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EPEE: A TOOL TO COMPARE THE MODERATING EFFICIENCY OF A MATERIAL TO THE ONE OF WATER

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ABSTRACT

To carry out the criticality safety assessment of a nuclear facility or a process, it is necessary to distinguish not only the fissile materials which are handled inside, but also the non-fissile materials which may be close or even mixed with the fissile material. These non-fissile materials can moderate/slow down and thermalize the neutrons and thus increase the neutron production rate within the fissile material: they are referred to as “moderating materials”. This is particularly the case for non-fissile materials containing light nuclei such as hydrogen (and its isotopes) and, to a lesser extent, carbon (e.g. water, oils, polymers, solvents, lubricants, fire-extinguishing powders, etc.).

Criticality safety assessments usually take into account water as moderating material. Although there are materials with better moderating efficiency (e.g. high-density (0.96) polyethylene), water is indeed the historical moderator as its hydrogen mass content makes it a conservative moderator in many configurations. The moderating efficiency of a material, compared with water, depends on its composition and its density.

This paper presents a new approach to predict if a moderating material is covered by water when criticality safety is ensured by mass control, at optimal moderation. This methodology is based on criticality calculations performed using the APOLLO2-Sn route of CRISTAL V2.0. The approach involves simulating different non-fissile fictitious materials, with varying hydrogen, carbon and oxygen contents as well as density, and comparing them to water in terms of moderating efficiency for mass control, at optimal moderation. Elementary validation data are also presented by testing a large number of real moderating materials.

In addition, a user-friendly tool named “EPEE” has been developed to implement this methodology by entering either its density and molecular formula or the mass concentrations of its constituent elements.

KEYWORDS

ICNC2023, Criticality safety, moderation

1. INTRODUCTION

The CEA (Commissariat à l'énergie atomique et aux énergies alternatives / French Alternative Energies and Atomic Energy Commission) is a French public research establishment founded in 1945. Its missions cover the use of atomic energy for science, industry and national defense. One of the CEA's missions is to apply the necessary resources in order to ensure the safety of its own nuclear installations.

As presented in [1], one of the issues frequently encountered in nuclear criticality safety is the presence of materials which may have a better moderating efficiency than water when mixed more or less intimately with the fissile materials. This topic was originally encountered in fuel fabrication facilities, in which various materials are added during the process (porogen, lubricant, etc.). Over the past thirty years, it has spread to other fuel cycle installations such as hot laboratories, solid waste processing facilities and consequent waste storage facilities, including those operated by the CEA.

This paper presents a new approach to presumably predict if a moderating material is covered by water regarding criticality safety mass limits. After a brief reminder of the concept of moderating efficiency (see section 2), this paper details two methods (called "simplified" and "extensive" hereafter) that may be used to determine whether a material is more or less moderating than water (see sections 3 and 4); some elementary validation data are also presented by comparing, for several non-fissile materials, the results of these methods with those from state-of-the-art criticality calculations. Finally, this paper presents a user-friendly tool named "EPEE" ("Estimateur Pénalisant pour matériau Enveloppé par l'Eau" (in French), i.e. penalizing estimator for material less moderating than water) and its GUI (graphical user interface), which was developed in order to facilitate the use of this methodology (see section 5).

2. MODERATING EFFICIENCY

To perform the criticality safety assessment of a nuclear facility, it is necessary to distinguish not only the fissile materials which are handled inside, but also the non-fissile materials which may be close or even mixed with the fissile material. These materials can moderate (slow down) the neutrons and thus increase the production rate of neutrons within the fissile material: they are referred to as "moderating materials". This is the case for those containing light nuclei such as hydrogen (and its isotopes) and, to a lesser extent, carbon (e.g. water, oils, polymers, solvents, fire-extinguishing powders, lubricants, etc.).

Criticality safety assessments usually take into account water as moderating material. It is indeed the historical moderator as its hydrogen mass content makes it a conservative moderator in many configurations regarding criticality safety limits determination (e.g. mass of fissile material, etc.).

