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Impact of fissile minor actinides on the criticality safety of transport of fissile material

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Abstract

In the current IAEA regulation for the safe transport of radioactive material [1] and all previous editions of this regulation, the fissile materials are defined as the materials containing any of the 4 following nuclides ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu. It is well-known that other nuclides are able to sustain a fission chain reaction and that some of them have a lower subcritical mass limit than the four usual fissile nuclides, for example ^{242m}Am, ^{243}Cm and ^{251}Cf. In cases where sufficient quantities of these nuclides would be present in a material, the guidance of the regulation [2] advices to perform a criticality safety assessment as per the requirements of paragraphs 673-686.

When these minor actinides are not separated from fission products, uranium or plutonium (for example, in irradiated UOX or MOX fuel), they have no impact on nuclear criticality safety and the formulation of SSR-6 is acceptable. However, if these fissile minor actinides come from an enhanced separation process, they could have a significant impact on reactivity even for limited quantities (several grams), in particular for some nuclides.

Moreover, even if many minor actinides have a significant subcritical mass limit (much higher than ^{235}U), when these nuclides are irradiated (for example, as experimental fuel), it becomes more complicated to justify subcriticality if their initial mass is important. Indeed, during irradiation, neutron capture by these nuclides, for example ^{241}Am, will produce fissile minor actinides with very low subcritical mass limits.

Introduction

Paragraph 222 of the current IAEA regulation for the safe transport of radioactive material [1] defines fissile isotopes as the 4 following nuclides ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu. Indeed, theses nuclides are the main fissile nuclides encountered during the transport of radioactive material and in nuclear facilities.

Nevertheless, as stated by paragraph 222.3 of the guidance of the regulation [2], other nuclides are fissionable[1], or even fissile[1], and some of them have a lower subcritical mass limit than the four usual fissile nuclides. These nuclides are the following: ^{237}Np, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{247}Cm, ^{249}Cf and ^{251}Cf. When these minor actinides are not separated from fission products, uranium or plutonium (for example, in an irradiated UOX or MOX fuel), they are present in small quantities compared to those of uranium and plutonium, which is covered by the margins provided by the calculation assumptions (such as not taking the fission products into account).

However, research are being conducted on enhanced separation process to extract some minor actinides from irradiated fuel, the objective being to optimize final disposal of waste and reduce their amount by transmutation or fission. In this field, research fuel pins enriched in minor actinides are produced in order to study their behavior in a reactor. During the transport of such material, due to the presence of these nuclides in sufficient quantities, the guidance of the regulation [2] advices to perform a criticality safety assessment as per the requirements of paragraphs 673-686.

The goal of this paper is to provide comments on the application of paragraph 222.3 of the guidance [2] on the basis of comparison of the minimal critical mass of several actinides and depletion studies of minor actinides.

1 Comparison of the minimal critical mass

In the past, many studies, such as [4] to [6], have been carried out to estimate the minimal critical mass of fissionable actinides. Table 1 gives metallic critical masses (unmoderated material reflected by 30 cm of stainless steel) and minimal critical masses in solution (moderation by water and reflection by 20 cm of water) for the four usual fissile isotopes and the minor actinides listed in paragraph 222.3 of [2]. Except those of ^{249}Cf and ^{251}Cf which are taken from [6], all values of table 1 are extracted from [4].

As stated by the guidance [2], some of these values, especially for the heaviest isotopes, require special attention due to “the lack of critical experimental data, the limited knowledge of the behavior of these nuclides under different moderator and reflection conditions and the uncertainty in the cross-section data”.

It can be noted that all the metallic critical masses presented in table 1 are higher than 2 kg. In actual industrial facilities, the presence of such amount of pure neptunium, americium, curium and californium is not usual (and is very unlikely for the heaviest elements). However, the minimal critical mass, when it exists, is much lower than the metallic critical mass (for example, 25 g for ^{251}Cf). Hence, the risk of criticality during transport of minor actinides is mainly present for fissile

[1] ‘Fissionable’ nuclides are those “capable of undergoing fission” [3]. Among these nuclides, ‘fissile’ nuclides are those “capable of undergoing fission by interaction with slow neutron” [3]. Some nuclides (such as ^{238}Pu) may be fissionable not fissile, meaning that they are capable of undergoing fission after capturing a fast neutron but not a slow neutron.

nuclides and not for fissionable nuclides, in other words ^{242m}Am, ^{243}Cm, ^{245}Cm, ^{247}Cm, ^{249}Cf and ^{251}Cf. With regard to its high minimal critical mass, which is around 2 kg, ^{247}Cm will not be considered thereafter.

