Selection of exception limits for all actinide nuclides based on revised criteria for safe international transport and including storage delay

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Since 1998, there have been some speculations about future transport of significant quantities and concentrations of other actinide nuclides than the four currently listed in the regulation for the safe transport of the radioactive material [1]. Therefore, it raised a need to specify exception limits for such actinides.
In order to define credible exception limits, it was necessary to have reasonably accurate data for all actinide nuclides. Then the DGTREN/participants decided to perform calculations with different codes (MONK, MCNP, CRISTAL and SCALE) and different cross-section libraries (JEF2.2, ENDFB, etc.). The parameters of interest (such as k-infinite, critical masses) were determined.
This article presents the work achieved and the questions raised, e.g. related to the effect of the radioactive decay of the isotopes on the criticality risks. It also points out the need for an evolution of the regulation of the safe transport of radioactive materials and gives a proposition of modification for the IAEA requirements related to, firstly, the list of the fissile materials, secondly, the rule to determine the quantities of actinide nuclides that can be excepted from the requirements for the packages containing fissile materials.

1. Introduction

Currently, the IAEA Regulation for the Safe Transport of Radioactive Material [1] only considers ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu as fissile materials whereas other actinide nuclides are capable of sustaining a chain reaction on their own. In order to propose a modification of the regulation to take into account additional actinide nuclides, which present a potential hazard in terms of criticality, the European Community (DGTREN) decided to partially fund a working group gathering IRSN, EMS, DTLR (now DfT) and SERCO.
The first goal of this Group was to define the list of actinide nuclides able to sustain a chain-reaction and to calculate the minimum critical masses of those actinides. Then, as most of those actinides are supposed to be produced and transported in small quantities [2], some exception limits (mass limits) were determined; for a mass of nuclide per package lower than its exception limit, there will be no need to assess the criticality risk.
The current fissile exception rule, in terms of excepted masses, is based on Woodcock and Paxton calculations performed in the 1960's [4]. Since the maximum number of packages per consignment increased (the Woodcock and Paxton calculations were performed with 5x50 packages) and the case of accidental configurations need to be assessed (the dimension of the package in Woodcock and Paxton calculations is always greater or equal to 10x10x10 cm^3), a new method [5] to define fissile exception criteria, was discussed during IAEA Consulting in March 2004 (Paris).

2. List of fissionable actinides

The evaluations only include nuclides with half-lives greater or equal to 45 days although shorter half-lives may need to be considered in transport. Radioactive decay of some nuclides into more reactive nuclides is an additional consideration that needs to be evaluated. An example is ^{233}Pa, which cannot support criticality directly but decays into ^{233}U. Minimum "cooling time" after production of the nuclide as well as maximum length of transport should be established before a detailed evaluation is carried out.
In this article the term fissile is for nuclides that can sustain a chain reaction with thermal neutrons and fissible is used for nuclides that can only support a chain reaction as metal.
In order to define the criticality hazard of each actinide nuclide, different parameters have been studied:

- the ETA parameter (η) as a function of energy; it gives the capability of a given isotope to increase the number of neutrons in a system for any incident neutron energy ($\eta(E)$ is equal to $\sigma f(E)*\nu(E)/\sigma a(E)$),
- the k-infinite parameter of a given actinide; it gives the possibility for a single nuclide to reach the criticality state in an infinite medium.

Different cross-section evaluations were studied, such as JEF2.2, ENDF/B-VI.8, JENDL3.3.
More precise information on the work achieved is given in article [5].
Finally, the following list was obtained.

