



Courrier : BP 17, 92262 FONTENAY-AUX-ROSES CEDEX, France - Téléphone : (33) 01.58.35.74.21 - Télécopie : (33) 01.46.57.29.98

Siège social : 77-83, avenue du Général de Gaulle - 92140 CLAMART - Standard : (33) 01 58 35 88 88 - RCS Nanterre B 440 546 018

Demandeur	DG TREN Mme Van den Berghe
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**TITRE :** **Final report**  
**Evaluation of nuclear criticality safety data and limits for actinides in transport**  
**C4/TMR2001/200-1**

**RESUME :**  
**This final report gives the results of the project including the data of the interim report and the work achieved in the last phases of the project.**  
**First, the report gives a preliminary list of actinide nuclides that should be introduced in the regulations of the transport of radioactive materials, considering their potential effect on reactivity.**  
**Secondly, as many of those actinide nuclides are supposed to be transported in very small quantities, a rule to except them from the fissile requirements was set. This rule is based on calculations for Normal and Accidental Conditions of Transport and takes into account: different parameters that may have an influence on reactivity (like reflectors or moderators) and some safety factors to include the discrepancies between the results obtained by the participants (IRSN, EMS and SERCO).**

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Noms	C. LAVARENNE / D. MENNERDAHL / C. DEAN	V. ROUYER	P. COUSINOU	p.i. M. POURPRIX	J. REPUSSARD
Dates					
Signatures					

\* rapport sous assurance de la qualité

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## 1. INTRODUCTION

Until the 1996 Revision of the IAEA Safety Standards series no.TS-R-1, there were five nuclides that were listed in the Regulations as capable, on their own or in mixtures, of supporting criticality events with neutrons. These nuclides were  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$ . Other nuclides were known to have similar properties but were not considered a threat since they only existed in small quantities or concentrations. Those nuclides belong to the element group *actinides*.

In 1995, the international criticality safety community did not express any worries about those new nuclides. Instead,  $^{238}\text{Pu}$  was removed from the list. A major reason was that  $^{238}\text{Pu}$  cannot sustain a chain reaction in a thermal spectrum; it can only support criticality in a fast neutron energy spectrum. The reason why  $^{238}\text{Pu}$  was not considered a criticality safety threat in 1995 was that it is a very strong heat source. A critical mass during transport was not considered credible.

Since 1998, there have been some speculations about future transport of significant quantities and concentrations of other actinide nuclides than the four already listed. Therefore, it raised a need, first, to define a list of actinide nuclides presenting a criticality hazard, secondly, as most of those actinide nuclides will be shipped in very small quantities, to specify exception limits from the criticality safety requirements of the regulation for such actinide nuclides. But, to set such a list and define credible exception limits, it is necessary to have reasonably accurate data for all actinide nuclides.

To establish exception limits for new nuclides it is also necessary to understand and validate the assumed criteria for the current exception limits.

During the revision process to establish the 1996 version of the IAEA standards, it was decided to add to the limit of 15 g of an excepted isotope per package a general limit on the total mass per consignment [7]:

$$\frac{\text{Mass}(^{235}\text{U})}{X} + \frac{\text{Mass}(\text{other excepted isotopes})}{Y} < 1^1.$$

This additional limit improves the safety of the transport for one consignment but it can be noted that several consignments can be shipped on the same conveyance.

Moreover it appears that the 15 g value has been determined with the Woodcock and Paxton method (described in reference [4]) considering a minimum volume of  $10 \times 10 \times 10 \text{ cm}^3$  for each package. This value per package has to be guaranteed whatever the package is (excepted, industrial, A, B, C) since para.634 [7] applies for all types of packages which contain fissile materials (for excepted packages see para.515(c)). Those dimensions are guaranteed only for non-accidental conditions.

The ICNC article in reference [3] showed that the "15 g" rule is not acceptable if the number of packages is unlimited. The "15 g" limit is linked to the maximal number of packages that can be shipped together. When the Woodcock and Paxton rule was set, the maximum number of packages shipped at the same time was equal to 50; a safety factor of 5 was considered and, then, the rules were set for 250 packages. Influence of the other materials than water (e.g. carbon) and damaged packages were not considered. This rule has therefore to be updated.