The "moderating efficiency" of a material matters when establishing the criticality safety limits for a work unit or piece of equipment. It relies on its propensity to moderate (slow down) neutrons relative to its absorption capacity, with regard to the scattering and absorption cross-sections of its various constituents as well as their proportions. It is well known that some materials have a better moderating efficiency than water: for example, it concerns high-density polyethylene ((CH₂)_n, 0.96 g.cm⁻³) which results on the one hand from a higher mass content of hydrogen in CH₂ than in water, and on the other hand from absorption cross-sections of oxygen higher than those of carbon. More generally, the moderating efficiency of a material, compared with water, depends on its composition and its density. Several examples of moderating materials are given in the CEA "Nuclear Criticality Safety Guide Sheets" [2] (see Sheets 4 and 12 in particular [3] – available upon request from the authors by e-mail, in French until now with an English translation in progress).

At this stage, for the remainder of this article, it can be noted that the following study only relates to configurations for which criticality safety is based on limiting only the fissile material (F) mass, regardless of the quantity of moderating materials (i.e. at optimal moderation). Thus, affirming that "a material (M) has a lower moderating efficiency than the one of water (W)" implies that "the Maximum Permissible Mass (MPM, i.e. at optimal moderation) for a mixture between a fissile material and this

material ($F+M$) is higher than the one obtained for a mixture between this same fissile material and water ($F+W$)" (for a given value of k_{eff} and for the same criticality calculation route, assuming there is no calculation bias).

The issue addressed here is to be able to easily predict, without carrying out a criticality calculation, whether a material has a lower moderating efficiency than water or not. To fulfill this objective, two methods are described and then tested:

- A "simplified method", based on the comparison of hydrogen mass concentrations;
- An "extensive method", based on extensive calculations taking into account many other chemical elements.

The two following sections present each of these methods.

3. SIMPLIFIED METHOD

3.1. Introduction and notations

A "historical" approach to verify that a non-fissile material has a lower moderating efficiency than that of water consists in comparing their Hydrogen Mass Concentrations (HMCs), given that hydrogen is present in many molecules and that it is an effective neutron moderator due to its low molar mass¹. The HMC of a non-fissile material M , noted $[H](M)$, can be calculated using Eq.1:

$$[H](M) = \%wt.H(M) * \rho(M) \quad (1)$$

where:

- $\%wt.H(M)$ is the mass fraction of hydrogen in the material M ;
- $\rho(M)$ is the density of the material M .

From this notation, it is possible to compare the HMC of a material (M) to the one of water (W) by means of the Hydrogen Mass Concentration Ratio (HMCR) as shown in Eq.2:

$$HMCR(M) = \frac{[H](M)}{[H](W)} \quad (2)$$

The simplified method then consists in assuming that a material M has a lower moderating efficiency than the one of water if:

$$HMCR(M) < 1$$

The advantage of this method is that it only requires calculating the HMC of the material concerned, for example from its molecular formula and its density. However, this amounts to neglecting the influence of atoms other than hydrogen, which may question the validity of the method in all cases.

3.2. Criticality calculation configuration

The validity of the simplified method is tested on the following calculation configuration:

- Fissile material (F): pure homogeneous metallic ²³⁹Pu (at maximum theoretical density);
- Optimal moderation by a moderating material (M) or water (W);
- Spherical shape of the fissile/moderator mixture ($F+M$ or $F+W$) surrounded by a 20 cm-thick layer of water;
- k_{eff} criterion: 0.95.

A representation of the calculation model is presented in Figure 1.

¹ This approach cannot therefore be applied to non-hydrogenated materials (i.e. containing ¹H) such as graphite, heavy water, Teflon, beryllium, etc.

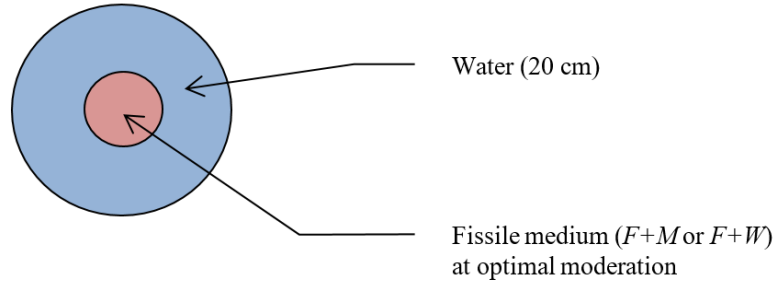


Figure 1. Criticality calculation model

For a given moderator-to-fuel ratio, the radius of fissile medium is determined for the given k_{eff} value (0.95) using the APOLLO2-Sn route of the French criticality package CRISTAL V2.0, using the bias reduction procedure [4]. The Maximum Permissible Masses (MPMs) of plutonium are calculated at optimal moderation, both for mixtures of plutonium with moderator M ($MPM(M)$) and water ($MPM(W)$).