Element	Nuclide	Spectrum	Element	Nuclide	Spectrum	Element	Nuclide	Spectrum
Actinium	**^{227}Ac**	NONE	**Plutonium**	**^{236}Pu**	SLOW	**Berkelium**	**^{247}Bk**	SLOW
Thorium	**^{228}Th**	NONE		**^{237}Pu**	SLOW		**^{248}Bk**	?
	^{229}Th	FAST		**^{238}Pu**	FAST		**^{249}Bk**	FAST
	^{230}Th	NONE		**^{239}Pu**	SLOW	**Californium**	**^{248}Cf**	FAST
	^{232}Th	NONE		**^{240}Pu**	FAST		**^{249}Cf**	SLOW
Protactinium	**^{231}Pa**	NONE		**^{241}Pu**	SLOW		**^{250}Cf**	FAST
Uranium	**^{232}U**	SLOW		**^{242}Pu**	FAST		**^{251}Cf**	SLOW
	^{233}U	SLOW		**^{244}Pu**	FAST		**^{252}Cf**	SLOW
	^{234}U	FAST	**Americium**	**^{241}Am**	FAST		**^{254}Cf**	FAST
	^{235}U	SLOW		**^{242m}Am**	SLOW	**Einsteinium**	**^{252}Es**	?
	^{236}U	NONE		**^{243}Am**	FAST		**^{254}Es**	SLOW
	^{238}U	NONE	**Curium**	**^{242}Cm**	FAST	**Fermium**	**^{257}Fm**	?
Neptunium	**^{235}Np**	FAST		**^{243}Cm**	SLOW	**Mendelevium**	**^{258}Md**	?
	^{236}Np	SLOW		**^{244}Cm**	FAST			
	^{237}Np	FAST		**^{245}Cm**	SLOW			
				^{246}Cm	FAST			
				^{247}Cm	SLOW			
				^{248}Cm	FAST			
				^{250}Cm	FAST			

Table 1. List of fissile/fissible actinide nuclides

3. Critical masses obtained

Given adequate cross sections, it is easy to evaluate the individual properties of each actinide isotope. The properties selected here are k_{inf}, the infinite neutron multiplication factor and k_{eff}, the effective neutron multiplication factor for spheres of metals and of mixtures with water. Calculations first focussed on bare systems and systems reflected by water and steel. Calculations include the best estimates of each property using available information.
The values of the critical masses have been calculated with:
- SCALE 4.4 with sequence CSAS1X and the ENDF/B-V 238-groups library (EMS),
- MCNP4C2 with various releases of ENDF/B (-V, -VI.2, -VI.5 and -VI.8) and JENDL (-3.2 and –3.3) as well as JEF 2.2 and ENDL-92 continuous energy cross section libraries (EMS),
- JEF2.2 evaluation, APOLLO2-Sn (CRISTAL V0.1) and 172-groups (IRSN),
- JEF2.2 evaluation and TRIPOLI4.1 Monte Carlo (continuous) code (IRSN).

The values obtained with Monte Carlo codes (MCNP or CRISTAL) were determined with a standard deviation (σ) on the k-effective lower or equal to 0.001 (0.1% in Δk); most of IRSN results are based on calculations presented in details in [3].
It appears that the values of critical masses are very similar when a continuous code or a multi-groups code is used with JEF2.2, except for ^{236}Pu.

The same values are obtained with ENDF/B and JEF2.2 for: ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{238}Pu, ^{239}Pu and ^{240}Pu. The values differ:

- by less than a factor 2 for ^{237}Np, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{247}Cm,
- by a factor 2 to 7 for ^{235}Np, ^{236}Pu,
- by a factor 30 for ^{242}Cm.

Tentative explanations of the discrepancies are given in [5].
The discrepancies point out the importance of the safety factors that will be used for the definition of the exception limits, depending on the confidence in the data.