The DGTREN/project copes with the problems raised above. The report is divided as follows:

- the physical properties of the actinide nuclides of interest are given in paragraph 2; it concerns densities, half lives, k infinite, cross-section parameters; the values of the critical masses and other criticality properties that have been calculated are given in § 2.5. Those studies give an evaluation of the available data and the confidence one can have in the calculated critical mass,

<sup>1</sup> the values of X and Y are roughly equal to 50% of the critical mass

- Paragraph 3 gives the calculations achieved with 8 actinide nuclides for normal and accidental conditions of transport, with various quantities of actinide nuclides per package,
- Paragraph 4 presents the principles of an exception rule and its application to the actinide nuclides studied, using the results of the calculations achieved for normal and accident conditions of transport and taking into account the confidence in the criticality calculations for the actinide nuclides.

This document is a collaboration of IRSN – EMS (see reference [1]) and DTLR and has been written with the help and the information provided by SERCO.

The authors of the document would like to acknowledge valuable and timely support from a few individuals around the world.

First of all, Frédéric Jean who performed a great number of calculations in support to this project and whose help was appreciated to answer many questions. Jim Stewart, Nicholas Barton from DTLR (UK), Veronique Rouyer, Gilles Sert and Marie Thérèse Lizot from IRSN (France) who helped to cope with the problems raised and participate to the meetings.

Dr. Choon-Sup Gil and Dr. Do Heon Kim from KAERI in Korea who, for the benefit of this project, provided Dennis Mennerdhal with a sub-set of JENDL-3.3 in MCNP format and also gave him fast access to their generation of an ENDF/B-VI.8 library in MCNP-format. Dr. Srinivasan Ganesan at BARC in Trombay, India gave valuable comments and results of recent work on minor actinides. Calvin Hopper from ORNL, USA updated D. Mennerdhal on a recent discussion on definitions of fissile and fissionable materials in the U.S. Dr. Hiroshi Okuno at JAERI, Japan and Dr. Enrico Sartori and colleagues at the NEA Data Bank helped to arrange for a revised version of JENDL-3.2 to be released from the Data Bank in a short time.

The ANS 8.15 WG for providing some data and conclusions on actinides nuclides.

Jacques Anno from IRSN and member of ANS 8.15 has been helpful in discussing issues and providing materials of interest for this project.

Dennis Mennerdahl also thanks Eric Häggblom, Swedish Nuclear Power Inspectorate for direct support of the actinide project and its continuation.

## **2. THE NUCLIDES PROPERTIES**

The ANS 8.15 standards working group, who study the actinides, limits its selection of actinides according to radioactive half-life of more than 40 days and to the capability to support criticality alone or with non-fissionable nuclides. In addition to the ANS 8.15 actinide nuclides, other nuclides have been included in the scope of this project. The new nuclides, on their own or in combinations, may have quite different properties than the nuclides that are common today and will require open-minded evaluation<sup>2</sup>.

The main parameters we have been interested in are:

- the densities of the isotopes of interest; the maximum densities for a given isotope will then be used in the calculations (§ 2.1),
- the half lives of the actinide nuclides (§ 2.2),
- the radioprotection properties of the actinide nuclides; it has been checked to determine the probability to have a steel wrapper in the package (§ 2.3),
- the cross sections evaluation; the capability of sustaining a chain reaction for different types of spectrum will be discussed then (§ 2.4),
- finally the critical masses have been calculated (§ 2.5).

### **2.1 THEORETICAL DENSITIES OF SINGLE NUCLIDES, ELEMENTS AND MIXTURES**

The theoretical densities for individual nuclides or for elements are taken from various handbooks and other references. For actinides, the maximum theoretical densities are expected for metals. Mixtures of some metals and water are probably not normal or even realistic during transport but will be assumed possible.

When a theoretical density is given in a reference, it is assumed that it refers to a specific nuclide or to an element with a given isotope distribution. The theory commonly used is based on the assumption that the theoretical atomic number density for all isotopes of an element is a constant. Given the theoretical density for one isotope, the corresponding theoretical densities for other isotopes can be calculated easily using the atomic masses of each isotope.

In mixtures of isotopes and of elements, the assumed theoretical density of the mixture is based on the assumption that the sum over all elements and isotopes of the fractions of theoretical density for each isotope will add to the total of unity. Less conservative approaches should to be verified by experimental data.

The ANS 8-15 standards working group has extensive experience with actinide properties. The densities proposed in current work by this working group in reference [2] should be used. Newer information can be used whenever it is considered reliable.

The different values are given in the **Table 1**, next paragraph.