A Maximum Permissible Mass Ratio (MPMR) can be calculated for a material M as shown in Eq.3:

$$MPMR(M) = \frac{MPM(W)}{MPM(M)} \quad (3)$$

The MPMR is the true safety criterion, indeed calculations demonstrate that a material M has a lower moderating efficiency than water if:

$$MPMR(M) < 1$$

3.3. Application of the simplified method: elementary validation data

The simplified method is tested on a selection of some moderating materials of interest:

- Polyethylene, which is commonly used to account for the presence of polymers or vinyl;
- TBP (tributylphosphate), which is a liquid-liquid extraction solvent in fuel reprocessing process;
- Ethanol, used for cleaning purposes in laboratories;
- “Araldite”, which refers to polymer fuel encapsulating polymer resins (see [1] for more details);
- TETA (triethylenetetramine), which can be used as hardening agent for epoxy resins;
- Ethylene glycol, which can be used as antifreeze in cooling circuits.

For each of these materials, the simplified method is tested by calculating its HMCR and MPMR. Results are shown in Table I.

Note: to facilitate reading, for HMCR or MPMR values greater than 1, corresponding boxes are hatched.

Table I. Results of the simplified method for metallic ^{239}Pu at optimal moderation, surrounded by a 20 cm-thick layer of water

Moderating material			Application of the simplified method		Criticality safety calculation results	
Name	Formula	Density (g.cm ⁻³)	HMC=[H] (g.cm ⁻³)	HMCR	MPM (g Pu)	MPMR
Water	H ₂ O	0.998	0.1117	-	403	-
Polyethylene	CH ₂	0.960 (max)	0.1380	1.24	271	1.49
		0.75	0.1078	0.97	419	0.96
TBP	C ₁₂ H ₂₇ O ₄ P	0.972	0.0993	0.89	452	0.89
Ethanol	C ₂ H ₆ O	0.7893	0.1036	0.93	452	0.89
“Araldite”	C ₅ H ₈ O ₂	1.180	0.0950	0.85	406	0.99
TETA	C ₆ H ₁₈ N ₄	0.982	0.1218	1.09	651	0.62
Ethylene glycol	C ₂ H ₆ O ₂	1.114	0.1085	0.97	373	1.08

These results show the validity of the simplified method for most of the considered materials:

- High-density polyethylene has a greater moderating efficiency than water;
- When the density of polyethylene varies up to 0.75 g.cm^{-3} , the simplified method predicts that polyethylene is less moderating than water: this result is consistent with that presented in [1];
- TBP, ethanol and “araldite” are correctly predicted as less moderating than water; however, it should be noted that HMCR and MPMR values are not equal, in particular for “araldite” (its MPMR is just below 1 whereas its HMCR is about 0.85); this can be related to the variety and number of atoms (carbon, oxygen) in the chemical formulas, as well as various densities;
- Although TETA has a HMCR greater than 1, calculations show that it is far less moderating than water (MPMR = 0.62): this is due to the presence of absorbing nitrogen atoms in the chemical formula, which are not taken into account in the simplified method;
- Ethylene glycol is also incorrectly predicted but in an unsafe sense: indeed, its HMCR is just below 1 whereas its MPMR is not.

Regardless of chemical compositions, it can also be noticed that MPMR values seem higher than the HMCR ones when the density of the compound is higher (or close) than the one of water.

3.4. Conclusion

The simplified method gives a preliminary idea of the moderating efficiency of a material compared to the one of water but may sometimes be imprecise, depending on the chemical composition of the material and its density. A more robust method is therefore needed.

4. EXTENSIVE METHOD

The “extensive method” involves simulating different fictitious materials, by varying the concentrations of different atoms as well as density, and comparing them to water in terms of moderating efficiency.