4. Definition of some exception limits

To prevent criticality hazards during a transport of excepted fissile material, both normal conditions (array of packages) and accidental conditions of transport (eventually fissile material of many packages is mixed together) must be considered. The configurations studied are given in the **Fig.1.**

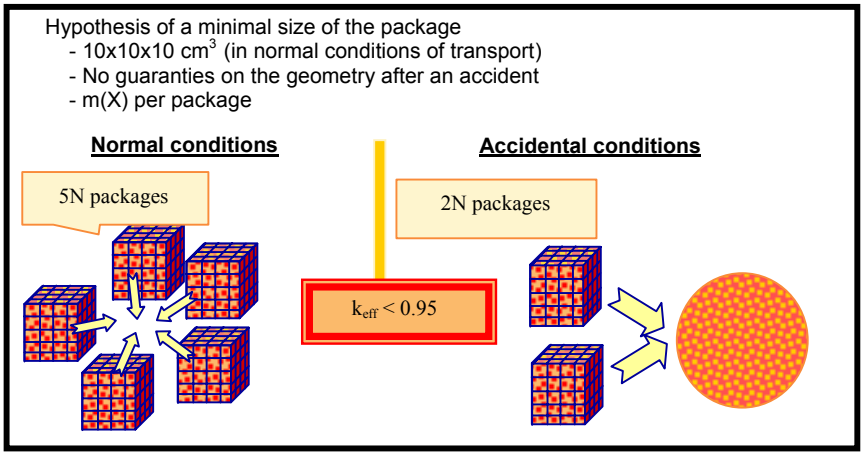

Fig. 1 Configurations studied

4.1 Calculations achieved

Calculations have been performed with: ^{235}U, ^{233}U, ^{239}Pu, ^{241}Pu and ^{242m}Am in thermal energy spectrum and ^{237}Np, ^{244}Cm and ^{238}Pu in fast energy spectrum; those nuclides correspond to the actinide nuclides already present in the regulation (^{235}U, ^{233}U, ^{239}Pu, ^{241}Pu and, also, ^{238}Pu) and the ones where large discrepancies with the different calculations between CRISTAL (IRSN) and SCALE or MCNP (EMS) appeared (^{237}Np, ^{244}Cm and ^{242m}Am).

4.1.2. Normal conditions of transport

The number N_1 of packages that could lead to a criticality hazard in normal conditions was determined. Each cubic package dimension is greater or equal to $10\times10\times10$ cm^3. In order to be consistent with the regulation, it is postulated to have $5N_1$ packages close together.
Then different calculations were performed, depending on the mass m(X) per package, to find the number of $5N_1$ packages leading to $k_{eff} + 3\sigma = 0.95$ (CSI equal to 50).
The aim is to find the relations $N_1 = f(m(X))$ that lead to no criticality hazard.
The calculations are precisely described in [5].

Calculations were also carried out with a steel wrapper. ^{238}Pu has a particular behavior in this case: the reactivity increases with the steel thickness. There is a better reflection in each package when there is a steel wrapper around the package (steel is a good reflector in fast spectrum). For the other fissible isotopes (^{244}Cm and ^{237}Np), the presence of a steel wrapper gives an opposite effect, as the reactivity is lower with a steel wrapper.

Finally:

- the crystal form is the most constraining form for fissible nuclides, while an homogeneous concentration of the fissile isotope in the package leads to the maximum k-effective;

- the steel wrapper increases the reactivity for the case of ^{238}Pu with 1 kg of ^{238}Pu per package, while it reduces the reactivity for every other isotopes (fissible/fissile) studied.

4.1.2. Accidental conditions of transport

The number N_2 of packages that could lead to a criticality hazard in accidental conditions was determined. In order to be consistent with the transport regulation for fissile material, $2N_2$ damaged packages into the most constraining form (sphere) were considered.
Then, the number of packages $2N_2$ leading to k_{eff} = 0.95 (maximum allowable mass) is determined.
Maximum allowable masses were determined with CRISTAL (APOLLO2 - Sn 2D (20 gr)) in order to define the N_2 value. For the cases with steel, the steel wrapper is removed, in order to take the most constraining case (when steel is homogeneously mixed with the fissile medium, the reactivity decreases due to steel absorption). The fact of taking steel into account would allow carrying a higher number of packages.
N_2 is calculated as follows: N_2 = (1/2)*(Allowable Mass(X) / m(X)), with m(X) the mass of actinide nuclide X per package and the Allowable Mass is the minimum mass of nuclide X in a sphere (with the reflector - water, steel or lead- that gives the lowest mass value) that gives a k-effective equal to 0.95.
Fig. 2, gives the numbers N_1 and N_2 depending on the mass m(X) per package (for the 8 actinide nuclides studied). It shows that the accidental conditions are always more limiting than the normal conditions up to a mass of actinide nuclide per package. For large masses of actinide nuclides per packages, the normal cases become more constraining.