Table 1 Critical masses

Isotope	**^{233}U**	**^{235}U**	**^{239}Pu**	**^{241}Pu**	**^{237}Np**	**^{238}Pu**	**^{240}Pu**	**^{242}Pu**	**^{241}Am**
Metallic critical mass (g)	6032	17079	4655	5309	48073	4421	21694	42164	40066
Minimal critical mass (g)	553	779	498	267	NF	NF	NF	NF	NF
Isotope	**^{242m}Am**	**^{243}Am**	**^{243}Cm**	**^{244}Cm**	**^{245}Cm**	**^{247}Cm**	**^{249}Cf**	**^{251}Cf**	
Metallic critical mass (g)	4505	122540	2758	16007	2657	3600	2390	2270	
Minimal critical mass (g)	23	NF	264	NF	47	2104	60	25	

NF: non fissile.

In the regulation [1], a material is defined as fissile material if the total mass of the four usual fissile nuclides is higher than 0.25 g (paragraph 222(c)). Fissile material and packages containing fissile material are classified as FISSILE if the total mass of the four usual fissile nuclides is higher than 2 g (paragraph 417(d)). These two limits are respectively around 2000 and 250 times lower than the ^{239}Pu minimal critical mass[2]. When applying the same coefficients for the fissile minor actinides listed previously, the maximal masses corresponding to the requirements of the paragraphs 222 and 417 would be the following.

Table 2 Limit masses for minor actinides

Isotope	**^{242m}Am**	**^{243}Cm**	**^{245}Cm**	**^{249}Cf**	**^{251}Cf**
Limit corresponding to paragraph 222(c) (g)	0.012	0.132	0.020	0.030	0.013
Limit corresponding to paragraph 417(d) (g)	0.09	1.06	0.19	0.24	0.10

Thus, considering the same principles as the IAEA regulation, a material would be defined as a fissile material when containing more than about 10 mg of these nuclides. Similarly, a fissile material would be classified as FISSILE when containing more than about 0.1 g of some of these nuclides. These very low mass limits demonstrate the need to perform a specific criticality safety analysis

[2] ^{241}Pu is not considered because its presence is linked to the presence of ^{240}Pu which has a high capture cross section in thermal spectra. Thus, the penalizing isotopic composition of plutonium is generally 100 % ^{239}Pu.

regardless of the quantities of these minor actinides in a packaging when they have been separated from uranium, plutonium and fission products.

2 Irradiation of minor actinides

As mentioned above, some minor actinides have only a critical mass in the metallic form (they are 'fissionable' but not 'fissile'), so these actinides would not lead to a risk of criticality due to the unlikelihood to load such large quantities of these isotopes in a package. However, their irradiation may create non-negligible amount of fissile nuclides by neutron capture and decay. For this reason, the present paragraph investigates the irradiation of minor actinides.

2.1 Calculation method

VESTA [7] is a generic Monte-Carlo depletion interface used to calculate the evolution of a material subjected to radiation. This interface uses a transport code (MCNPX [8] for this study) coupled with a depletion code (ORIGEN2 [9] for this study). The depletion code iterates between a steady state transport calculation and a material evolution through the Bateman equations. The material composition in every zone is considered spatially constant when determining the particle flux in the steady state transport calculation and the neutron flux (and the associated spectrum) is assumed to be constant in space and time during a time step in the evolution calculation.

The beta version 2.2.0h of VESTA, used for the present study, also enables the use of a second approach considering tabulated burn-up dependent activation libraries instead of a transport calculation. These libraries contain all the data required to perform a depletion calculation without the need for new transport calculations. The standard method previously described is actually used to generate these libraries. The neutron fluence integrated over time is used instead of the burn-up, which enables depletion calculations on non-fissile materials. The advantage of this approach is to allow calculating the evolution of a non-fissile actinide sample subjected to irradiation in a reactor.