### **2.2 ACTINIDE NUCLIDES WAYS OF FORMATION AND RADIOACTIVE HALF-LIVES**

All actinide nuclides are unstable. The natural ones that still exist on earth (<sup>238</sup>U, <sup>235</sup>U, ...) have very long decay periods. Most of the other are artificial nuclides created by men by irradiation of others actinide nuclides in reactors or in laboratories for different reasons (energy production, properties nuclides studies, nuclear physics, ...). Some of them are created in large amounts in reactors and others in small quantities (also in reactors and in laboratories). As they are given by neutron capture

<sup>2</sup> Two fissionable nuclides, which individually cannot support criticality, may cause criticality together in some configurations. This is not expected, but a large variety of new actinides with different properties require open-minded evaluation.

or others nuclear reactions like (n, 2n) reaction, they disappear by radioactive decay ( $\alpha$ ,  $\beta$ - or  $\beta^+$  reaction). Some of them might have a high number of neutrons and protons (Californium (Z = 98), Einsteinium (Z = 99), ...). During the decay of those actinide nuclides, some might decrease from a low-reactive isotope to a high-reactive one. This phenomenon has to be taken into account for the calculation of exception limits criteria for the transport of fissionable material. Mass exception limits have to include the isotopes that will be formed during a certain period after analyse of the package and could increase the reactivity of the package (**reference [8]**).

**Table 1** lists isotopes of the various actinide nuclides with indications of radioactive half-lives. Nuclides with shorter half-lives than 40 days are excluded.

This selection is in agreement with the criteria selected by the ANS 8.15 Standards working group (N.L. Pruvost, ANS Trans. Winter 2000 and presented in reference **[2]**).

**Table 1. General data for actinide isotopes**

Element	Nuclide <sup>3</sup>	Atomic weight	Density (g/cm <sup>3</sup> )	Atoms/barn -cm	T <sub>1/2</sub> (y)	
Actinium <sup>4</sup>	<sup>227</sup> Ac	227.027750	10.043	0.02664	21.77	
Thorium	<sup>228</sup> Th	228.028731	11.524	0.03043	1.913	
	<sup>229</sup> Th	229.031754	11.575	0.03043	7340	
	<sup>230</sup> Th	230.033126	11.626	0.03043	75400	
	<sup>232</sup> Th	232.038050	11.727	0.03043	1.4 x 10 <sup>10</sup>	
	Protactinium	<sup>231</sup> Pa	231.03588	15.336	0.03997	32500
Uranium	<sup>232</sup> U	232.03715	18.681	0.04848	69.8	
	<sup>233</sup> U	233.039628	18.762	0.04848	1.59 x 10 <sup>5</sup>	
	<sup>234</sup> U	234.040946	18.842	0.04848	2.45 x 10 <sup>5</sup>	
	<sup>235</sup> U	235.043924	18.923	0.04848	7.04 x 10 <sup>8</sup>	
	<sup>236</sup> U	236.045561	19.004	0.04848	2.34 x 10 <sup>7</sup>	
	<sup>238</sup> U	238.050784	19.165	0.04848	4.46 x 10 <sup>9</sup>	
	Neptunium	<sup>235</sup> Np	235.04406	20.303	0.05202	1.058
		<sup>236</sup> Np	236.04657	20.389	0.05202	155000
<sup>237</sup> Np		237.048167	20.476	0.05202	2.14 x 10 <sup>6</sup>	
Plutonium	<sup>236</sup> Pu	236.04605	19.601	0.05001	2.87	
	<sup>237</sup> Pu	237.04840	19.685	0.05001	45.7 d	
	<sup>238</sup> Pu	238.04955	19.768	0.05001	87.74	
	<sup>239</sup> Pu	239.05216	19.851	0.05001	24110	
	<sup>240</sup> Pu	240.05381	19.934	0.05001	6537	
	<sup>241</sup> Pu	241.05684	20.017	0.05001	14.4	
	<sup>242</sup> Pu	242.05874	20.101	0.05001	3.76 x 10 <sup>5</sup>	
	<sup>244</sup> Pu	244.064199	20.267	0.05001	8.2 x 10 <sup>7</sup>	
Americium	<sup>241</sup> Am	241.05682	13.662	0.03413	432.2	
	<sup>242m</sup> Am	242.059543	13.717	0.03413	141	
	<sup>243</sup> Am	243.061375	13.774	0.03413	7370	
Curium	<sup>242</sup> Cm	242.05883	13.407	0.03335	162.8 d	
	<sup>243</sup> Cm	243.06138	13.463	0.03335	28.5	
	<sup>244</sup> Cm	244.06275	13.518	0.03335	18.11	
	<sup>245</sup> Cm	245.06548	13.574	0.03335	8500	
	<sup>246</sup> Cm	246.06722	13.629	0.03335	4780	
	<sup>247</sup> Cm	247.070347	13.685	0.03335	1.56 x 10 <sup>7</sup>	
	<sup>248</sup> Cm	248.07234	13.740	0.03335	3.4 x 10 <sup>5</sup>	
	<sup>250</sup> Cm	250.07835	13.851	0.03335	9700	

