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THE CRITICALITY-SAFETY DURING TRANSPORT OF MATERIALS CONTAINING LOW CONCENTRATIONS OF SOLID FISSILE MATERIAL

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ABSTRACT

With the aging of nuclear installations, the need to transport large volumes of waste draws the community to inquire after the criticality risk of materials containing low concentrations of solid fissile material. The main issue about it is to determine the ratio between the solid non-fissile mass and the solid fissile mass that ensures an infinite multiplication factor k_∞ lower than 0.95. US calculations on ^{235}U in a SiO_2 matrix establish that this ratio should be 2000. This paper presents the IRSN approach studying any solid non-fissile matrix and the different fissile isotopes considered in the IAEA Regulation for the Safe Transport of Radioactive Material which are ^{235}U, ^{233}U, ^{239}Pu, ^{241}Pu. In the plutonium case, moreover, the existence of a positive temperature coefficient is brought to light.

Key Words: Transport, IAEA, Fissile Exception, Positive Temperature Coefficient, Criticality

1 INTRODUCTION

The national and international radioactive material transportation is regulated by the IAEA safety standards [1]. These standards include, among others, some rules under which, packages containing fissile material can be excepted from the requirements of the regulation to be transported.

The need to transport large volumes of waste, originally from the decommissioning of nuclear facilities, brought some member states to propose an addition to the excepted fissile material list of the regulation concerning low concentrations of solid fissile material in solid non-fissile material. The definition of this new type of excepted fissile material is the following (proposal USA 04/03 §e):

"Low concentrations of solid fissile material commingled with solid non-fissile material (e.g. consolidated wastes, activated materials) provided that:

- the fissile material is essentially uniformly distributed throughout a non-fissile solid collection of non-solid objects, or is essentially uniformly distributed in a non-fissile solid compact binding agent (such as concrete, soil, ceramic, etc.).
- the fissile material is relatively inseparable, or it is intrinsically contained in a relatively inseparable matrix; inseparable means inseparable by leaching, burning and mechanical impact.

- the estimated average mass of fissile material within any 100 kg of non-fissile solid, excluding any reflecting material, does not exceed 1g fissile nuclide / 2000 g of non-fissile solid (for the purpose of calculation excluding beryllium, graphite and substances enriched in deuterium)."

At present, the dilution factor of fissile material in non-fissile material proposed is 1/2000. This factor corresponds roughly to 60% of the minimum critical fissile material concentration of 1.33 g ^{235}U/l in a 1600 g SiO_2/l matrix (see reference [2] from the ORNL-USA team). SiO_2 was chosen in this study because it is the most representative matrix associated with transport of waste materials and has a very low probability for absorbing neutrons. The 60% value allows to take into account non-fissile materials other than SiO_2.

This demonstration mainly refers to waste disposal of uranium blended with soil. It is not directly applicable to excepted fissile materials that can concern all types of material. The present work objective is to make sure that the use of a ratio 1/2000 is appropriate to prevent a critical situation for any type of material by examining any non-fissile matrix and the different fissile isotopes considered in the IAEA Regulation for the Safe Transport of Radioactive Material which are ^{235}U, ^{233}U, ^{239}Pu, ^{241}Pu.

2 CALCULATION METHOD

For the purpose of calculation, we considered an homogeneous mixture of fissile material (denoted F) and non-fissile material (denoted NF) with a fixed non fissile to fissile mass ratio (M_{NF}/M_F). We also considered an infinite medium since the main interest of the proposal USA 04/03 §e is the absence of a limit on mass or volume of the waste.

Let us note here that the risks associated with the gathering of fissile material can be ruled out because the proposal specifies that within any 100kg of non-fissile material, there cannot be more than 50g of fissile material. Therefore, the gathering of 8 casks, for example, will not lead to the gathering of more than 400 g of fissile material which is considered as acceptable due to the values of the minimum critical masses[i].

The neutron multiplication in a medium so defined, was studied with the APOLLO2 cell code from the CRISTAL V0.2 package [3] using a 172-group energy structure. The infinite multiplication factor (k_∞) was thus extracted as a function of the moderator to fissile atomic ratio (H/F) for different values of M_{NF}/M_F. This allows the determination of M_{NF}/M_F for which the medium is sub-critical, whatever the moderation. Regarding these calculations, it is important to keep in mind that there are few or no benchmarks allowing codes validation for fissile material moderations other than water (such beryllium, bismuth, concrete, silicon oxide, etc...). Therefore, the CRISTAL package is not validated for most studied moderator materials. As a consequence, results should be considered as qualitative rather than quantitative and will need to be confirmed by calculations performed with other codes and other nuclear data libraries.