4.1.3. Other points

Other parameters that may influence the reactivity, have been studied, such as:
- other moderators than “standard” water moderator or other reflectors than “standard” water (thermal spectrum) or steel (fast spectrum) reflectors,
- non-uniform repartition of fissile material,
- mixture of actinide nuclides.

These effects need to be assessed for the definition of the excepted mass limits.

4.2 Exception limits proposition

As calculations were performed to determine the most limiting case between normal and accidental conditions of transport for 8 nuclides, it was then possible to set a rule and give an exception mass limit for each of them. Then, the rule and the exception limits will be extended to the other actinide nuclides.
The principles to set the rule are the following:
1. the exception rules will be set considering the accidental conditions only and it will be verified, for the mass limit per package obtained, that the accidental conditions are more limiting than the normal ones (fig 2); thus it is possible to derive the mass limits from the maximum allowable mass calculated for each isotope;
2. every possible environment and configurations (reflectors, moderators, mixtures …) will be considered; actually, if the package is excepted, there will be no chance to re-check the real configurations of transport;
3. the minimum of packages leading to a criticality hazard will be greater or equal to 10 (N > 10); this will guarantee a dilution of the medium and reduce the consequences of human errors;
4. the current limit of 15 g per package for ^{235}U in the regulation [1] will be kept.

During the discussions above, it appeared that the exception limit of mass per package should take into account:
- the effect of different types of reflectors, more limiting than the ones required in the current IAEA regulations; actually the excepted packages will not be checked by the safety authorities, in terms of criticality; thus, most of the configurations must be taken into account; the effect of different types of moderators also has to be considered; the critical mass will be reduced with a safety factor F1,
- a safety margin of 5% on k-effective, to take into account all unknown phenomena (mixture, repartition of the fissile medium); the critical mass will be reduced with a safety factor F2,
- the knowledge on the cross-sections has to be taken into account (reduction of the critical mass by a factor F3),

- the number of packages that can lead to criticality in accidental conditions must be greater than 10 (a human error leading to a 2x10 package array should be safe).

