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Effect of low temperatures on criticality calculation for the transport of fissile material

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ABSTRACT

Fissile package design has to meet requirements of IAEA (International Atomic Energy Agency) regulation n°SSR-6 ([1]) in order to ensure subcriticality in usual normal and accidental conditions of transport. For these packages, these requirements are to be taken into account for temperatures of the package between -40°C (233 K) and +38°C (311 K). However, criticality calculation tools presently used do not allow to perform calculations over the required range of temperature. Criticality calculations are mainly performed at room temperature (~ +20 °C) which is the temperature generally encountered in benchmark experiments used for nuclear data validation. In the past, this hypothesis has not been questioned since the neutron effective multiplication factor (Keff) decreases with temperature for temperature above +20°C and because of the models simplifications generally taken into account. However, since neutron cross-section for low temperatures are available on a theoretical basis, it is possible nowadays to explore if the assumption of using +20°C data is really valid over the whole range of temperature.

This paper shows that a decrease of temperature leads to a decrease of Keff due to the decrease of H_2O density when water is in the solid phase (ice) compared to the liquid phase on the base of two simplified cases considering water as the moderator. On the other hand, considering nuclear data, a decrease of temperature could lead to an increase of reactivity in particular due to the Doppler effect on ^{238}U capture cross section.
Nevertheless, new studies should be performed in order to better understand the global effects of low temperature on nuclear data (for example, with more temperatures and nuclides). In particular, the impact of density variation of polyethylene as moderator must be assessed as its behaviour could be very different from those of water.

INTRODUCTION

IAEA (International Atomic Energy Agency) is responsible for prescribing regulations for safety and protection of persons and the environment. Among those standards, the regulation n°SSR-6 ([1]) stipulates that packages used to transport radioactive materials have to meet specific requirements. For fissile package design, the respect of these requirements ensures subcriticality in normal and accidental conditions of transport. The requirements are to be ensured for an initial temperature of the package between -40°C and +38°C (cf. para. 679 of [**Erreur ! Source du renvoi introuvable.**]). However, criticality calculation tools presently used do not allow to perform calculations over the required range of temperature in routine. Indeed, the nuclear data are mainly generated for positive temperature and validated primarily at room temperature (293.6 K / ~+ 20°C) and sometimes, for specific applications, at higher temperature. This hypothesis has not been questioned since the neutron effective multiplication factor (Keff) decreases when temperature increases for temperature above +20°C (due to the decrease of water density with temperature and Doppler effect…) and because of the model simplifications taken into account that generate margins allowing to compensate the absence of temperature variation. However, since neutron cross-sections for low temperatures are available on a theoretical basis, even if not perfect and not validated over benchmarks, it is now possible to explore if this statement is really valid over the whole range of temperature.

The study in this paper is related to the impact of low temperatures on Keff for two simplified configurations:

- Case n°1: a cylindrical containment vessel loaded with moderated uranium and reflected by water,
- Case n°2: an UO_2 fuel assembly into water and surrounded by boronated resin.

The study will take into consideration the effects of temperature on nuclear data (cross sections, $S(\alpha,\beta)$) and material density. The temperatures studied are 233 K (-40°C), 273 K (0°C) and 293 K (20°C).

1. Methodology used in this study

a. Nuclear data

Evaluated Nuclear Data Files (ENDF) store nuclear data which are used in many applications. These files contain, for example, data on microscopic effective sections which are used to perform criticality calculations. The evaluation of these data is carried out by "expert evaluators" using experiments or models. All these elements are gathered in international files known as ENDF. Each international library has its own data evaluations (JENDL in Japan, CENDL in China, JEFF in Europe or ENDF/B in the USA). Nuclear data evaluations are commonly performed based on nuclear data taken at room temperature or temperatures above. The evaluations have generally been done in response to reactor applications, which in turn, do not require calculations at temperature below room temperatures. In order to obtain results at low temperatures (for example at 233 K), it is therefore necessary to extrapolate the results from existing databases at 293 K. For this study, the JEFF 3.3 library at 293 K is used (European database, 2017). The results obtained with these nuclear data should be taken with caution, as they are not based on experimental measurements but on extrapolation of data at 293 K. Studies or experimental measurements should be carried out to validate these data at low temperatures.

