Proceedings of the 17th International Symposium on the Packaging and Transportation of Radioactive Materials
PATRAM 2013
August 18-23, 2013, San Francisco, CA, USA

INFLUENCE ON CRITICALITY SAFETY OF THE PENETRATION OF HYDROCARBONS INTO PACKAGES CONTAINING FISSILE MATERIALS

C. Lenepveu
Institut de Radioprotection et de Sûreté Nucléaire (IRSN)
BP 17 - 92262 Fontenay-aux-Roses Cedex, France
celine.lenepveu@irsn.fr

ABSTRACT
According to the IAEA transport regulation, the nuclear criticality safety demonstration for a package containing fissile materials must take into account water presence in it. But some other materials as hydrocarbons, which could be better neutron moderators than water, could ingress and mix with the fissile materials, in the case of a road accident involving, for example, a portable tank container. The aim of this paper is then to show influence on the reactivity of this kind of scenario. The present work is focused on a study for IF type packages which, in general, contain non irradiated UO_2 fuel in physical form of powder or sintered pellets conditioned in fuel assembly and, therefore, do not need water or containment barriers of high standard level.

First of all, to cover the existing different types of hydrocarbons, a bounding composition is determined based on literature gathered information. This composition is defined by the following chemical formula $CH_{2.1}$ and by a density equal to 0.86 g/cm^3. Criticality calculations show low impact of the slight presence of impurities (oxygen and sulphur) in hydrocarbons.

Comparisons between critical masses and volumes are performed for UO_2 powder (for several ^{235}U enrichment) and between the k_{eff} of an isolated UO_2 assembly (for several ^{235}U enrichment too) considering water, CH_2 and hydrocarbon as moderators. Results show that, without being as efficient as CH_2, the considered hydrocarbon is a better moderator than water.

Finally, the same work is made for packages for UO_2 powder or assemblies. Results show that, for one case, an isolated package can be critical in case of penetration of hydrocarbons. In other cases, the penetration of hydrocarbons can only lead to critical conditions for an array of packages, but not for an isolated package.

This study demonstrates the interest in considering penetration of hydrocarbons into packages containing fissile materials for criticality safety assessment, which potentially might be reflected in the IAEA transport regulation.

INTRODUCTION
Due to the likelihood of water spray or immersion during transportation, the IAEA transport regulation [1] requires to consider water inside packages containing fissile materials for the criticality safety demonstrations. The amount of water taken into account depends on the results of the representative tests of the normal and accident conditions of transport and on the presence or not of water barriers of high standard level. Indeed, this assumption is penalizing

in terms of criticality safety because of the neutron slowing down effect of water which, most of the time, increases the fission probability in fissile media. The proposed study assumes the penetration into packages of other liquids which could be better neutron moderators than water, due for example to the higher hydrogen concentration. In particular, the penetration of hydrocarbons is considered, in the case of a road accident involving, for example, a portable tank container. The impact of this scenario is analyzed by comparing the reactivity of a system composed of fissile materials moderated by water, hydrocarbons or polyethylene (CH_2) which is actually considered as the best hydrogenous moderator for criticality safety. First of all, hydrocarbons bounding composition was assessed based on literature review. Then comparison exercise was first done on simple geometric configurations (critical masses and volumes, and k_{eff} for isolated PWR assembly) and then on real packages configurations. In order to be realistic as much as possible, the packages chosen in this study (IF type) do not include multiple high standard water barriers. Thus, the concerned fissile materials are essentially fresh uranium oxide in physical form of powder or sintered pellets conditioned in fuel assembly.

HYDROCARBONS

Hydrocarbons are organic compounds used in particular as fuel for transportation. Given that they are essentially made of hydrogen and carbon, their chemical formula can be written as follows: $(CH_y)_n$. In order to determine a bounding composition, considering criticality aspects, the concentration of hydrogen (g/cm^3) has to be maximised, specifically compared to water. This can be simply evaluated by the following formula:

$\frac{[H]_{hydrocarbon}}{[H]_{water}} \approx d_{hydrocarbon} \times \frac{H/C}{12 + H/C} \times \frac{18}{2}$, where $d_{hydrocarbon}$ is the hydrocarbon density (g/cm^3) and H/C is the ratio between hydrogen and carbon atoms in the hydrocarbon (equivalent to "y" in the chemical formula). According to literature [2], the maximal density and H/C ratio for hydrocarbons used for transportation are equal respectively to 0.86 g/cm^3 and 2.1 (corresponding to diesel oil). This leads to a $\frac{[H]_{hydrocarbon}}{[H]_{water}}$ ratio equal to 1.15, which means that the concentration of hydrogen in this hydrocarbon is higher than in water. It can be noticed in another hand that hydrogen concentration ratio between diesel oil and CH_2 $\frac{[H]_{hydrocarbon}}{[H]_{CH_2}}$ is equal to 0.93.