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[i] For ^{241}Pu, whose critical mass is below 400 g, the risk to have ^{241}Pu alone can be reasonably excluded.

3 URANIUM 235 RESULTS AND NON-FISSILE MATRIX PROPERTIES

The case of the fissile isotope uranium 235 was studied first. Calculations performed refer to:

- the case considered by the US, namely an SiO_2 matrix,
- matrix composed of nuclei (A) with small or large atomic number possibly in presence of oxygen (A_xO_y type),
- so called “mixed” matrix of concrete, soils, or ceramics.

3.1 SiO_2 matrix

3.1.1 Moderation optimum and neutron spectrum as a function of M_{NF}/M_F

Several calculations carried out for various SiO_2 to ^{235}U mass ratios (M_{SiO2}/M_{U5}) show that the maximum of the infinite multiplication factor (k_∞) is obtained at H/U equal to zero for high mass ratios (about 2000), whereas it is obtained at H/U different from zero for low mass ratios (about 200).

The origin of this difference can be explained by observing the corresponding neutron spectra. Indeed:

- for high mass ratios (about 2000), neutrons emitted by uranium are slowed down by the numerous SiO_2 molecules and the neutron spectrum is characteristic of thermal neutrons ($E_n \sim 0.1$ eV with E_n the neutron energy),
- for low mass ratios (about 200), the neutron spectrum is characteristic of fast neutrons ($E_n > 100$ eV).

In the thermal region, when H/U increases, neutrons are absorbed by hydrogen atoms whereas the absorption cross-sections of SiO_2 as well as the fission and absorption cross-sections of ^{235}U behave as one over the neutron velocity so that the ratio $\sigma_f^{U5}/(\sigma_a^{U5}+\sigma_a^{SiO2})$[ii] is constant as a function of the neutron energy in a first approximation. Therefore, k_∞ defined as $\sigma_f^{U5}/(\sigma_a^{U5}+\sigma_a^{SiO2}+\sigma_a^{H})$ decreases. As a result, the maximum of k_∞ is generally obtained for H/U equal to zero when the neutron spectrum of a unmoderated medium is thermal.

Concerning low mass ratios, the neutron energy for H/U equal to zero corresponds to absorbing resonances of Si. The fact that neutrons can be trapped in a resonance infers a fall-off of k_∞ for H/U equal to zero. This explains why the maximum of k_∞ is not obtained for H/U equal to zero in this case.

3.1.2 Sub-critical M_{NF}/M_F estimate

In any case, it is possible to determine the acceptable SiO_2 to ^{235}U mass ratio (M_{SiO2}/M_{U5}), corresponding to the acceptable criterion ($k_\infty \leq 0.95$), by varying this ratio. This acceptable mass ratio is equal to 1200 as it can be seen in Figure 1.

This value is congruent with the value obtained by the US team in the reference [2]. In addition, the US team chose to apply a factor of 60% to this value in order to take into account codes validation and non-fissile materials other than SiO_2 leading to the value of 2000 of their proposal.

[ii] σ_f^E and σ_a^E refer to the fission and absorption microscopic cross-sections of E.