The microscopic cross sections for ^{1}H diffusion and ^{238}U capture at different temperatures are presented in Figures 1 and 2.

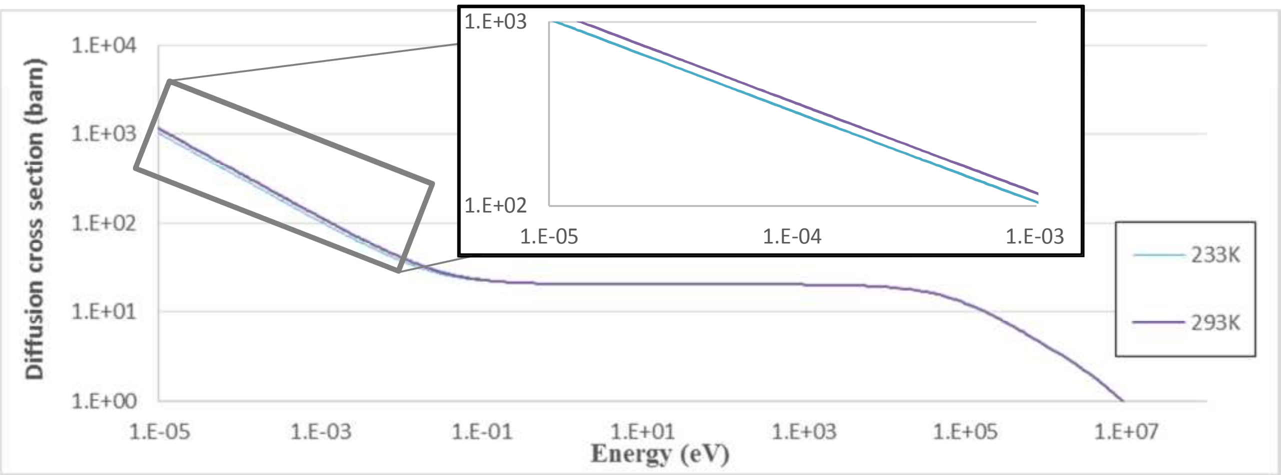


Figure 1 – ^{1}H diffusion cross section

As shown in Figure 1, for the ^{1}H diffusion cross sections, temperature has no impact for fast and epithermal spectra. But, for thermal spectrum, temperature has a visible impact due to the 1/v behavior of the cross section in this neutron energy area.

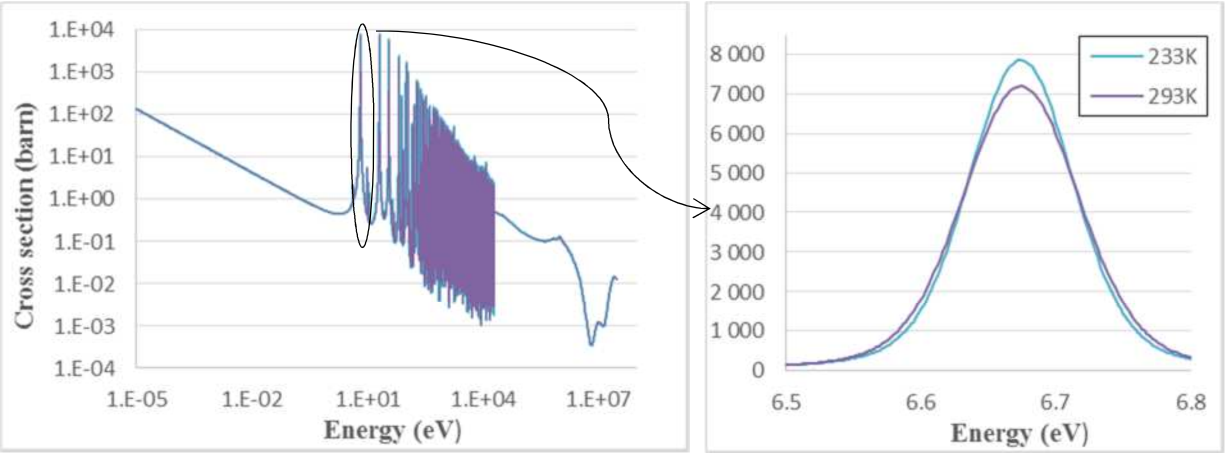


Figure 2 – ^{238}U capture cross section

As shown in Figure 2, for ^{238}U capture cross sections, temperature has no impact outside the resonance range. In the resonance range, the Doppler effect applies, which results in a rise of the peaks and a narrowing of the energy band with a decrease of temperature: the probability that the neutron "jumps" the resonance increases, thus the probability of capture by ^{238}U decreases. This effect is particularly visible at the first resonance of this cross section at 6.68 eV (cf. Figure 2).