Hydrocarbons might also contain some impurities as oxygen and sulphur, leading to the following chemical formula: $(CH_yO_zS_t)_n$, with $z \leq 0.03$ and $t \leq 0.02$. Given the low concentration of these impurities, their presence in hydrocarbons has no significant impact on the fissile media reactivity as shown in calculations presented in figure 1 for an isolated fresh UO_2 PWR assembly. The different observed behaviours for higher concentrations of impurities can be explained by the fact that sulphur has a capture cross-section higher than oxygen, meanwhile their diffusion cross-sections are very close.

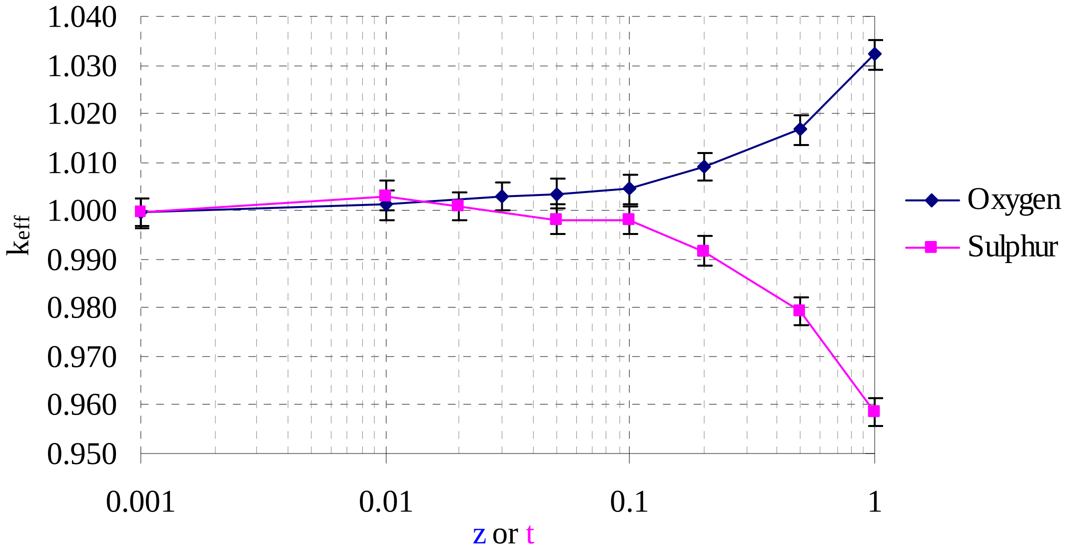

Figure 1. Impact of the presence of impurities in hydrocarbons ($CH_{2.1}O_zS_t$) on the reactivity of an isolated UO_2 PWR assembly

CALCULATION METHODS

Calculations are performed with APOLLO2-MORET 4 and APOLLO2-SN calculation schemes from the CRISTAL V1 package [3], using JEF 2.2 nuclear data library.
The fissile materials considered in this paper are those usually encountered in IF type packages, corresponding to non irradiated uranium oxide in physical form of powder (with a maximal density considered equal to 11 g/cm^3) or sintered pellets conditioned in PWR fuel assembly. The uranium enrichment in ^{235}U isotope is considered variable.