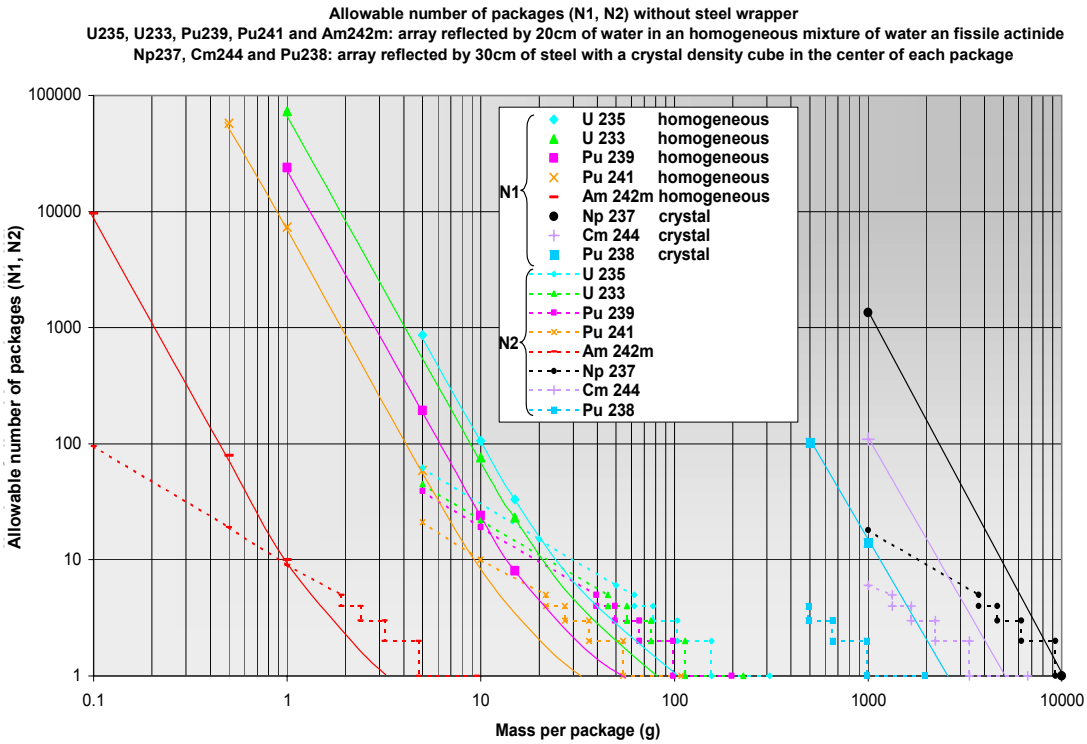

Fig. 2 Allowable Number of Packages depending on the mass per package

The proposed method to determine each consignment limit and each mass limit per package is described in article [5].
An additional rule states “if the total mass of a group of actinide nuclides is less than 10 mg, they don’t need to be taken into account” (10 mg is less than 1/30 of the minimum m*(X) obtained for any of the actinide nuclides X).
The values obtained are given in the following table, giving the mass limit per consignment (Mconsignment) and the mass limit per package (m*), they can be compared to the current limits used in the current para. 672 a) of the regulatory text [1].

Nuclide	Mconsign-ment (g)	m* (g)	Nuclide	Mconsign-ment (g)	m* (g)	Nuclide	Mconsign-ment (g)	m* (g)	Nuclide	Mconsign-ment (g)	m* (g)
^{229}Th	-	-	^{238}Pu	1880	150	^{246}Cm	1760	140	^{251}Cf	1	0.08
^{232}U	700	55	^{239}Pu	110	9	^{247}Cm	50	4	^{252}Cf	230	20
^{233}U	130	10	^{240}Pu	6200	510	^{248}Cm	1700	140	^{254}Cf	180	15
^{234}U	31500	2600	^{241}Pu	60	5	^{250}Cm	1180	100	^{252}Es	-	-
^{235}U	190	15	^{242}Pu	12200	1000	^{247}Bk	2800	230	^{254}Es	2.5	0.2
^{235}Np	490	40	^{244}Pu	4800	400	^{248}Bk	-	-	^{257}Fm	-	-
^{236}Np	4	0.3	^{242}Cm	610	50	^{249}Bk	10400	850	^{258a}Md	-	-
^{237}Np	10100	840	^{243}Cm	16	1.3	^{248}Cf	-	-	^{241}Am	8800	730
^{236}Pu	350	30	^{244}Cm	4200	350	^{249}Cf	3	0.25	^{242}Am	4.5	0.3
^{237}Pu	7	0.5	^{245}Cm	10	0.9	^{250}Cf	250	20	^{243}Am	29400	2400

Table 2. Values of mass limits per consignment and per package.

5. Risks due to radioactive decay

As it was mentioned above for the definition of the fissionable isotopes, the risk to obtain a more reactive mixture after radioactive decays must be addressed.
Therefore, for every nuclide X (on every chain of disintegration), the mass limit (limits in table 2) should be related to the most reactive "daughter" isotope of X.

While considering the α, β+ and β- decays reactions, 6 different chains were identified, including almost every actinide nuclides. Those chains lead to ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{240}Pu and ^{242}Pu.
For the purpose of the study, nuclides with a radioactive period lower than 2 days were not considered (if the package is transported after 20 days, those nuclides will have already decreased into their "daughters").
In the chains, the decay reactions that lead to a more (plain arrow) or less (dotted arrow) reactive isotope can be identified.