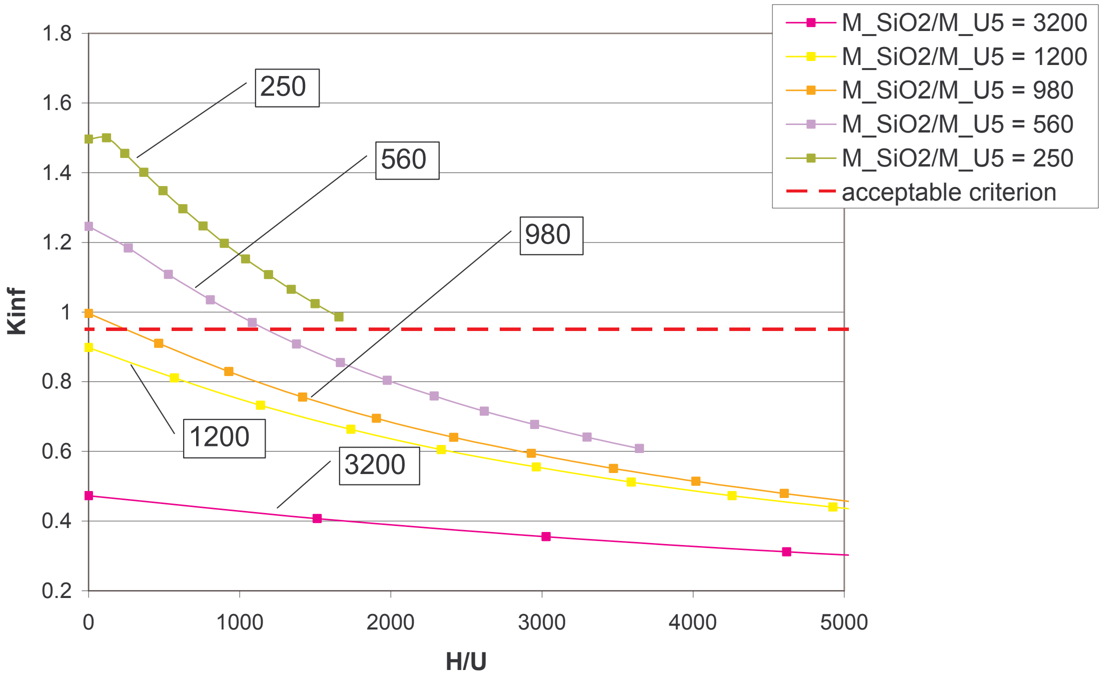

Figure 1. Reactivity of an ^{235}U-SiO$_2$ mixture moderated with water for different SiO$_2$ to ^{235}U mass ratios.

3.2 Matrix of small atomic number nuclei

3.2.1 Comparison of the medium reactivity

The results of the calculations with media of type A_xO_y where the atomic number of A element is small, demonstrate that for A_xO_y to ^{235}U mass ratios around 2000 (corresponding to the proposal USA 04/03), the neutron spectrum for H/U equal to zero is thermal. Thus as it was explained in section 3.1.1, k_∞ is maximum for H/U equal to zero. Moreover, in the thermal region, k_∞ which reads $\sigma_f^{U5}/(\sigma_a^{U5}+\sigma_a^{A}+\sigma_a^{O})$ is constant in a first approximation. For these reasons, the medium reactivity can be compared by comparing the absorption cross-sections of A elements. These cross-sections are reported on Figure 2.

From this analysis, one notes that some elements such as ^{31}P, Mg$_{nat}$, ^{7}Li, ^{9}Be, ^{11}B and C$_{nat}$ are less conservatives than Si. Logically, calculations concerning MgO, LiO$_2$, BeO and CO$_2$ materials show that the acceptable criterion is not respected for M_{AxOy}/M_{U5} equal to 2000.

3.2.2 Example of the beryllium oxide

In the particular case of the beryllium oxide (BeO), the mass ratio that allows to avoid the criticality risk is M_{BeO}/M_{U5} equal to 10000. In accordance with the proposal USA 04/03 that recommends to exclude the beryllium atoms in the mass ratio calculation, this acceptable ratio (M_{BeO}(except Be)/M_{U5}) would be shifted to 6400 which is still above the limit proposed by the US.

Since the infinite multiplication factor with beryllium as a moderator could be a problem, its minimum critical volume was estimated to be compared with the volume of containers for transportation which is 10 to 20 m^3. For M_{BeO}/M_{U5} equal to 3200 and a density of 3.01 g/cm^3, this volume is found to be 1.25 m^3. This volume is conceivable.