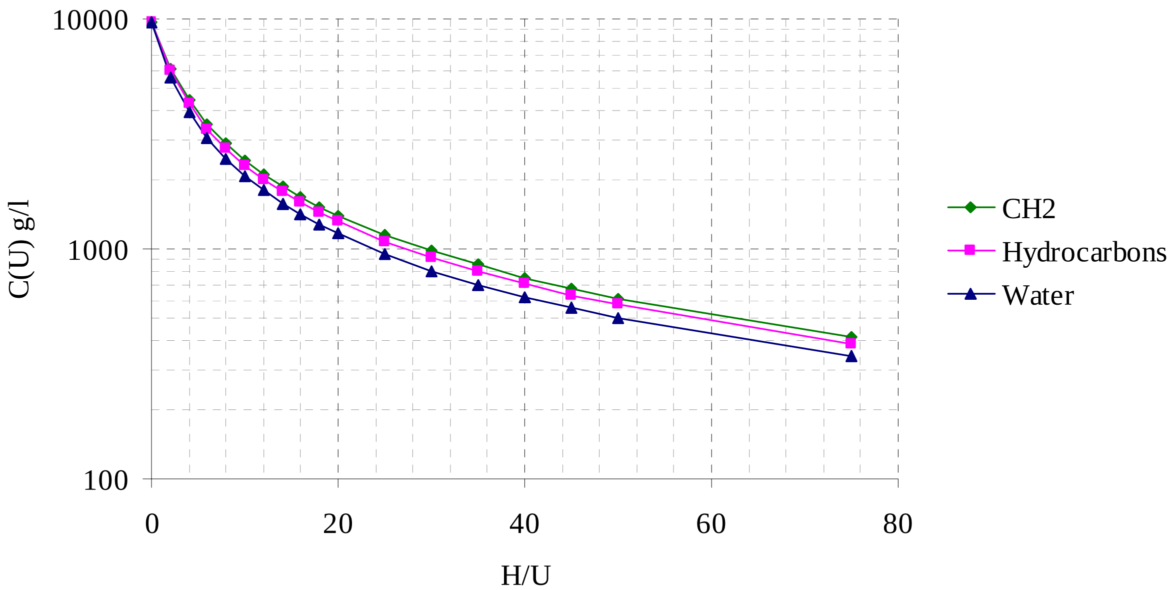

Figure 2. Comparison of dilution laws for UO_2 powder moderated by water, hydrocarbons or CH_2

Three types of moderator are taken into account: water, hydrocarbons as defined here above ($CH_{2.1}$ with a density equal to 0.86 g/cm^3) and polyethylene (CH_2 with a density equal to 0.96 g/cm^3). CH_2 is classically taken into account as moderator in criticality safety demonstrations to cover all the hydrogenous materials, in particular those showing a higher hydrogen concentration than water. CH_2 and hydrocarbons are modelled with cross-sections taking into account bonds between hydrogen and carbon atoms.

For UO_2 powder, hydrocarbons are supposed intimately mixed with fissile materials, in the same way that water. In this regard, comparison of dilution laws (based on the volume addition law) used in this study for the three considered moderators is given in figure 2 and already shows the penalizing nature of CH_2 and the selected hydrocarbon composition.

SIMPLE GEOMETRIC CONFIGURATIONS

As a first step, the comparison of the three defined moderators is performed for simple geometric configurations, that is to say UO_2 powder sphere or UO_2 PWR assembly reflected by 20 cm of water.

<u>UO_2 powder:</u>

The comparison of minimal critical masses and radii shows that the lower values are obtained considering first CH_2, then hydrocarbons and finally water, as shown in figure 3, which is consistent with the comparison of the dilution laws.

Results show that discrepancies for minimal critical mass are close to 20 % between hydrocarbons and water and 30 % between CH_2 and water, whatever the uranium enrichment. For minimal critical spherical radius, discrepancies are close to 10 % between hydrocarbons and water and 15 % between CH_2 and water, whatever the uranium enrichment except for very high enriched uranium. Actually, for a 100 % enrichment, the minimal critical spherical radius is obtained for dry configuration considering water, unlike CH_2 and hydrocarbons. Discrepancy is then around 5 % between hydrocarbons and “water” and around 10 % between CH_2 and “water”.

Greater discrepancies on the minimal critical mass are observed for low enrichment when geometry is imposed, especially when the radius of a cylinder of fissile materials is close to the minimal critical radius.

Other calculations show that taking into account CH_2 or hydrocarbons as reflector instead of water leads to a slight decrease of the critical values (up to 5 %).