Given its high diffusion capacity and low "capturing" power, light water is the most widely used moderator in reactor. In criticality safety, H_2O is commonly considered due to its presence in facilities and laboratories or its potential presence during transport of fissile material. It is therefore important to have reliable effective diffusion cross-section data for criticality safety studies for this material. Indeed, the diffusion reaction, and therefore the associated cross sections, are impacted by the chemical bond (between hydrogen and oxygen atoms). This effect is taken into account by means of the so-called $S(\alpha,\beta)$. $S(\alpha,\beta)$, where α is the momentum and β the energy transfer (unitless quantities), are derived from physical models, allowing to describe low-energy neutron interactions by taking into account molecular binding effects in diffusion processes. The use of $S(\alpha,\beta)$ in calculations significantly modifies the hydrogen diffusion cross sections ([2]). Concerning $S(\alpha,\beta)$ of H-H_2O, when water is in the solid form (ice), the resulting diffusion cross section at low temperatures (233 K and 273 K) are obtained from JEFF 3.3 $S(\alpha,\beta)$ nuclear data at these temperatures. The results obtained with these $S(\alpha,\beta)$ nuclear data at low temperature have to be taken with caution, as they are not based on experimental measurements but only on extrapolation of data at other temperatures. Studies or experimental measurements should be carried out to validate these data at low temperatures.

b. Material density

Evolution of water density between -60°C (213 K) and 100°C (373 K) and evolution of UO_2 density between 0 K and ~ 3000 K are shown in Figure 3 (left and right respectively).

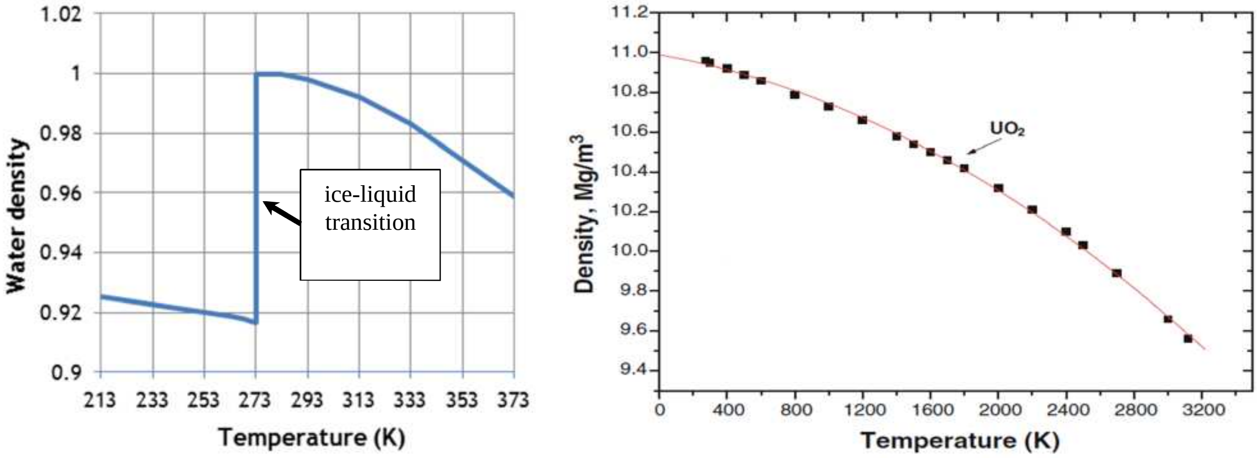


Figure 3 - Density of water (left) and UO$_2$ (right) as a function of temperature

Water density evolution is quite important in particular between the liquid state and the solid state (ice). Concerning UO$_2$, it could be noted that the density variation between 233 K and 293 K is very limited. So, in criticality calculation, this effect is not taken into account and the density at 293 K is considered. Concerning other materials considered in this paper (for example stainless steel or resin), the density evolution in function of temperature is as limited as UO$_2$. Consequently, densities for these materials are also considered at 293 K.

c. Criticality calculation code

Calculations were performed using the French MORET 5 continuous energy Monte Carlo code [3]. In the results, the standard deviation on reactivity is equal to 10 pcm.