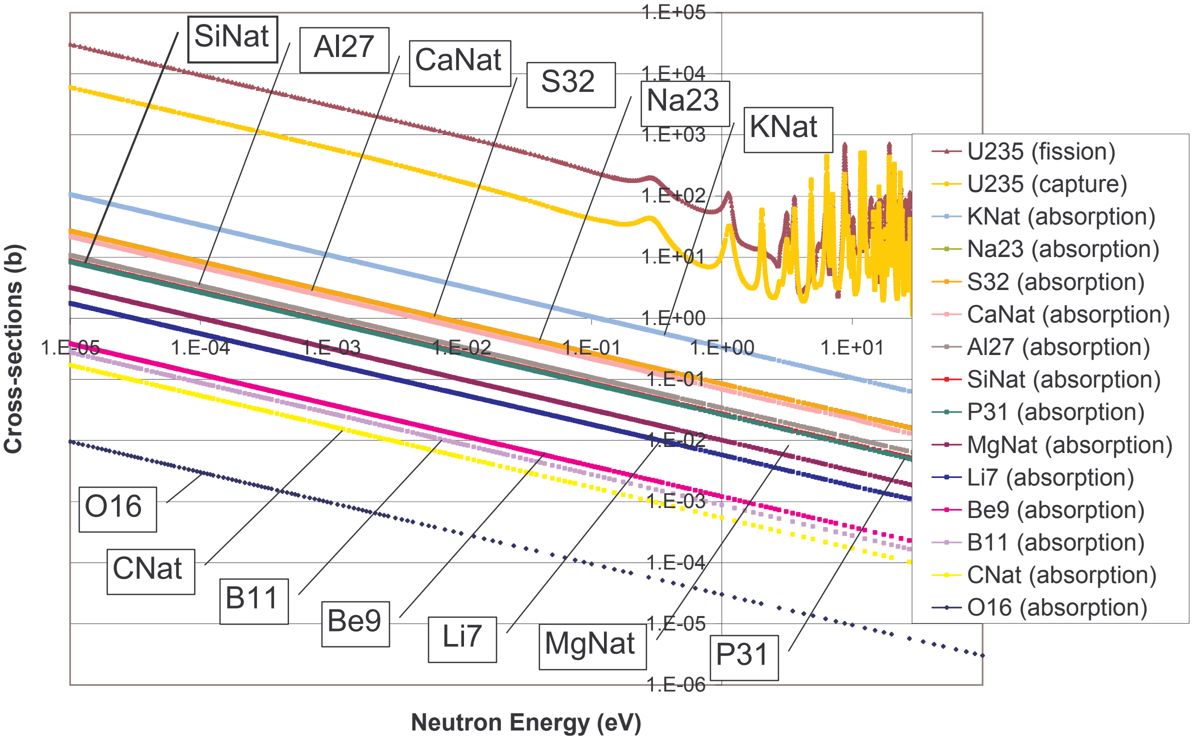

Figure 2. Absorption cross-sections of elements with small atomic number.

3.2.3 Impact of the oxidation degree

Taking into account the fact that the absorption cross-section of ^{16}O is particularly low (see Figure 2), the oxidation degree of A elements, i.e. the weight percentage of ^{16}O in A_xO_y molecule, will impact the reactivity.

From a very restrictive point of view, a matrix, strictly composed of ^{16}O atoms, would then be an enveloping matrix. Calculations estimate that a ^{16}O to ^{235}U mass ratio greater than 100000 is required to respect the acceptable criterion of k_∞.

3.3 Matrix of large atomic number nuclei

3.3.1 Large atomic number and neutron spectrum

Broadly speaking, the larger the atomic number of elements are, the less neutrons are slowed down. Thus, for large M_{AxOy}/M_{U5} (~2000), neutron spectrum can be fast for H/U equal to 0 and the mean absorption energy coincide with the resonance region of A elements. Therefore, the maximum of k_∞ is not necessarily obtained at H/U equal to zero, but rather when neutrons are slowed down with the hydrogen atoms in the thermal domain.

3.3.2 Example of the bismuth

In the particular case of the bismuth (Bi), whose absorption cross-section is lower than the one of the silicon, the maximum of k_∞ for M_{Bi}/M_{U5} equal to 3200, is obtained for a moderator to uranium ratio (H/U) between 1500 and 2000 and it does not respect the acceptable criterion and becomes even supercritical. This point is illustrated on Figure 3.

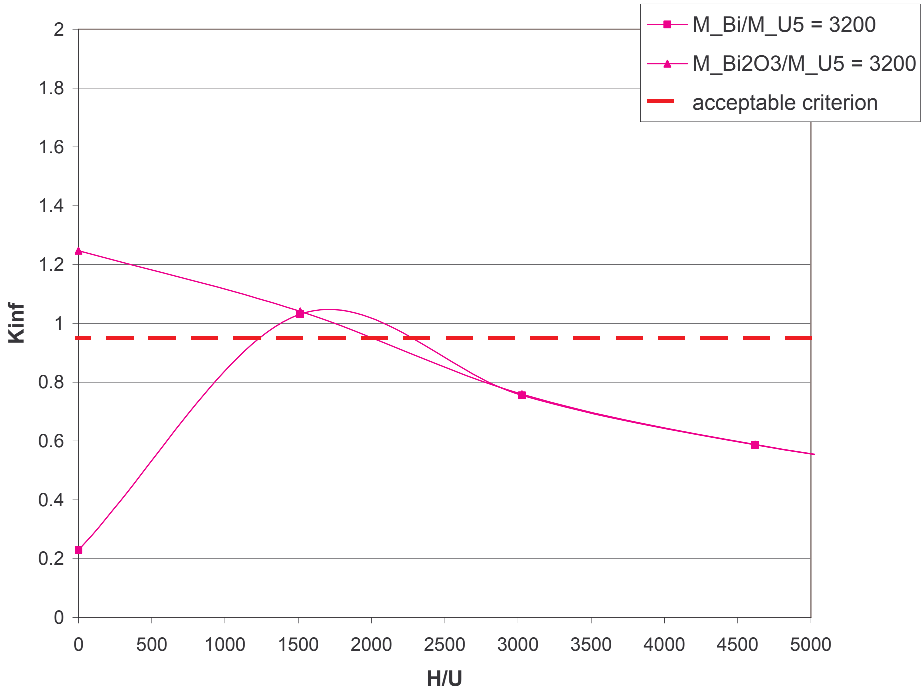

Figure 3. Reactivity of ^{235}U-Bi and ^{235}U-Bi_2O_3 mixtures moderated with water for Bi/Bi_2O_3 to ^{235}U mass ratio equal to 3200.