As stated above, VESTA calculations using the transport code MCNPX are first performed in order to generate activation libraries. Two models are considered for this study: a Sodium-cooled Fast Reactor (SFR) assembly and a Pressurized Water Reactor (PWR) assembly. The SFR is loaded with UO_2-PuO_2 fuel pins[3] irradiated up to a burnup of 124 GWd/tHM. The PWR is loaded with 17x17 UO_2 assemblies with 3.7% enriched uranium irradiated up to a burnup of 70 GWd/tHM. All the cross section data have been taken from JEFF3.1.1 [10].

Then, the activation libraries containing the neutron fluence in the SFR fuel and the PWR fuel at every burn-up step are used in order to perform evolution calculations on different samples (no cooling time is considered). The corresponding flux values are used for the irradiation. The samples studied are either a minor actinide (^{241}Am, ^{243}Am, ^{237}Np, ^{242}Cm or ^{243}Cm) or natural uranium (0.72%

[3] The plutonium mass content (Pu/(U+Pu)) is equal to 19,3 %wt, with a fissile plutonium mass content (^{239}Pu+^{241}Pu)/Pu equals to 84.1 %wt. The uranium is natural uranium (0.72%wt ^{235}U).

^{235}U) or ^{232}Th. With regard to ^{238}Pu, its presence is taken into account in paragraph 675 of the regulation [1], therefore this isotope is not considered hereafter. Every sample is considered in the metallic form and its volume is 1 cm^3. The composition of these samples after irradiation is analysed in paragraph 2.2.

Finally, the composition of the samples at different steps of irradiation is used to perform criticality calculations (no fission products are considered, and the composition of the actinides does not take into account correction factors). The infinite multiplication factor (Kinf) is calculated by the transport code APOLLO [4] for different moderation ratios (H/X, X being the initial atomic concentration in the sample). The Kinf values are analysed in paragraph 2.3.

This article intends to provide general tendencies on the behavior - in terms of criticality safety - of minor actinides during irradiation. For this purpose, considerations about Monte-Carlo calculation statistical uncertainties are omitted as well as considerations about computer code validation (no assessment of a possible bias). Thus, the results presented and discussed in this article should be seen as illustrative critical values rather than “true” critical values.

Furthermore, this method assumes that the sample has no influence on the neutron spectrum, which is realistic for small samples only. Besides, the approach considered may lead to errors for evolution studies in a PWR, in which the neutron mean free path is much shorter than in a fast spectrum reactor as an SFR.

2.2 Irradiation results

The figure 1 below presents the evolution of the three main fissile isotopes produced by the irradiation in an SFR of two non-fissile nuclides, ^{241}Am and ^{242}Cm, with a respective mass of 13.7 g and 13.4 g corresponding to 1 cm^3. The burnup provided represents the burnup of the UO_2-PuO_2 fuel pins of the SFR.