2. Case n°1: cylindrical moderated uranium

a. Presentation

This configuration is a cylindrical containment vessel of 12 cm of radius loaded with 13 kg of uranium (enriched at 20% in ^{235}U) moderated by water. The cylinder is reflected either with 2.5 cm of water or 20 cm of water which corresponds to the usual “fixed reflection” considered in criticality safety studies. The uranium mass has been chosen in order to have a Keff near 1 at the optimum moderation for a reflection by 2.5 cm of water and a temperature of 293 K. For this study, the concentration of uranium of 560 g/L, corresponding to the optimum of moderation at 293 K, is used in all calculations in order to have the same geometry size.

Based on calculations performed for this study but not presented in this paper, the optimum of moderation ratio is not modified when using only nuclear data at low temperatures. But, when water is in the solid form (ice), H_2O density is lower than the one for liquid water. This results in a slight decrease of the corresponding uranium optimum concentration at low temperatures from 560 g/L to 520 g/L. However, as the reactivity difference between these 2 concentrations is very limited (78 pcm), it doesn’t affect the conclusions of the following paragraphs.

The Figure 4 presents the studied configuration.

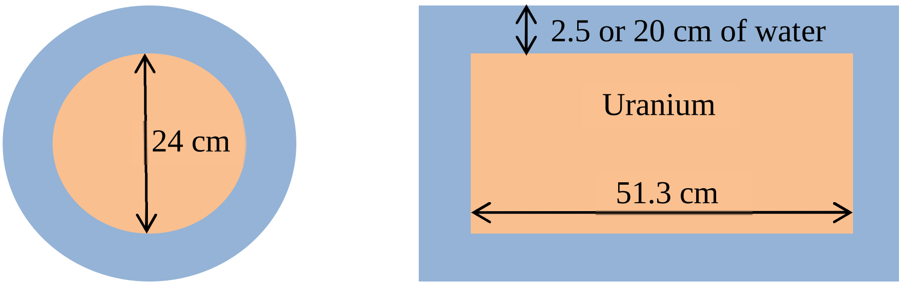


Figure 4 - Case n°1 configuration

Concerning the water reflector, the density is fixed in the calculations and is equal to the one at 293 K in order to separate the effects in this study. However, the nuclear data used for the reflector correspond to the studied temperature.

b. Results

Table 1 presents the reactivity results for the case n°1. When only nuclear data is studied, the water density used is those at 293 K. When only water density is studied, the nuclear data are at 293 K.

Reflector thickness	Temperature	Nuclear data effect	H_2O density effect	Nuclear data & H_2O density combined effect
2.5 cm	**233 K**	1.00296 (+334pcm)	0.95469 (-4493pcm)	0.95764 (-4198pcm)
	273 K (ice)	1.00007 (+45pcm)	0.95091 (-4871pcm)	0.95112 (-4850pcm)
	273 K (liquid)*	*	1.00054 (+92pcm)	*
	293 K	0.99962		
20 cm	**233 K**	1.08047 (-272pcm)	1.04846 (-3473pcm)	1.04514 (-3805pcm)
	273 K (ice)	1.08283 (-36pcm)	1.04559 (-3760pcm)	1.04482 (-3837pcm)
	273 K (liquid)*	*	1.08389 (+70pcm)	*
	293 K	1.08319		

* The S(α,β) for H-H_2O liquid at 273 K have not yet been processed.