For the same element, oxygen moderation has the effect of slowing down neutrons to the thermal region and leads to a more reactive situation for a fixed non fissile to fissile mass ratio. As a consequence, the maximum of k_∞ of a ^{235}U-Bi_2O_3 mixture for M_{Bi2O3}/M_{U5} equal to 3200 does not respect the acceptable criterion (see Figure 3).

3.4 “Mixed” matrix

The determination of the maximum reactivity of a “mixed” matrix, composed of several non-fissile elements, with different masses and different absorbing cross-section values, is a complex issue.

Some “mixed” matrix, such as concrete, soils, and ceramic whose compositions are reported in appendix A, have been processed. For “standard” compositions concerning constituent elements of these “mixed” matrixes, no critical situation was found for M_X/M_{U5} equal to 2000. The case of low hydrate or dehydrate concrete has been examined as well. The less concrete is hydrate, the more reactivity is large. However, k_∞ keeps respecting the admissible criterion.

3.5 Conclusions

The determination of an enveloping non fissile to ^{235}U mass ratio, being able to cover any ^{235}U-NF mixture, turns out to be a complicated task. Obtained results, in particular those concerning beryllium oxide and bismuth oxide, demonstrate that the proposal USA 04/03 should be modified to be acceptable.

However, results obtained with "mixed" matrix such as concrete, soils and ceramic don't show any criticality risk. Let us note here, though, that this conclusion does not take into account any additional margin to the admissible criterion ($k_\infty \leq 0.95$) regarding the CRISTAL package validation.

4 PLUTONIUM 239 RESULTS AND TEMPERATURE EFFECT

The case of the fissile isotope plutonium 239 was then examined. Similar calculations to the uranium 235 case were performed. In addition, taking into consideration the fact that ^{239}Pu exhibits a resonance around 0.3 eV, the effect of a temperature increase was studied.

4.1 21°C results

4.1.1 SiO_2 matrix

Figure 4 illustrates the k_∞ dependence as a function of the moderator to plutonium atomic ratio of a ^{239}Pu-SiO_2 mixture for different M_{SiO2}/M_{Pu9}. The acceptable mass ratio is equal to 2400 which is 2 times the mass ratio found in the ^{235}U case. This observation can be explained by the fact that the ^{239}Pu fission cross-section is larger than the ^{235}U one in the thermal region. As a result, the admissible criterion is not respected for M_{SiO2}/M_{Pu9} equal to 2000 and this ratio must be increased at least to 2450.