For example, ^{232}U production chain is given below:

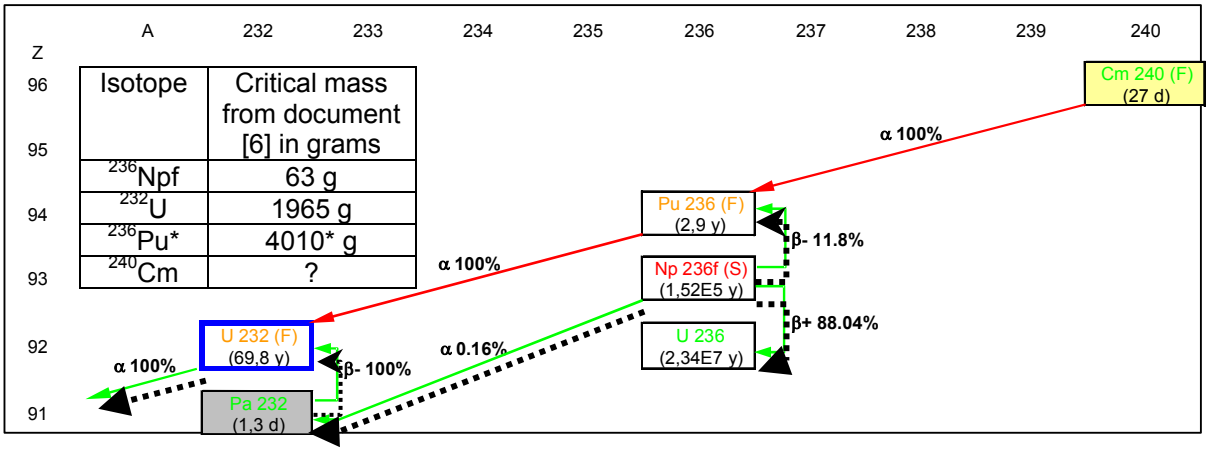

Fig 3. ^{232}U production chain

*This evaluation of ^{236}Pu and ^{232}U was based on earlier cross section data. More recent data indicate that the critical mass for ^{236}Pu is lower than that for ^{232}U, see tables 2 and 3. This will reverse the conclusions made here but the principles for considering radioactive decay still applies.

For example, while considering the chain above:
- ^{236f}Np is the most reactive isotope (it has the smallest critical mass in the chain), its mass limit will be derived from its own critical mass (the mass limits will be obtained with the critical mass and the safety factors described in paragraph 4 above);
- For ^{236}Pu, its mass limit will be derived from ^{232}U critical mass, because this "daughter" nuclide is more reactive than ^{236}Pu. Indeed, the mass of ^{236}Pu transported at a time t = 0 will present a higher risk in terms of criticality after 30 years.

As a first conservative approach, it could be proposed that, for every chain, the smallest mass limit of the daughters should be retained for the mass limits of the fathers.

However, a less constraining approach could be proposed: if the father isotope X decreases into a more reactive isotope Y, the "daughter" isotope Y has its own radioactive disintegration; then it increases until a certain time but then starts to decrease: the maximum quantity of the "daughter" isotope Y depends on the radioactive periods of both "father "and "daughter" isotopes. Then, it is possible to apply a factor f (less than 1) to the quantity of the daughter isotope produced from a previous mass of X (Mass of produced Y will always be lower than f times initial mass of X):
- for father isotopes that have small decay periods regarding to the daughters, this coefficient is equal to 1 as the "father" isotopes decreases by 100% into its daughters in a short time (e.g. ^{242}Cm into ^{238}Pu (T1/2 = 163 days) or ^{235}Np into ^{235}U (T1/2 = 1.08 year)), while no decay of the daughter occurs.

- for the “father” isotopes that have long decay periods, a coefficient could be used to consider the fact that the quantity of daughter formed will be lower than the initial quantity of the father. As the father isotope decreases, it isn’t necessary to consider the whole quantity of father.