Table 1 – Keff (σ = 10 pcm) and discrepancy with the Keff at 293 K for case n°1

Results show that water density has a more significant impact on Keff than nuclear data (~4000-5000 pcm compare to 0-400 pcm for both cases) as it could be expected. Indeed, the transition from liquid water to ice leads to a consequent decrease in density and thus to a decrease in Keff (from 1.00054 to 0.95091 for 2,5 cm reflector and from 1.08389 to 1.04559 for 20 cm reflector). In order to understand these results, the Keff could be expressed by the following formula:

$$K_{eff} = K_{\infty}.P_{NL}.(abs\ FM), \text{ where :}$$

- K_{∞} is the infinite Keff of the fissile medium (solution of uranium);
- P_{NL} is the probability of non-leakage of the neutron from the configuration (the probability of neutron leakage is equal to "1- P_{NL}");
- $(abs\ FM)$ is the ratio of neutron absorption in the fissile medium on the total neutron absorption.

These specific results for each term are presented in Table 2.

Effect of nuclear data						
Reflector	**2.5 cm of water**			**20 cm of water**		
Temperature	**233 K**	**273 K (ice)**	**293 K**	**233 K**	**273 K (ice)**	**293 K**
K_{∞}	1.67932	1.67615	1.67442	1.69194	1.68982	1.68840
P_{NL}	0.61898	0.61729	0.61720	0.97470	0.97343	0.97317
$(abs\ FM)$	0.96495	0.96661	0.96711	0.65534	0.65842	0.65910

Effect of H_2O density								
Reflector	**2.5 cm of water**				**20 cm of water**			
Temperature	**233 K**	**273 K (ice)**	**273 K (liquid)**	**293 K**	**233 K**	**273 K (ice)**	**273 K (liquid)**	**293 K**
K_{∞}	1.68076	1.68113	1.67432	1.67441	1.69711	1.69785	1.68823	1.68843
P_{NL}	0.58914	0.58677	0.61798	0.61733	0.97131	0.97119	0.97323	0.97318
$(abs\ FM)$	0.96422	0.96400	0.96719	0.96709	0.63619	0.63412	0.65984	0.65932

Table 2 – Detailed results for case n°1

Based on this table, the following main conclusions could be drawn:

- for a variation of nuclear data only, a decrease of temperature leads to an increase of K_∞ (this effect might be explained by the decrease of neutron capture by ^{238}U (cf. Table 3). Concerning the effect of nuclear data on the probability of non-leakage and neutron absorption in fissile medium, it is difficult to conclude, as H_2O is present in the fissile medium and the reflector. In order to quantify only the effect of H_2O mixed with the fissile medium, a new study should is underway [4] considering fixed nuclear data for reflector (no variation in function of temperature);
- for a variation of H_2O density in fissile medium, a decrease of temperature and a change of H_2O phase (from liquid to ice) leads to a slight increase of K_∞ due to the decrease of hydrogen nucleus per volume unit in the fissile medium. However, this lower concentration on hydrogen nucleus explains also the increase of neutron leakage for a reflection by 2.5 cm of water and the lower neutron absorption in the fissile medium, which explains the decrease of Keff.

In order to show the impact on reactivity of each nuclide, a study is performed considering, for the configuration with a reflection by 2.5 cm of water, the nuclear data of the nuclide of interest at 233 K, all the others being at a temperature of 293 K. These results are presented in Table 3.

Isotope at 233 K	**H-ICE**	**^{16}O**	**^{235}U**	**^{238}U**	**(all at 293 K)**
Keff	1.00127 (+165 pcm)	0.99962 (0 pcm)	0.99944 (-18 pcm)	1.00163 (+201 pcm)	0.99962

Table 3 – Isotope impact on Keff for case n°1

Based on this table, the following conclusions can be drawn:

- effect of temperature on nuclear data of ^{16}O and ^{235}U have negligible impact on reactivity;
- decrease of temperature on nuclear data of ^{238}U leads to an increase of Keff due to the Doppler effect on capture cross section;
- decrease of temperature on nuclear data of H-H_2O leads to an increase of Keff. A new study [4] will allow to determine if this impact is due either to H_2O in the fissile medium or H_2O in the reflector or a combination of both.

3. Case n°2: simplified package loaded with UO2 PWR assembly

a. Presentation

This case models a simplified infinite array of packages loaded with one UO_2 PWR (pressurized water reactor) fuel assembly enriched at 5% in ^{235}U. The Figure 5 presents the studied configuration.
This assembly is a 17x17 (1.26 cm square pitch) rods array with 264 fuel rods and 25 “holes” (corresponding to missing rods). Each UO_2 rods has a diameter of 0.844 cm surrounded by a zirconium thickness of 0.53 mm and a height of 4.2 m (the foot, plenum and head of rods are not modelled).
The simplified packaging consists only of a 2.3 cm thick boronated resin with an internal cavity of the size of the assembly. The infinite packages array is modelled by applying a neutron reflection on the boronated resin.