4.1. Principles

The objective is to be able to represent any moderating material in a conservative way (in terms of criticality safety, e.g. guarantee that $\text{MPMR} < 1$), with the smallest number of atoms. In order to ensure that this approach is conservative, the two following rules have to be respected:

- Every atom has to be represented by (itself or) another one whose moderating efficiency is equal or higher than the one of the atom it represents;
- There must be conservation of mass concentrations (in g.cm^{-3} for instance) of each atom.

The first rule is quite trivial, for the consistency of the conservation. The second rule exists because the mass concentration is directly and linearly proportional to the macroscopic cross-sections of the material: it must therefore be kept identical. This rule could, in theory, be disregarded or tweaked. For example the mass concentration of an atom that has a weak moderating efficiency can be decreased when representing this atom by another one way more moderating. This would require a lot more calculations to have enough confidence in the new model.

A two-dimensional approach considering only carbon and hydrogen could have been studied but would probably lead to substantial margins. It was therefore chosen to select three different atoms to represent all chemical elements, as they can all be found within most moderating materials and are rather light: hydrogen, carbon and oxygen. Hydrogen, as the lightest element, has the best moderating efficiency and is therefore necessarily in this list. Beryllium could have been used, but given its physico-chemical properties and its low representativeness among the various moderators, carbon was preferred: indeed, carbon is present in the majority of moderators and is only slightly more massive than beryllium. Oxygen was chosen for the same reasons as carbon: it is one of the lightest elements and it is present in a large quantity of moderators, despite having a slight neutron absorbing capacity.

With this selection of three atoms (H, C, O), it is now necessary to determine the specific sorting rules to assimilate any material to a fictitious chemical compound only composed of hydrogen, carbon and oxygen, while still being completely sure to remain conservative regarding the initial moderator.

A brief reminder of some physico-chemical properties of the elements from the three first rows of the periodic table of elements, that are relevant to the neutrons, is given hereafter:

- Elements from the last group (helium, neon, argon, etc...) are the noble gases, they are not taken into account because they are not usually present in moderators;
- Lithium, boron, nitrogen and chlorine are not taken into account because they are neutrons “absorbers”, it will be discussed in section 4.4;
- Beryllium has a better moderating efficiency than carbon so, it must then be represented, like deuterium and tritium, by hydrogen to remain conservative;
- Fluorine has a better moderating efficiency than oxygen (lower inelastic collision and neutron absorption cross-sections), but yet lower than the one of carbon, so it is represented by carbon.

Also, all other atoms in the third period have been tested and have respectively a lower moderator efficiency compared to oxygen, so they are represented as such. It is also the case for the atoms in the following rows of the periodic table due to their high molar mass. Figure 2 presents a summary of these sorting rules for the first three rows of the periodic table.

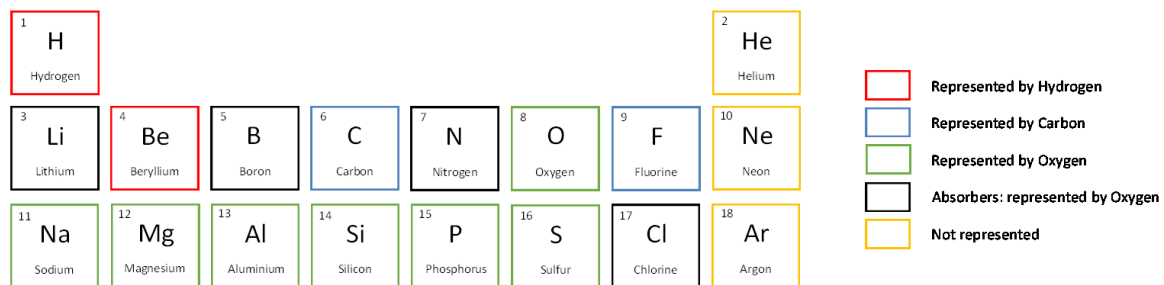


Figure 2. Sorting rules for the first three rows of the periodic table of elements

An example of the use of these sorting rules, to form the fictitious compound used in the extensive method from the composition of a hypothetical moderator and its “equivalent” mass concentrations of hydrogen, carbon and oxygen (noted $[H]_{eq}$, $[C]_{eq}$ and $[O]_{eq}$), is given in Table II.