For example, in the ^{235}U production chain, a factor for ^{243}Am (f_{Am243}) could be applied. This factor is linked to ^{243}Am decay (by 100%) into ^{239}Pu (^{239}Np is neglected) with a long decay period: the maximum quantity of ^{239}Pu is obtained after 19000 years and corresponds to 60% of the initial value of the quantity of ^{243}Am (see **Fig. 4**):

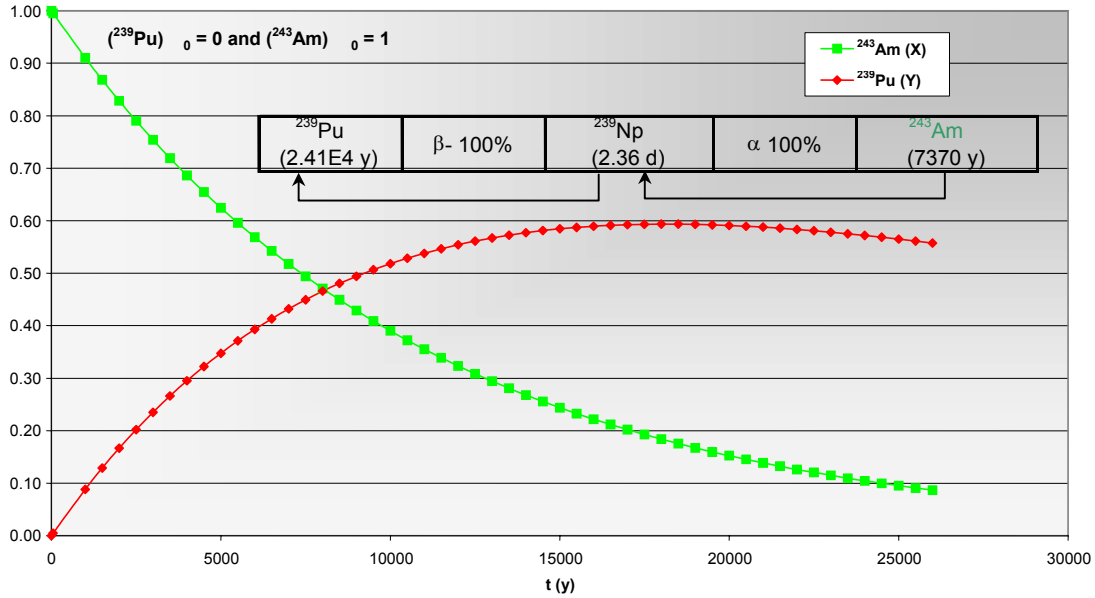

Fig 4. ^{239}Pu formation from ^{243}Am decay (^{235}U formation from the ^{239}Pu has been neglected).

Moreover, it seems possible to retain the quantity of ^{239}Pu obtained from the ^{243}Am after 100 years (after 100 years, the content of the package is assumed to be checked again before transportation). Then, the maximum amount allowed of ^{243}Am can be determined:

- with the penalizing method, the mass limit for ^{243}Am will be derived from ^{239}Pu Critical Mass and would be a very small mass limit: Mass limit of ^{239}Pu equal to 110 g in table 2 (instead of 29,4 kg for ^{243}Am);
- with a factor f_{243Am} considering that after 100 years the amount of ^{243}Am changed in ^{239}Pu is less than 1% of the initial quantity of ^{243}Am, the mass limit for ^{243}Am will be $100*M_{lim\ Pu239}$ = 11 kg; this limit can be applied to ^{243}Am as the mass limit obtained is smaller than the mass limit obtained from ^{243}Am mass limits in table 2 (29,4 kg).