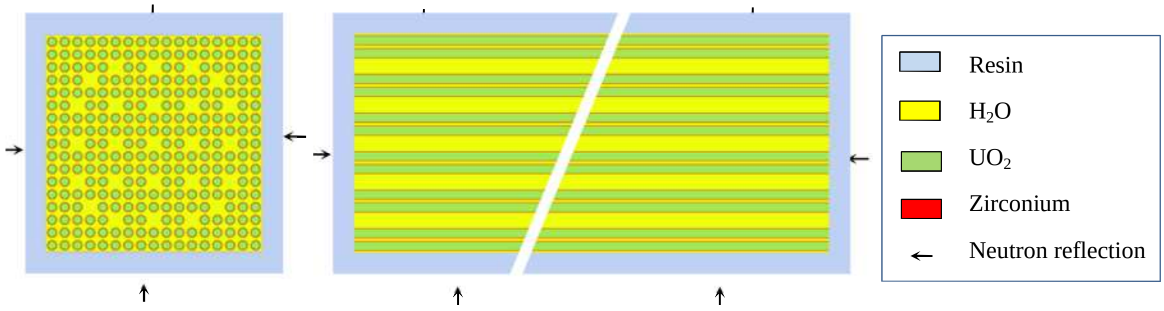


Figure 5 - Case n°2 configuration

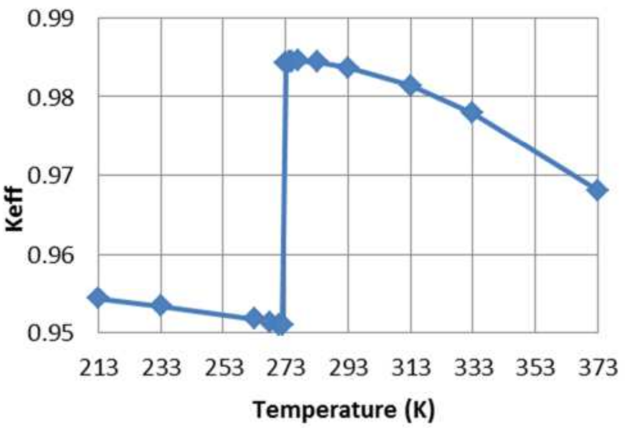
b. Results

The following table and figure present the reactivity results for the case n°2. When only nuclear data is studied, the water density used is those at 293 K. When only water density is studied, the nuclear data is considered at 293 K.

Temperature	Nuclear data effect	H_2O density effect	Nuclear data & H_2O density combined effect
233 K	0.98627 (+265pcm)	0.95314 (-3048pcm)	0.95582 (-2780pcm)
273 K (ice)	0.98435 (+73pcm)	0.95108 (-3254pcm)	0.95144 (-3218pcm)
273 K (liquid)	*	0.98444 (+82pcm)	*
293 K	0.98362		

* The S(α,β) for H-H_2O liquid at 273 K have not yet been processed.

Table 4 – Keff (σ = 10 pcm) and discrepancy with the Keff at 293 K for case n°2



Temp. (K)	Keff	Keff – Keff (293K)
213	0.95442	-2920 pcm
233	0.95314	-3048 pcm
263	0.95180	-3182 pcm
268	0.95140	-3222 pcm
273 (ice)	0.95108	-3254 pcm
273 (liq)	0.98444	82 pcm
277	0.98458	96 pcm
283	0.98445	83 pcm
293	0.98362	-
313	0.98138	-224 pcm

Figure 6 – Keff as a function of H_2O temperature (only H_2O density effect, nuclear data at 293 K)

As for case n°1, it could be seen that water density has a more significant impact on reactivity than nuclear data (~3000 pcm compare to 100-300 pcm). Indeed, the transition from liquid water to ice leads to a consequent decrease in density and thus to a significant decrease in Keff. It could also be seen that the Keff on Figure 6 has exactly the same shape as the H_2O density on Figure 3 as a function of temperature and that the maximum of Keff correspond to the maximum of H_2O density at 277 K (4°C) (increase of 96 pcm compare to Keff at 293 K). As for case n°1, the expression of Keff by the formula $K_{eff} = K_{\infty}.(abs\ FM)$ is presented in the following table (in this case, P_{NL} = 1 due to the neutron reflection on the boronated resin).