Table II. Example of the use of the sorting rules: calculation of equivalent mass concentrations

Composition of a hypothetical moderator to be tested	Composition of the fictitious moderator considered in the extensive method
$[H] = 0.10 \text{ g.cm}^{-3}$	$[H]_{eq} = 0.11 \text{ g.cm}^{-3}$
$[Be] = 0.01 \text{ g.cm}^{-3}$	
$[C] = 0.56 \text{ g.cm}^{-3}$	$[C]_{eq} = 0.58 \text{ g.cm}^{-3}$
$[F] = 0.02 \text{ g.cm}^{-3}$	
$[O] = 0.45 \text{ g.cm}^{-3}$	$[O]_{eq} = 0.48 \text{ g.cm}^{-3}$
$[Si] = 0.03 \text{ g.cm}^{-3}$	
Density = $[H] + [Be] + [C] + [F] + [O] + [Si] = 1.17 \text{ g.cm}^{-3}$	Density = $[H]_{eq} + [C]_{eq} + [O]_{eq} = 1.17 \text{ g.cm}^{-3}$

4.2. Calculations

With this sorting rules being defined, the next step is to determine the relationship between the three mass concentrations that will allow predictions on the moderating efficiency of a material to be made.

The criticality calculation model used for the following is the same than the one described in section 3.2.

The process to find the relations between these three atoms is the following one:

- Start by testing a fictitious compound with only two of the three atoms (for instance, C and H);
- Set a specific value of density for the compound;
- Run criticality safety calculations to determine the MPMR of the compound;
- Fit the mass concentrations and restart the calculations until the results are close enough to $MPMR = 1$ (with an accuracy of about $10^{-4} \text{ g.cm}^{-3}$);
- Identify the two closest pairs of mass concentrations (leading to MPMR values just below and just above 1) and then linearly interpolate them to become a single point in the system $\{[C], [H], \text{density}\}$ that corresponds to a MPMR value equal to 1;
- Repeat for other density values (in steps of 0.05 g.cm^{-3});
- Repeat for the two other pairs of mass concentrations (H and O; C and O).

Those results are obtained for a specific set of initial parameters (sphere of metallic ^{239}Pu at optimal moderation, surrounded by a 20 cm-thick layer of water, with a k_{eff} criterion equal to 0.95). All those parameters have an impact on the neutrons overall behaviour. This makes the results depending on those parameters, and it implies that they therefore cannot be generalised for another set of parameters. Additional calculations have been performed using the same process for three other fissile materials and three other reflectors (resultats are not reported in this paper).

4.3. Results

The plot of the results obtained has a form whose schematic appearance is shown in Figure 3.

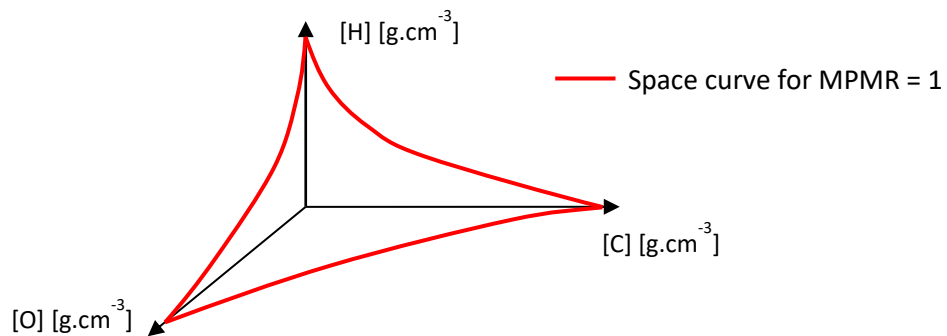


Figure 3. Shape of the space curve for MPMR = 1

Considering the shape of this “space curve” (set of points leading to $MPMR = 1$), a linear interpolation of the curve (to define a plane equation to be respected) would lead to the “MPMR equal to 1” criterion being slightly exceeded, that’s why a tangent lines to the curve is preferred to be conservative from a safety point of view. In order for a tangent to remain at any point under the curve, a domain of validity must necessarily be defined, because the domain between the two points of the tangent will lead to a violation of the criterion ($MPMR$ superior to 1). A density of 0.5 g.cm^{-3} and lower was chosen, as a higher density would have limited the range of validity. A lower density would have increased the margins between the model and reality. Therefore no moderator should be simulated for a density between 0.45 g.cm^{-3} (second point used for the tangent line) and 0.5 g.cm^{-3} .