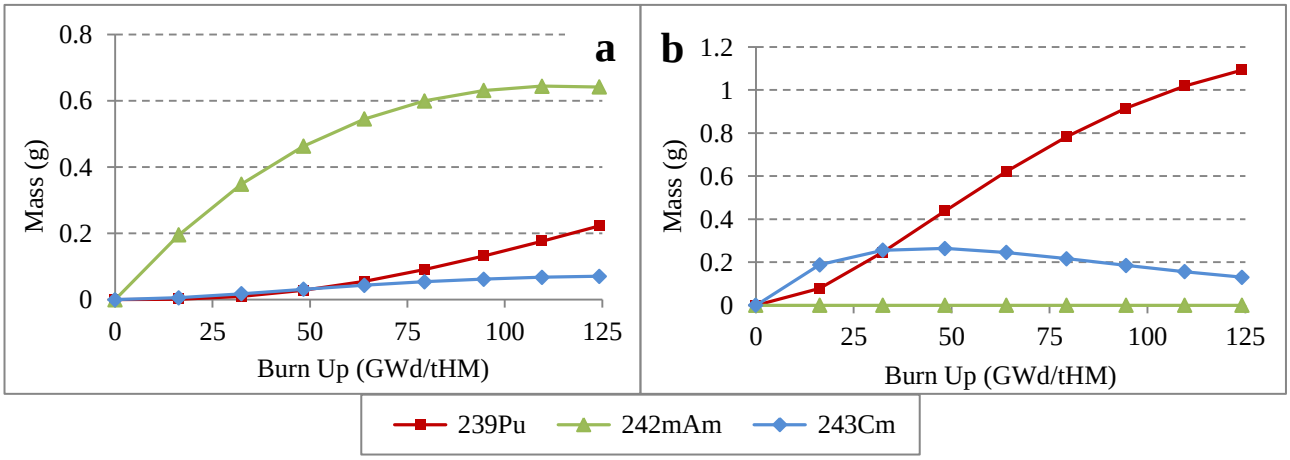

Figure 1 Production of fissile isotopes from ^{241}Am (a) and ^{242}Cm (b) after irradiation in an SFR

The figure 1-a shows that, even for a low burn-up, the irradiation of a small amount of ^{241}Am leads to a mass of ^{242m}Am which is higher than the limits presented in table 2. Moreover, the irradiation of ^{242}Cm leads to the production of ^{239}Pu which can represent more than 5 % of the irradiated sample mass for an irradiation higher than 75 GWd/tHM. These results highlight the importance to perform a specific criticality safety analysis in presence of minor actinides separated from uranium, plutonium and fission products, even for fissionable actinides which are not considered as fissile nuclides before irradiation.

2.3 Kinf results

Figures 2 and 3 show the evolution of the Kinf for a fissile sample (^{243}Cm) or a non-fissile sample (^{241}Am) as a function of the moderation ratio, for different fuel burnups (the irradiation spectrum corresponding to UO_2-PuO_2 pins for the SFR and UO_2 rods for the PWR).

As shown in paragraph 1, ^{243}Cm is a fissile nuclide[1] and has a low minimal critical mass. When irradiated in a fast (SFR) or a thermal (PWR) neutron spectrum, its Kinf decreases (see Fig. 2) as a function of the burnup. This is due to the decrease of the amount of ^{243}Cm by fission and relatively low creation of fissile nuclide by capture and decay (fission cross sections are higher than capture).

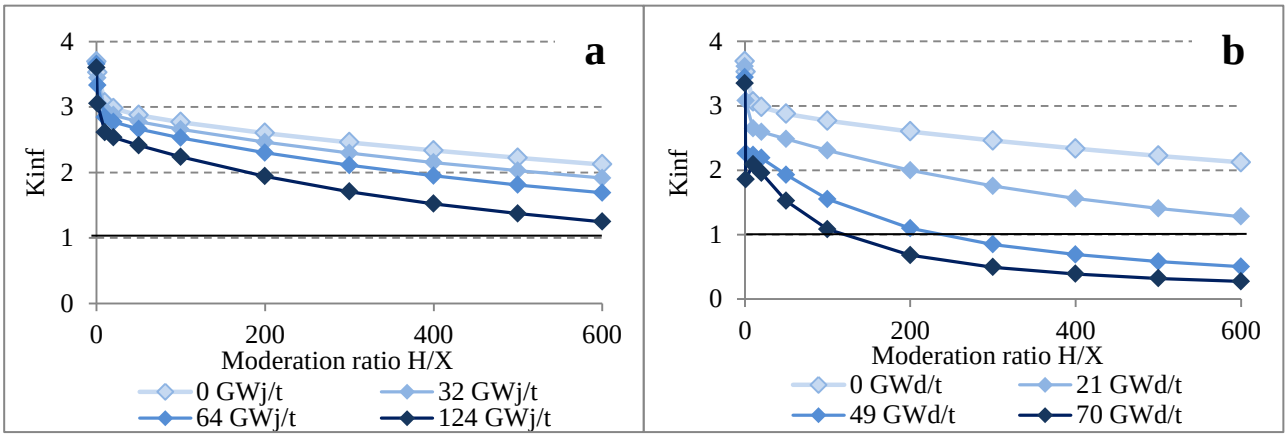

Figure 2 Kinf values of ^{243}Cm irradiated in an SFR (a) or a PWR (b)