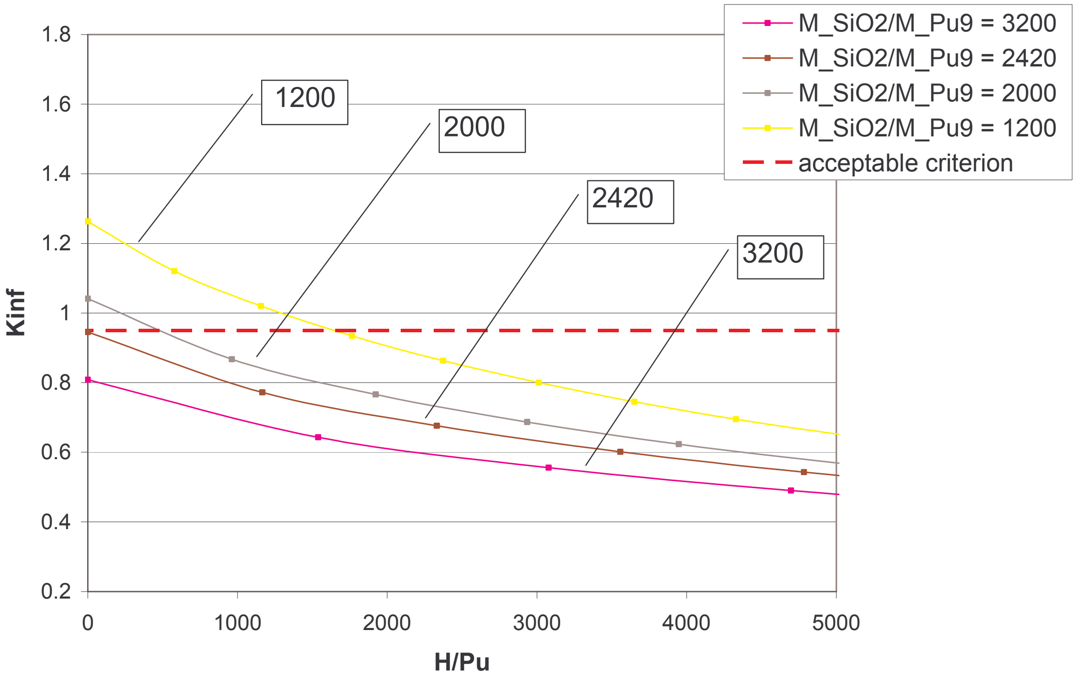

Figure 4. Reactivity of an ^{239}Pu-SiO_2 mixture moderated with water for different SiO_2 to ^{239}Pu mass ratios.

4.1.2 A_xO_y matrix

The comparison between ^{235}U-SiO_2 and ^{239}Pu-SiO_2 mixtures reveals that ^{239}Pu is more restrictive than ^{235}U. Cases of none respect of the acceptable criterion raised in sections 3.2 and 3.3, in the ^{235}U case are therefore also valid in the ^{239}Pu case. This demonstrates again that the proposal USA 04/03 should be modified to be acceptable.

4.1.3 "Mixed" matrix

Main results of calculations on ^{239}Pu-"mixed" matrix mixtures are the following:

- Just as for the ^{235}U case, ^{239}Pu-concrete mixtures, even when concrete is dehydrated, don't show any criticality risk for $M_{concrete}/M_{Pu9}$ equal to 2000.
- Although the acceptable criterion is not respected for a ^{239}Pu-SiO_2 mixture such as M_{SiO2}/M_{Pu9} equal to 2000, it is respected for a ^{239}Pu-soil mixture. Indeed, the soil composition is such as - the number of atoms much less absorbing than silicon (carbon and magnesium) is small enough to have no contribution, - the ^{16}O to other elements atomic ratio is smaller than the ^{16}O to Si atomic ratio in SiO_2 molecules, this leads to a less reactive situation, - these "other elements" include elements (such as aluminum and steel), more absorbing than silicon, which leads to an even less reactive situation.
- However, in the ceramic composition, the absorbing elements proportion is not sufficient to compensate for the low absorption of silicon. Then the acceptable criterion is not respected for $M_{ceramic}/M_{Pu9}$ equal to 2000. Therefore, it is necessary to establish a ratio value larger than 2000. Such a ratio will be defined in the following sections and will take into account a possible increase of the temperature.

4.2 Temperature effect

4.2.1 Principle

Calculations made by several countries (USA, Japan, UK and France) have highlighted a possible positive reactivity temperature coefficient in the case of dilute plutonium solutions (for example, see reference [4]).

The mechanism is the following:

- the neutron spectrum in the thermal energy range shifts to a slightly higher energy due to the temperature rise,
- the ^{239}Pu exhibits a broad resonance around 0.3 eV,
- as a result, the neutron importance increases with the neutron energy in the thermal energy range and the solution has a positive temperature coefficient.

For that reason, we investigated the temperature effect on the reactivity of mixtures containing plutonium.

4.2.2 Temperature effect for a SiO_2 matrix

The temperature effect in a ^{239}Pu-SiO_2 mixture is shown in Figure 5. For high M_{SiO2}/M_{Pu9} (1200, 2000 and 3200), the reactivity clearly increases as a function of the temperature, as in the case of dilute plutonium solutions. Indeed, as it was indicated in section 3.1.1, the neutron spectrum is characteristic of thermal neutrons for these ratios. Then the mechanism described above can operate.

