eff} of the PWR assembly decreases when lower temperatures are considered.

The uncertainty associated to the H in Ice TSL have been evaluated and propagated on one case of the benchmark. The reference method to do so consists in a direct propagation of the LEAPR model parameters uncertainties and leads to an uncertainty of 111 pcm. Another method, easier to implement and based on covariance matrix of the H in Ice scattering cross section, was also used in order to evaluate its relevance. It leads to an uncertainty of around 47 pcm. This indicates that, although this method can provide orders of magnitude, the direct propagation of the LEAPR model parameters uncertainties is the only reliable method available.

Finally, in the criticality-safety context, those uncertainties remain low whatever the method used. This conclusion is strengthened as the effect of the temperature on the k_{eff} and the impact of the hydrogen bound nature are both low regarding density effects.

ACKNOWLEDGMENTS

The authors wish to thank ORANO and EDF for their partial financial support.

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