It is then necessary to determine the mass concentration pairs $\{[H] \text{ and } [O]\}$ or $\{[H] \text{ and } [C]\}$ for the two points of the tangent, i.e. the two points that have a MPMR equal to 1 for densities of 0.45 and 0.5 g.cm^{-3} . To do this, proceed in the same way as for defining the curve in Figure 3 (see section 4.2).

It has been studied that for a mass concentration of carbon close to the one of oxygen which serves as a reference for the points of the tangent (couples at $d=0.45$ and 0.5 which lead to $MPMR=1$), the points of the plane passing through $[H, O]$ ($d=0.5 \text{ g.cm}^{-3}$), $[H, O]$ ($d=0.45 \text{ g.cm}^{-3}$) and $[H, C]$ ($d=0.5 \text{ g.cm}^{-3}$) are

necessarily under the "space curve" defined by the points d, [C], [H] which leads to a MPMR value equal to 1. No pair {[H], [C], [O]} that respects the equation of the plane and the validity domain can therefore lead to a MPMR superior to 1.

All this work leads to the plane equation indicated in Eq.4, where $[H]_{eq}$, $[C]_{eq}$ and $[O]_{eq}$ are the mass concentrations of H, C and O using the sorting rules defined in section 4.1, expressed in $g.cm^{-3}$:

$$[H]_{eq} + 0.04434685.[C]_{eq} + 0.03149435.[O]_{eq} - 0.13856741 = 0 \quad (4)$$

Note: the respective multiplier coefficients in Eq.4 underline that hydrogen has a much higher moderating efficiency than carbon, which is also itself slightly more efficient than oxygen.

To test a moderating material with the extensive method, it is therefore sufficient to:

- Calculate its $[H]_{eq}$, $[C]_{eq}$ and $[O]_{eq}$ values using the sorting rules;
- Then use the formula of Eq.4: one can reasonably predict that this material has a less moderating efficiency than water if the following inequality (Eq.5) is satisfied:

$$[H]_{eq} + 0.04434685.[C]_{eq} + 0.03149435.[O]_{eq} - 0.13856741 < 0 \quad (5)$$

As indicated before, this plane equation was obtained for a sphere of metallic ^{239}Pu at optimal moderation, surrounded by a 20 cm-thick layer of water. To obtain the plane equations for other fissile material and reflectors, the same calculations have been performed, for each couple of fissile material and reflector, and led to other coefficients in the plane equation.

Note: as the principle of this method makes the variables $[H]_{eq}$, $[C]_{eq}$, $[O]_{eq}$ and ρ (density) related ($[H]_{eq} + [C]_{eq} + [O]_{eq} = \rho$), it is also possible to write Eq.4 with another set of three variables. Thus, the plane equation may be represented as a function of density, $[H]_{eq}$ and $[C]_{eq}$ as in the figure in Appendix A.

4.4. Addition to the method in case of the presence of neutron absorbers

All atoms have scattering cross-sections resulting in slowing down neutrons, as well as an absorption cross-section (n,γ) which makes neutrons disappear from the fissile medium. Those two cross-section are involved in the k_{eff} formula.

Some atoms have a very high absorption neutron cross-section: they will be referred as "absorbers". The following atoms are part of this absorbers group: Li, B, N, Cl, Ti, Tc, Sm, In, Hf, Gd, Fe, Eu, Er, Dy, Co, Cd, Ag. Representing an absorber as a regular moderator constitutes a double penalty. When an absorber is added to a regular moderator, in general, the maximum permissible mass significantly increases. This phenomenon may be related to the fact that the absorber's absorption neutron cross-section is not compensated by the scattering cross-sections.

For calculations with absorbers within the moderator, neglecting the absorbers' mass concentration will therefore still correspond to a conservative situation. Doing so implies to neglect both the scattering cross-sections and the absorption neutron cross-section of the absorber (which is far larger than the scattering one, which remains conservative. To do this, the moderator must be completely removed from the mass concentration of the absorber, and the moderator's density is adjusted accordingly.