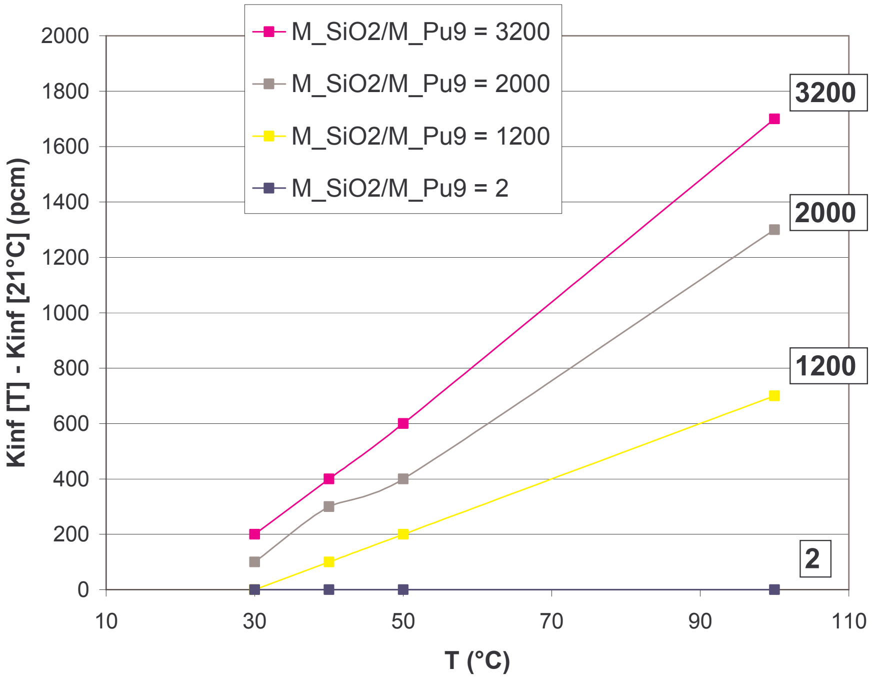

Figure 5. Reactivity of an ^{239}Pu-SiO$_2$ mixture as a function of temperature for different SiO$_2$ to ^{239}Pu mass ratios.

Moreover, the value of the positive temperature coefficient raises with the dilution. It is 20 pcm/°C for $M_{SiO2}/M_{U5} = 3200$, 18 pcm/°C for $M_{SiO2}/M_{U5} = 2000$, 10 pcm/°C for $M_{SiO2}/M_{U5} = 1200$ and 0 pcm/°C for $M_{SiO2}/M_{U5} = 2$. This observation can be explained by the fact that the thermal neutron importance increases with dilution (indeed, dilution goes together with SiO_2 molecules number and then with scattering importance). Since the positive temperature effect is linked to the presence of thermal neutrons, the larger the dilution is, the larger is the temperature effect. This assumption is valid in the studied dilution range.

4.2.3 Temperature effect for “mixed” matrix

A similar positive temperature effect is observed in the case of plutonium diluted in “mixed” matrix. The consequences are the following:

- The acceptable criterion is still respected for a mixture of ^{239}Pu and standard or even low hydrated concrete such as $M_{concrete}/M_{Pu9}$ equal to 2000 at 600°C[iii] (but not in the penalizing case when the concrete hydration falls to zero).

- The acceptable criterion not being respected for a mixture of ^{239}Pu and ceramic such as $M_{ceramic}/M_{Pu9}$ equal to 2000 at 21°C, is a fortiori not respected at 600°C. To guarantee the sub-criticality of this mixture at 600°C, a ratio $M_{ceramic}/M_{Pu9}$ equal to 3000 is needed.

[iii] Concerning temperatures that are likely to be detected in nuclear waste, existing measurements performed on the TN 28 package reveal that temperature can rise up to 500°C in normal conditions, and up to 550-570°C after the 15 minutes thermal test. To be conservative, a maximum temperature of 600°C has been considered for the present study.

Since the ^{239}Pu-ceramic mixture is enveloping the ^{239}Pu-concrete and ^{239}Pu-soil mixtures, a "mixed" matrix to fissile mass ratio greater than 3000 and a temperature inferior to 600°C are therefore required to ensure the criticality-safety.

4.3 Conclusions

In the ^{239}Pu case, in addition to the problems found in the ^{235}U case, section 3, concerning the determination of an enveloping non fissile to fissile mass ratio, the consideration of the positive temperature effect is added.

This effect is all the more important than the fissile material is diluted in the absorbing non-fissile matrix. Nevertheless, the more the fissile material is diluted, the more the mixture reactivity is large. In addition, as it was mentioned in section 2, the results concerning this effect should be considered as qualitative rather than quantitative.

Calculations performed show that mixtures such as the "mixed" matrix to fissile mass ratio is greater than 3000 with the temperature staying below 600°C, don't show any criticality risk. Let us remark here again that no additional margin to the admissible criterion ($k_\infty \leq 0.95$), regarding the CRISTAL package validation, has been taken into account in the determination of this value.

5 OTHER FISSILE ISOTOPES RESULTS: URANIUM 233 AND PLUTONIUM 241

Since the physics principles have been reviewed in details for ^{235}U and ^{239}Pu, calculations concerning ^{233}U and ^{241}Pu focused on the definition of a mass ratio that guarantees the criticality-safety in the case of "mixed" matrix of concrete, soil and ceramic (including the temperature effect in the ^{241}Pu case).