	Effect of nuclear data				**Effect of H_2O density**			
Temperature	**233 K**	**273 K (ice)**	**273 K (liquid)**	**293 K**	**233 K**	**273 K (ice)**	**273 K (liquid)**	**293 K**
$\boldsymbol{K_\infty}$	1.56839	1.56551	1.56521	1.56401	1.54507	1.54495	1.56447	1.56401
$(\boldsymbol{abs\ FM})$	0.62980	0.62968	0.62975	0.62921	0.61805	0.61674	0.63032	0.62921

Table 5 – Detailed results for case n°2

Based on this table, the following conclusions can be drawn:

- for a variation of nuclear data only, a decrease of temperature leads to an increase of K_∞ (due to Doppler effect) and to an increase of absorption in fissile medium which leads to a global increase of Keff;
- for a variation of H_2O density only, a decrease of temperature and a change of H_2O phase (from liquid to ice) leads to a decrease of K_∞ as this fuel assembly is still sub-moderated (this might be shown by the increase of the Energy corresponding to Average Lethargy of neutrons causing Fission (EALF) when temperature decreases: EALF = 0.5 eV at 293 K and EALF = 0.6 eV at 233 K) and a decrease of absorption in fissile medium (UO_2 rods). This last effect is due to the increase of neutron absorption in resin due to the fact that more neutrons enter the resin as there is less hydrogen atoms to diffuse neutron in the fuel assembly. These 2 effects explain the decrease of Keff.

In order to show the impact on reactivity of each nuclide, a study is performed considering the nuclear data of the nuclide of interest at 233 K, all the others being at a temperature of 293 K. These results are presented in Table 6.

Isotope at 233 K	**^{10}B in resin**	**^{1}H in resin**	**H-ICE**	**^{16}O in all materials**	**^{235}U**	**^{238}U**	**(all at 293 K)**
Keff	0.98357 (-5 pcm)	0.98299 (-63 pcm)	0.98525 (+163 pcm)	0.98367 (+5 pcm)	0.98369 (+7 pcm)	0.98530 (+168 pcm)	0.98362

Table 6 – Isotope impact on Keff for case n°2

Based on this table, the following conclusions can be drawn:

- effect of temperature on nuclear data of ^{16}O, ^{10}B and ^{235}U have negligible impact on reactivity;
- decrease of temperature on nuclear data of ^{238}U leads to an increase of Keff due to the Doppler effect on capture cross section;
- decrease of temperature on nuclear data of H-H_2O leads to an increase of Keff;
- decrease of temperature on nuclear data of ^{1}H in resin leads to a decrease of Keff.

CONCLUSIONS

Based on two simplified cases considering uranium moderated by H_2O, it has been shown that a decrease of temperature till negative ones leads to a decrease of Keff due to the decrease of H_2O density when water is in the solid phase (ice) compared to the liquid phase. These results validate the relevance of the achievement of criticality safety calculations for the transport of fissile material by considering nuclear data at room temperature when the fissile medium is moderated by water in the criticality safety analysis. However, other configurations considering water as a moderator should be performed in order to confirm these results, and, studies or experimental measurements should be carried out to validate nuclear data at low temperatures.

Moreover, in some criticality safety configurations, fissile materials can be moderated by plastics (modelled by polyethylene in criticality safety calculations) or other hydrogenated materials (for example HF for the transport of UF_6). As, for this type of material, the impact of density will be very different than the one for water (no phase change leading to a significant decrease in density), a new study should be carried out considering such materials.

Nuclear data could lead to an increase of reactivity when the temperature decreases in particular due to the Doppler effect on ^{238}U capture cross section. However, this increase is compensated, in the simplified cases of this study, by the decrease of Keff due to the decrease of H_2O density when water is in the solid form. In order to better understand the effects of nuclear data, new studies should be carried out to study more temperatures, and other configurations of calculations with other fissile media, other non-fissile materials or with other neutron spectra.

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