An example is given for TETA (containing nitrogen atoms) in section 4.5 hereafter.

4.5. Application of the extensive method: elementary validation data

The validity of the extensive method is tested on the calculation configuration presented in section 3.2 (metallic ^{239}Pu at optimal moderation, in spherical shape, surrounded by a 20 cm-thick layer of water. Table III presents the results for the materials already studied with the simplified method.

Table III. Results of the extensive method for metallic ^{239}Pu at optimal moderation, surrounded by a 20 cm-thick layer of water

Moderating material			Application of the extensive method				Criticality safety calculation results	
Name	Formula	Density (g.cm ⁻³)	[H] _{eq} (g.cm ⁻³)	[C] _{eq} (g.cm ⁻³)	[O] _{eq} (g.cm ⁻³)	Inequality "Eq.5"	MPM (g Pu)	MPMR
Water	H ₂ O	0.998	-	-	-	-	403	-
Polyethylene	CH ₂	0.960 (max)	0.1380	0.8220	0	> 0	271	1.49
		0.75	0.1078	0.6422	0	< 0	419	0.96
TBP	C ₁₂ H ₂₇ O ₄ P	0.972	0.0993	0.5260	0.3467	< 0	452	0.89
Ethanol	C ₂ H ₆ O	0.7893	0.1036	0.4116	0.2741	< 0	452	0.89
"Araldite"	C ₅ H ₈ O ₂	1.180	0.0950	0.7078	0.3772	< 0	406	0.99
TETA	C ₆ H ₁₈ N ₄	0.982	0.1218	0.4839	0	> 0	651	0.62
Ethylene glycol	C ₂ H ₆ O ₂	1.114	0.1085	0.4311	0.5744	> 0	373	1.08

Table III shows that the extensive method correctly predicted all materials that are covered by water, which fulfills its initial purpose, except TETA for which it remains too penalizing. In particular, ethylene glycol is now correctly predicted, which was not the case using the simplified method. TETA is not predicted as having a less moderating efficiency than the one of water: indeed, the method remains very conservative when there are neutron absorbers such as nitrogen in this case (the absorption cross-section is not taken into account), despite the elements presented in section 4.4. Taking into account these absorbers still requires further developments to make the method less penalizing and more accurate.

Additional calculations have been carried out and have demonstrated a correct prediction of the less moderating efficiency than the one of water for many materials (alcanes, epoxy resins...) with the extensive method, as long as they have no "absorber atom".

5. DEVELOPMENT OF A TOOL TO IMPLEMENT THE EXTENSIVE METHOD

As shown in section 4, the extensive method only requires to know the mass concentrations of the constituent elements of the material to be compared to water. In order to facilitate the use of this method, a user-friendly tool has been developed and named "EPEE" ("Estimateur Pénalisant pour matériau Enveloppé par l'Eau" (in French), i.e. penalizing estimator for material less moderating than water). This GUI enables to predict if a material has a less moderating efficiency than the one of water.

The input data of "EPEE" are:

- The fissile material (available at this stage: ^{239}Pu , U-93.5%, UO₂-5%, MOX);
- The reflection conditions (available at this stage: 20 cm of water; 1 m of concrete (with 3% or 9% wt. water content); 25 cm of lead followed by 20 cm of water);
- The moderating material:
 - o Either the mass concentrations of: [H+D+Be] (i.e. [H]_{eq}), [C+F] (i.e. [C]_{eq}), [absorbers], [other atoms];
 - o Or its density and molecular formula².

² A parser is implemented in "EPEE" to distinguish the number of atoms indicated after each element in the molecular formula (or 1 if there is no number): for instance, if the user enters "C12H27O4P", this involves in "EPEE" that there are 12 atoms of carbon, 27 atoms of hydrogen, 4 atoms of oxygen and 1 atom of phosphorus (considered as oxygen due to the extensive method). The mass concentrations of each element are then calculated taking the density of the material into account.

Its output data are:

- Mass concentrations of [H+D+Be], [C+F] (when molecular formula is used as input);
- A graphic representation of the position of the point (coordinates: density, [H+D+Be], [C+F]) with respect to the plane whose equation is determined by the extensive method (see section 4.3);
- The conclusion on the less or possibly more moderating efficiency of the material than the one of water.