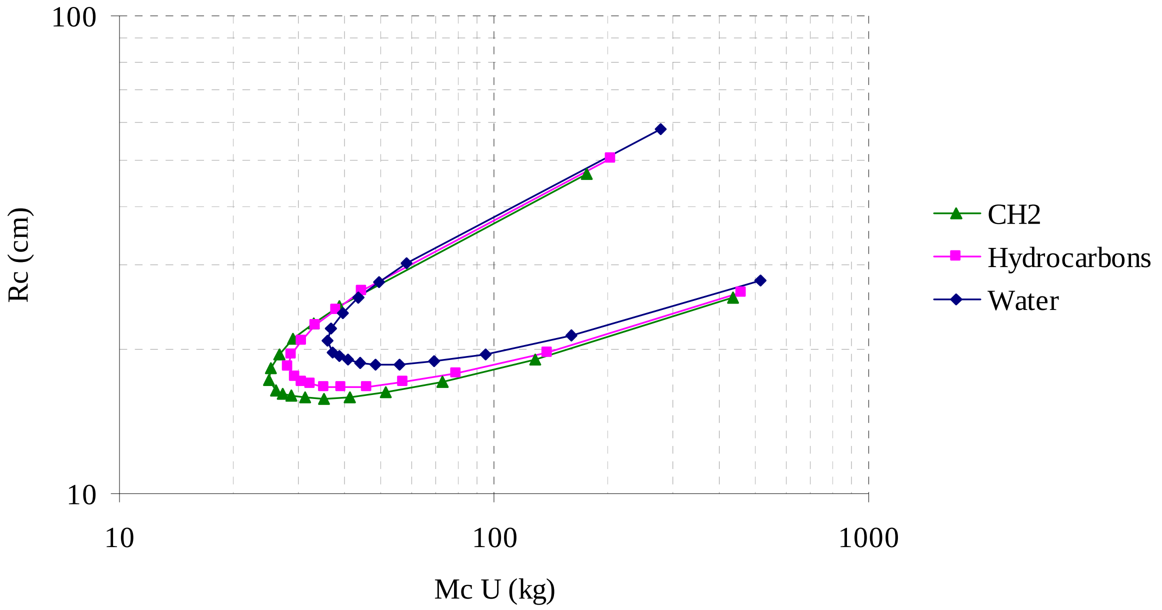

Figure 3. Comparison of critical masses and radii for UO_2 powder (5 % enrichment) moderated by water, hydrocarbons or CH_2 (for spherical geometry)

UO$_2$ PWR assembly:

Comparison of k_{eff} of an isolated 17x17 PWR fuel assembly moderated with each of previous moderators points out that the highest value is obtained considering first CH_2, then hydrocarbons and finally water, as shown in figure 4. Relative discrepancies are around 5 % between hydrocarbons and water and around 8 % between CH_2 and water. In terms of reactivity, for 5 % enrichment, the observed discrepancies are around 5300 pcm between hydrocarbons and water and around 7600 pcm between CH_2 and water.

Complementary calculations show that taking into account CH_2 or hydrocarbons as reflector instead of water leads to a slight increase of the k_{eff} values (up to 800 pcm).

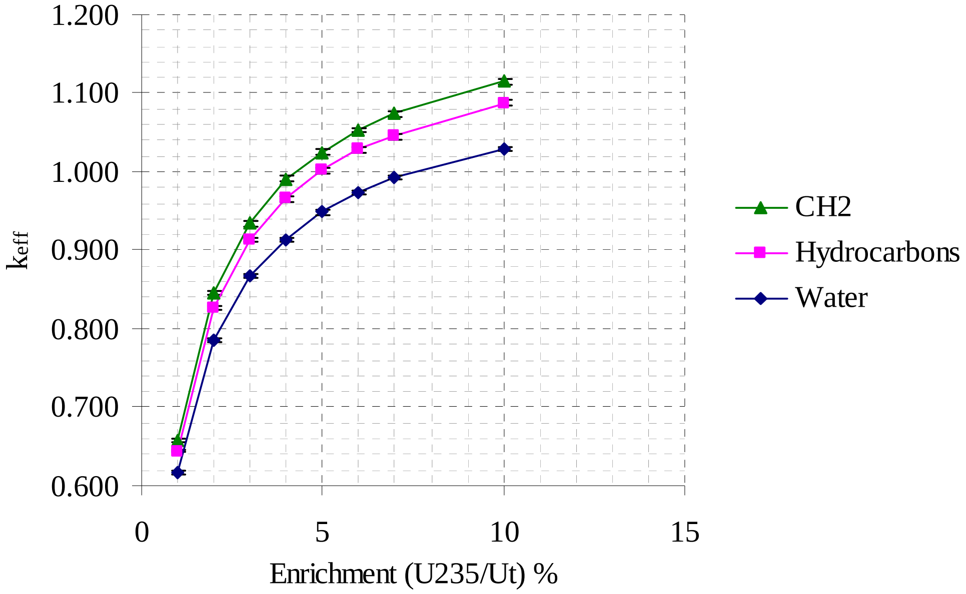

Figure 4. Comparison of k_{eff} values for an isolated UO$_2$ PWR assembly moderated by water, hydrocarbons or CH$_2$

IF TYPE PACKAGES CONFIGURATIONS

According to the previous paragraph, neutron moderation by hydrocarbons leads to increasing the fissile media reactivity compared to moderation by water. So, the purpose of this paragraph is to evaluate the impact of the penetration of hydrocarbons for real configurations.