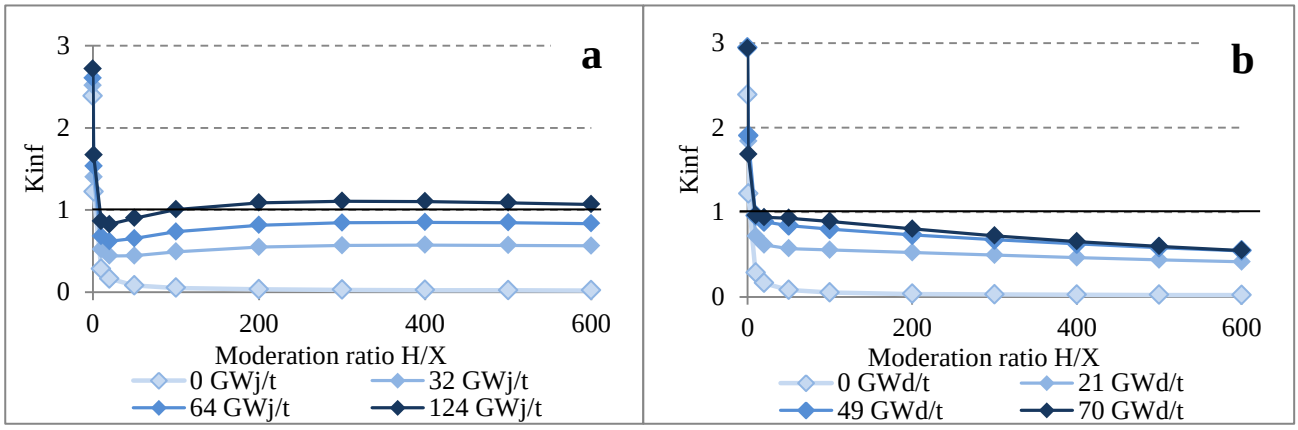

Figure 3 Kinf values of ^{241}Am irradiated in an SFR (a) or a PWR (b)

Unlike ^{243}Cm, pure ^{241}Am is not a fissile nuclide (fissionable nuclide), but the sample may become fissile when irradiated in an SFR (see Fig. 3-a). When irradiated in a PWR, it remains subcritical at thermal spectrum according to the calculations (see Fig. 3-b). However, considering uncertainties in the composition of the irradiated actinide and the lack of validation of calculation tools for minor actinides, subcriticality might not be certain. It is to be noted that the Kinf increases with irradiation, due to the production of fissile isotopes. The same conclusions applies to irradiation of a sample of ^{243}Am.

From the figure 3, it can be noted that the Kinf of a ^{241}Am sample, irradiated or not, is higher than 1 without moderation. Indeed, as stated in paragraph 1, ^{241}Am metal is fissionable. However, in such case, the amount of material needed to be critical is so huge that it is not realistic for a transport package (about 60 kg of ^{241}Am). Therefore, the present study focuses on the reactivity of the moderated samples.

Figure 4 presents the Kinf values after irradiation in an SFR or a PWR of different actinide samples which are not fissile before irradiation. Natural uranium and ^{232}Th are also presented in order to make comparison.