5.1 Uranium 233 results

Results for ^{233}U show that a ^{233}U-X mixture is more reactive than a ^{235}U-X mixture for a given matrix to fissile mass ratio. Nevertheless, for "mixed" matrix under consideration, mixtures such as M_X/M_{U3} equal to 2000 respect the admissible criterion. Therefore, conclusions obtained for ^{235}U are valid for ^{233}U for "mixed" matrix.

5.2 Plutonium 241 results

Results for ^{241}Pu reveal on one hand that a ^{241}Pu-X mixture is more reactive than a ^{239}Pu-X mixture for a given matrix to fissile mass ratio and on the other hand that the temperature effect is less important in the ^{241}Pu case than in the ^{239}Pu one. In order to determine the safe "mixed" matrix to ^{241}Pu mass ratio, a ^{241}Pu-ceramic mixture at 600°C, enveloping ^{241}Pu-concrete and ^{241}Pu-soil mixtures was considered. Calculations indicate that this ratio should be 3500.

6 CONCLUSIONS

The determination of a non-fissile to fissile mass ratio that ensures an infinite multiplication factor lower than 0.95 is not straight forward. Calculation results reveal that this ratio can vary a lot depending on the non-fissile matrix composition under consideration but also depending on the fissile isotope studied. Main results are summarized in Table I.

Table I.

Non-fissile matrix up to 600°C	**Fissile isotopes**				
	^{235}U	**^{239}Pu**	**^{233}U**	**^{241}Pu**	**all**
SiO_2	2000	ND (> 2450)	ND (> 2000)	ND (> 2450)	
A_xO_y type	10000	ND (> 10000)	ND (> 10000)	ND (> 10000)	
^{16}O (envelope)	> 100000	> 100000	> 100000	> 100000	
“mixed” matrix	2000	3000	2000	3500	3500

ND : not determined

Taking into account these observations, three alternatives can be envisaged for the definition of excepted fissile material concerning low concentration of solid fissile material in solid non-fissile material without any total mass or volume limit.

First alternative consists in excluding, in mass calculation, elements whose absorbing cross-section value is inferior to silicon one. This exclusion concerns at least Mg, ^{7}Li, ^{9}Be, ^{11}B, C and ^{16}O. Though, this proposition leads to problems in operational terms of content check. It is then settled.

Second alternative is to increase the non-fissile to fissile mass ratio (now proposed at 2000) to the value found for the enveloping ^{235}U-^{16}O mixture which is greater than 100000. However, the interest of this alternative is very limited.

In a third alternative, one could envisage to limit non-fissile matrix to well defined “mixed” matrix such as concrete, soils or ceramics for which no major problems were found. In this hypothesis, the non-fissile to plutonium mass ratio will have to be increased to 3500 for a temperature below 600°C, in order to take into account the fact that plutonium is more reactive than uranium and that it has positive temperature effect. In addition, discussions should continue to define concrete, soil and ceramic envelope compositions.

Finally, it is important to note that another approach could be envisaged to solve the problem. The idea is to limit the material volume in order to include leakages in the neutron multiplication factor calculation and to be less penalizing. Nevertheless, as it was mentioned section 3.2.2, a sphere of ^{235}U-BeO mixture with M_{BeO}/M_{U5} equal to 3200 is critical as soon as the volume is greater than 1.23 m^3 which is not large enough to exclude any risk of criticality.

In any case, since there are few or no benchmarks for fissile material moderations other than water and therefore the CRISTAL package is not well validated for most studied moderator materials, comparison calculations will have to be performed with different codes and different nuclear data libraries in order to confirm these results and to estimate the necessary additional margin to take into account regarding the CRISTAL package validation.

7 ACKNOWLEDGMENTS

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APPENDIX A

Table II. Concrete compositions

Elements	**Weight percentage**		
	Standard concrete	**Low hydrate concrete**	**Dehydrate concrete**
H_2O	8.9	3	0
Si	33.7	35.79	36.9
Ca	4.4	4.61	4.8
Na	4.6	4.92	5.1
Al	3.4	3.61	3.7
O	45	48.07	49.5

Table III. Soil composition

Elements	**Weight percentage**
C	4.29
O	49
Na	0.68
Mg	0.60
Al	7.1
Si	33
K	1.36
Ca	1.37
Fe	2.60

Table IV. Ceramic composition

Elements	**Weight percentage**
O	52.6
Al	0.5
Fe	0.7
Si	44.8
Ca	1.4