Finally, the conclusions obtained for this example are:

- the use of the critical mass of ^{243}Am would not *a priori* be safe for transportation period from 0 to 100 years: 132 kg of ^{243}Am (critical mass) would lead to 13,2 kg of ^{239}Pu after 100 years, which is greater than the critical mass of ^{239}Pu (the effect of the mixing of ^{239}Pu with ^{243}Am, which is supposed to reduce the reactivity, is not considered here),
- the use of the critical mass of ^{239}Pu to derive the mass limit for ^{243}Am would be very penalizing,
- with the assumption of a time period of interest of 100 years, it is proposed to use a mass limit for ^{243}Am derived from the critical mass of ^{239}Pu multiplied by a factor 100 (divided by 1%).

However, this coefficient cannot always be used: if the father mass limit obtained from the daughter critical mass with the coefficient f is higher than the critical mass of the father itself, the mass limit of the father must be considered (this is the case of ^{237}Np with ^{233}U):
$Mass\ limit_{\ father} = Min\ (Mass\ limit_{\ father\ (tab.2)}\ ;\ f_{daugthers}\ .\ Mass\ limit_{\ daughters})$

Finally, lists of mass limits for the actinides nuclides were set taking into account, with a very conservative approach, the possibility of an increase of reactivity due to the radioactive decay of nuclides. Due to the extreme reduction of the mass limits observed in some cases, a method, using

coefficients taking into account the maximum quantities of isotopes produced during a certain period of interest (we assumed 100 years), was applied when possible.
For future studies, calculations should be performed to test the evolution of reactivity during a certain period, as the daughter and father isotopes will be "melted" until the father nuclide disappears.

The new values obtained are given in next table (the modified values are given in bold italics).

Nuclide	Mconsign-ment (g)	m* (g)	Nuclide	Mconsign-ment (g)	m* (g)	Nuclide	Mconsign-ment (g)	m* (g)	Nuclide	Mconsign-ment (g)	m* (g)
^{229}Th	-	-	**^{238}Pu**	1880	150	**^{246}Cm**	1760	140	**^{251}Cf**	1	0.08
^{232}U	700	55	**^{239}Pu**	110	9	**^{247}Cm**	50	4	**^{252}Cf**	230	20
^{233}U	130	10	**^{240}Pu**	6200	510	**^{248}Cm**	*1700*	*140*	**^{254}Cf**	*180*	*15*
^{234}U	31500	2600	**^{241}Pu**	60	5	**^{250}Cm**	*1180*	*100*	**^{252}Es**	-	-
^{235}U	190	15	**^{242}Pu**	12200	1000	**^{247}Bk**	2800	230	**^{254}Es**	2.5	0.2
^{235}Np	***190***	***15***	**^{244}Pu**	4800	400	**^{248}Bk**	-	-	**^{257}Fm**	-	-
^{236}Np	4	0.3	**^{242}Cm**	610	50	**^{249}Bk**	*10400*	*850*	**^{258a}Md**	-	-
^{237}Np	10100	840	**^{243}Cm**	16	1.3	**^{248}Cf**	-	-	**^{241}Am**	8800	730
^{236}Pu	350	30	**^{244}Cm**	4200	350	**^{249}Cf**	3	0.25	**^{242}Am**	4.5	0.3
^{237}Pu	7	0.5	**^{245}Cm**	10	0.9	**^{250}Cf**	250	20	**^{243}Am**	***11000***	***900***

Table 3. Mass limits considering the risks of decay

6. Conclusion

A list of actinide nuclides that present a potential criticality hazard has been defined.
Minimum critical masses were calculated with various cross-section evaluations and various codes; some discrepancies were observed that pointed out the importance on safety margins for the evaluation of some isotopes.
Then, criticality calculations on arrays of packages containing various masses of isotopes per package were performed, giving the number of allowable packages for every quantity of mass per package.
Moreover, a tentative proposal to consider the potential risk for criticality of radioactive decay was presented.
Finally, a method is defined to set some exception criteria with a mass limit of actinide per package and a mass limit of actinide per consignment. The table 3 gives the values obtained for the mass limits.
The list of fissionable actinides and the mass limits for fissile excepted materials will be proposed for the next cycle of modification of the regulation.

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