Packages containing UO$_2$ powder (5 % enrichment)

This kind of packages is usually constituted of one (or several) cylindrical cavity containing UO_2 powder. Neutron absorbing material is usually around the cavity to reduce neutron interactions between packages (and eventually between cavities).

The studied package is constituted of one cavity containing a certain amount of uranium oxide. For the chosen content, the maximal evaluated k_{eff} for an isolated damaged package filled with water is low (less than 0.85). Consequently, even if it increases significantly reactivity, the moderation by hydrocarbons does not lead to a critical situation (discrepancy around 10000 pcm considering CH_2). On the other hand, the maximal evaluated k_{eff} for an infinite array of damaged packages filled with water being close to 0.95, the penetration of hydrocarbons leads to a k_{eff} higher than 1. To reach an acceptable k_{eff} value without changing the hypotheses on the content, in particular the fissile materials mass, the number of packages has be reduced (see figure 5), leading to increasing the criticality safety index (CSI) from 0 ($N = \infty$) to around 7 ($N \approx 7$).

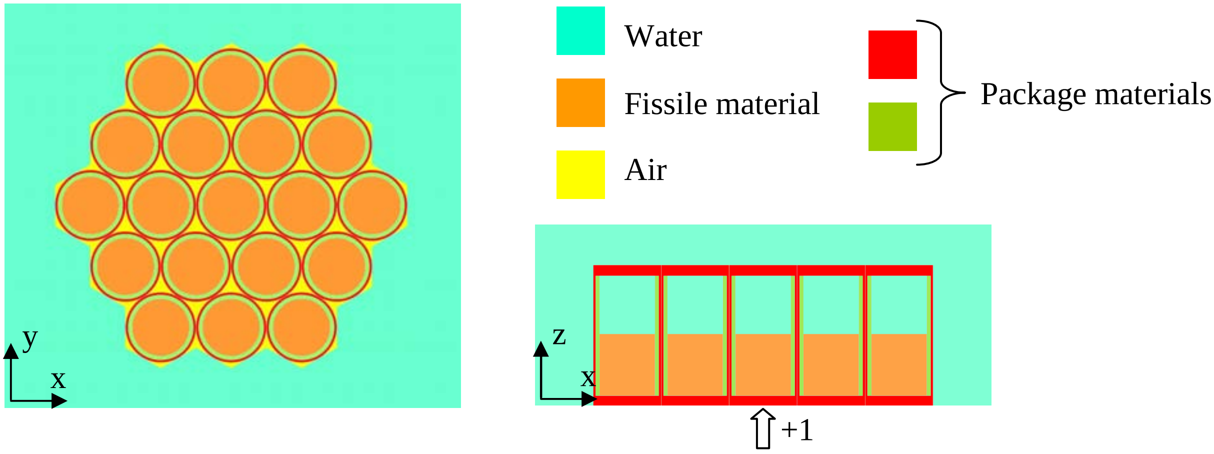

Figure 5. Array of 5N packages containing UO_2 powder

The loading of a higher quantity of fissile materials, keeping nevertheless the isolated package filled with water in a subcritical configuration, will reduce the margins towards the admissibility criteria, and thus could lead to a critical situation in the case of hydrocarbons penetration. As a matter of fact, this situation has been encountered for other studied packages.

Package containing fresh UO_2 PWR assembly (5 % enrichment)

This kind of package is usually constituted of one or two long compartments for the assembly(ies).