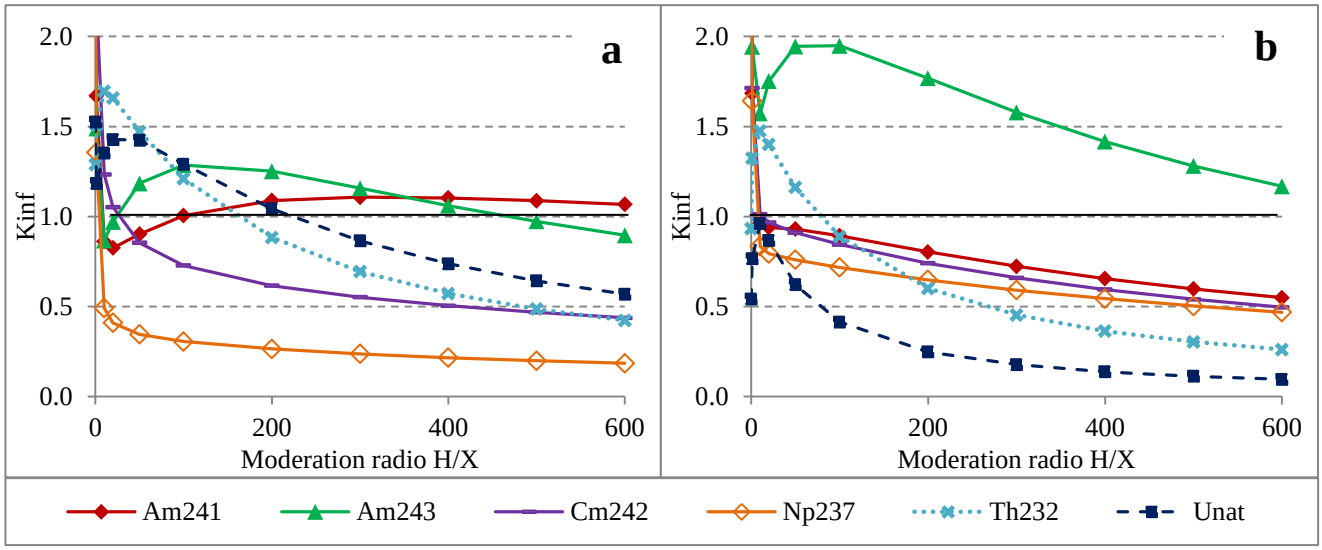

Figure 4 Kinf values of several isotopes after irradiation in an SFR (124 GWd/tHM) (a) or a PWR (70 GWd/tHM) (b)

From figure 4, the following conclusions can be drawn:

- As it is well known, natural uranium becomes fissile after irradiation in an SFR but remains non-fissile after irradiation in a PWR (though in this study it is near critical for H/X around 10). This result explains the paragraph 222(b) of IAEA regulation [1]: “natural uranium or depleted uranium that has been irradiated in thermal spectrum reactors is excluded from the

definition of fissile material";

- A sample of ^{232}Th irradiated in a fast or thermal spectrum becomes critical. In the IAEA regulation [1], this situation is taken into account by considering ^{233}U - which is the main fissile isotope created during ^{232}Th irradiation - as a fissile isotope in paragraph 222;
- ^{243}Am sample is critical in thermal spectrum after irradiation in an SFR and a PWR, mainly because of the formation of ^{245}Cm during the irradiation. For this isotope, it is interesting to note that the Kinf is higher after irradiation in a PWR than in an SFR, contrary to natural uranium and ^{232}Th;
- The Kinf of ^{241}Am sample irradiated in an SFR is higher than 1 over a large moderation ratio range (H/X between 100 and 800);
- ^{242}Cm and ^{237}Np samples irradiated in an SFR are not critical in thermal spectrum. After irradiation in a PWR, the Kinf is also inferior to 1 but the margin is lower than after irradiation in an SFR;
- After irradiation in a PWR, most of the minor actinides samples studied (^{241}Am, ^{242}Cm and ^{237}Np) remain subcritical in thermal spectrum, though they are more reactive than irradiated natural uranium in thermal spectrum for high moderation ratio.

These results point out that a material containing actinides which are not fissile before irradiation may become fissile after being irradiated in fast reactors but also in thermal reactors. This enhances the necessity to consider minor actinides that have been irradiated in any type of nuclear reactor when they are separated from uranium or plutonium, when the fuel is doped with one of these minor actinides, or when the fuel is made only of minor actinides.

Conclusions

Minor actinides are not defined as fissile nuclide by the IAEA regulation [1]. However, the paragraph 222.3 of the guidance [2] advices to perform a criticality safety analysis in presence of minor actinides separated from uranium, plutonium and fission products.

Indeed, minor actinides such as ^{242m}Am, ^{243}Cm, ^{245}Cm, ^{249}Cf and ^{251}Cf have a lower minimal critical mass than the usual fissile isotopes. Therefore, the usual mass limit use to define a material as a fissile material or to classify it as FISSILE is not relevant to these isotopes.

Moreover, irradiation of non-fissile minor actinides shows that a non-negligible quantity of fissile isotopes can be produced. Besides, after irradiation in both thermal and fast spectrum reactors, the infinite multiplication factor of these minor actinides can be higher than 1 in thermal spectrum.

These two points confirm the advice of paragraph 222.3 of the guidance [2].

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