Furthermore, an additional feature has been implemented: it consists of determining the density limit value below which a material has a lower moderating efficiency than water, based on the extensive method results. This density limit corresponds to the intersection between the envelope plane and the line which connects the origin to the coordinates of the material in space (d, [C], [H]) without absorbers. It is corrected to take into account any mass concentrations of neutron absorbers within the material.

A screenshot of the “EPEE” user interface is shown in Appendix A in the case of polyethylene (CH₂). In particular, one can find there the maximum density limit determined by the extensive method for CH₂, i.e. 0.7626 g.cm⁻³, which is consistent with the value of 0.75 g.cm⁻³ previously indicated.

6. CONCLUSIONS

Two methods can be used to presumably predict if a moderating material is covered by water when criticality safety is ensured by mass control, at optimal moderation:

- A simplified method, based on a comparison of hydrogen mass concentrations;
- An extensive method, based on criticality calculations by simulating different fictitious materials with varying hydrogen, carbon and oxygen contents as well as density, and comparing them to water. This method was built in a penalizing way regarding criticality safety, in order to be sure that a material is covered by water.

The simplified method allows to do calculations “by hand” but may sometimes lead to incorrect results.

The extensive method is more accurate and its validity was verified in most cases by testing a large number of real moderating materials. Further developments would remain necessary to make the method less penalizing and more precise in the case of the presence of absorbing atoms (nitrogen for example).

In addition, a user-friendly tool named “EPEE” has been developed to implement this method by entering either its density and molecular formula or the mass concentrations of its constituent elements.

Finally, this new method (with its “EPEE” tool) proves to be fast and reliable in determining whether a material is less moderating than water; it can therefore be used as a predictive approach but cannot however replace a real criticality calculation within the framework of a criticality safety demonstration.

REFERENCES

- [1] Kyriazidis, G., Dorval, A. Neutron moderation materials other than water: how, why and when the problems arose and the solutions proposed by the CEA. To be presented in Proceedings of the 12th International Conference on Nuclear Criticality Safety (ICNC 2023). Sendai, Japan, October 1-6, 2023.
- [2] Dorval, A., Prigniau, M., Casoli, P., Fillastre, E., Gagnier, E. The new version of the criticality safety guide sheets collection. Proceedings of the 11th International Conference on Nuclear Criticality Safety (ICNC 2019). Paris, France, September 15-20, 2019.
- [3] CEA. Nuclear criticality safety guide sheets, 2023 (available upon request by e-mail; English translated version in progress).
- [4] Gomit, J.M., Duhamel, I., Entringer, A. et al. CRISTAL V2.0: a new-generation criticality package. Proceedings of the 10th International Conference on Nuclear Criticality Safety (ICNC 2015). Charlotte, USA, September 13-17, 2015.

APPENDIX A: "EPEE" USER INTERFACE

EPEE
- □ ×

Fissile material and neutron reflector

Pu239
Water - 20 cm

Moderating material composition

Description by formula and density

Molecular formula Density g.cm⁻³

Description by mass concentrations

Concentration [H+D+Be] g.cm⁻³

Concentration [C+F] g.cm⁻³

Concentration [absorbers] g.cm⁻³

Concentration [other] g.cm⁻³

Results

[H]_{eq} = [H+D+Be] = 0.1380 g.cm⁻³

[C]_{eq} = [C+F] = 0.8220 g.cm⁻³

The moderation efficiency of the material appears to be **higher** than the one of water.

This material would be less moderating than water if its density were less than **0.7626 g.cm⁻³** (with the same mass fractions of each atom).

Limit values to be less moderating than water:

- ✓ Max. density limit = 0.7626 g.cm⁻³
- ✓ [H]_{eq} = [H+D+Be] = 0.1096 g.cm⁻³
- ✓ [C]_{eq} = [C+F] = 0.6530 g.cm⁻³

Values for the indicated material:

- ✓ Density = 0.96 g.cm⁻³
- ✓ [H]_{eq} = [H+D+Be] = 0.1380 g.cm⁻³
- ✓ [C]_{eq} = [C+F] = 0.8220 g.cm⁻³