The case considered here is able to carry two fuel assemblies. For the chosen content, the nominal k_{eff} of an isolated damaged package filled with water is less than 0.9 and about 0.935 for an array of damaged packages. Taking into account penetration of hydrocarbons does not reconsider the sub-criticality of the isolated package (discrepancy around 7000 pcm, higher than the evaluated one for an isolated assembly) but implies to decrease the number of packages for the array configuration to comply with the admissibility criteria. This would lead for this specific case to increasing the CSI from 0.625 (N = 80) initially to around 12.5 (N = 4), for an admissibility criteria fixed at 0.98 for the accident conditions of transport, as shown in figure 6.

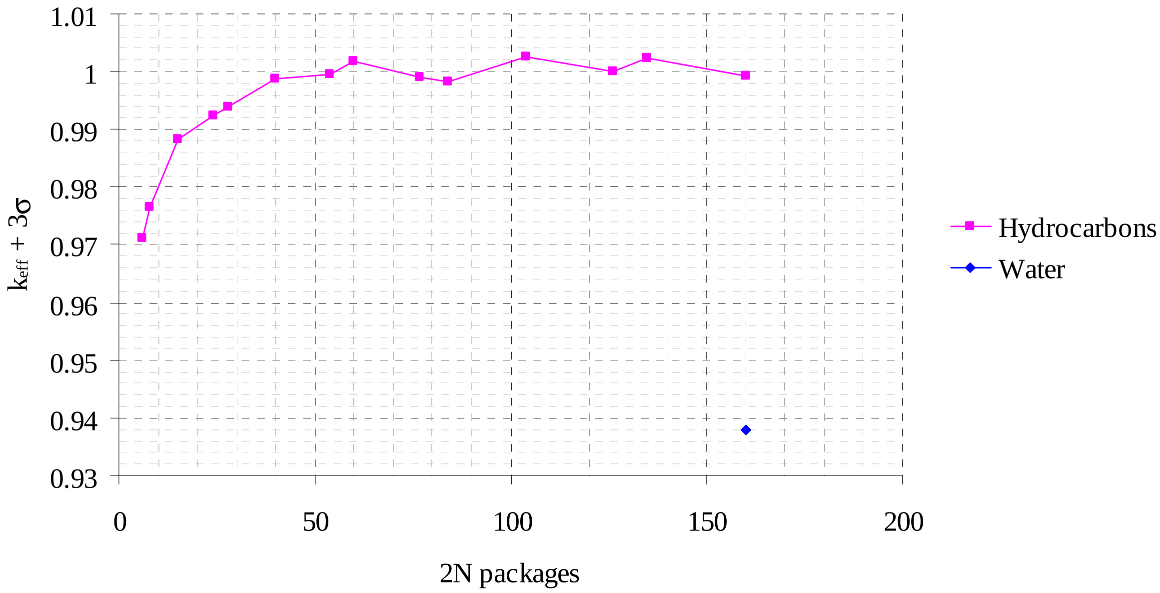

Figure 6. Evaluation of the admissible number of packages containing two UO_2 PWR assemblies in case of penetration of hydrocarbons

CONCLUSION

From this study, it appears that some hydrocarbons can be better neutron moderators than water, depending on their density and H/C ratio. The impact of their penetration into packages containing fissile materials in accident conditions of transport is not negligible in terms of criticality aspects. Actually, it can lead to decreasing the admissible number of packages or even the allowed quantity of fissile materials, depending in particular on the initial margins between the k_{eff} evaluated considering water and the admissibility criteria. Yet, this study is explorative and concerns a selected bounding hydrocarbon and a limited type of fissile materials and packages.

It might be a base of discussion to eventually make the IAEA regulation evolve in order to take into account hydrocarbons penetration into packages containing fissile materials under specific conditions, for example for packages without specific constraints on water tightness (in particular IF and A types).

Finally, although this study is about hydrocarbons penetration into non watertight packages, it could also be interesting for other kinds of packages and scenarios, in the case of penetration of oil pomp during outlet operation for example.

REFERENCES

[1] Regulations for the Safe Transport of Radioactive Material - 2012 Edition, Specific Safety Requirements No. SSR-6, IAEA Safety Standards

[2] Techniques de l'Ingénieur, traité Génie Mécanique, “Les carburants et la combustion”, Jean-Claude GUIBET (BM 2 520)

[3] “CRISTAL V1: Criticality Package for Burn up Credit calculations”, October 20th - 24th 2003, ICNC 2003, Tokaï-Mura, Japan, J.M. Gomit, P. Cousinou, C. Diop, G. Fernandez de Grado, F. Gantenbein, J.P. Grouiller, A. Marc, D. Mijuin, H. Toubon