<sup>3</sup> There are also elements with higher element numbers than 103 that have isotopes with long-half-lives.

<sup>4</sup> This nuclide is included for demonstration purpose only.

Element	Nuclide <sup>5</sup>	Atomic weight	Density (g/cm <sup>3</sup> )	Atoms/barn -cm	T <sub>1/2</sub> (y)
Berkelium	<sup>247</sup> Bk	247.070300	14.671	0.03576	1400
	<sup>248</sup> Bk	248.073080*	14.731	0.03576	9
	<sup>249</sup> Bk	249.07498	14.790	0.03576	320 d
Californium	<sup>248</sup> Cf	248.07218	15.050	0.03653	333.5 d
	<sup>249</sup> Cf	249.07485	15.110	0.03653	351
	<sup>250</sup> Cf	250.07640	15.171	0.03653	13.1
	<sup>251</sup> Cf	251.079580	15.232	0.03653	900
	<sup>252</sup> Cf	252.08162	15.292	0.03653	2.64
	<sup>254</sup> Cf	254.08732	15.412	0.03653	60.5 d
Einsteinium	<sup>252</sup> Es	252.082944	8.808	0.02104	1.29
	<sup>254</sup> Es	254.08802	8.878	0.02104	276 d
Fermium	<sup>257</sup> Fm	257.095099			100.5 d
	<sup>262</sup> Fm				250
Mendelevium	<sup>258</sup> Md	258.09857			51.5 d
	<sup>261</sup> Md				215d
	<sup>265</sup> Md				76d
Nobelium	<sup>258</sup> No	258.09857			1.2
	<sup>260</sup> No				49d

\* Value coming from 1995 Mass evaluation and Tuli January 2000 Wallet Card

The values used in the calculations during the project are the ones in the 5<sup>th</sup> column (Atoms/barn-cm).

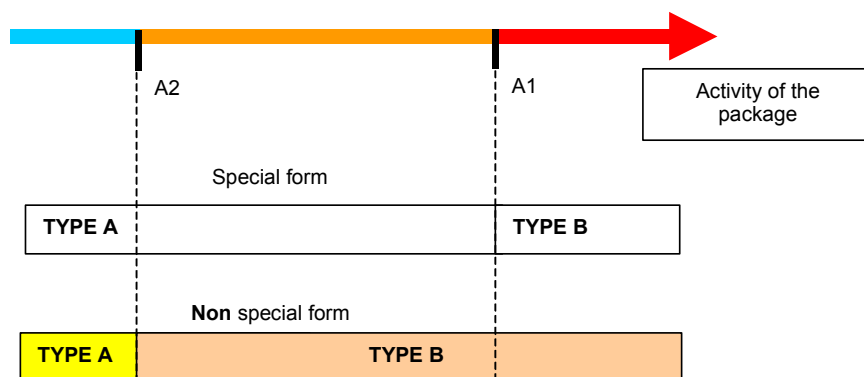
### 2.3 RADIOPROTECTION SAFETY RESTRICTIONS - TYPES OF PACKAGES

The radioactive properties of the actinide nuclides selected in **Table 2** (below – values derived from reference [6]) gives the type of the package (excepted packages, type A or B packages) depending on the quantity of actinide nuclide per package.

Actually the activity of the package, compared to the limits of activities A<sub>1</sub> and A<sub>2</sub> (reference [7] para 201.) and the specific activity (reference [9]), gives, for a special form or non-special form material, the type of the package.

The type of the package has some consequences for our study, as **Type B** packages are very likely to have a steel wrapper around the fissionable material. Further calculations tend to find out which cases (with and without steel) are the most reactive (§ 3).

For Special and non special form solid radioactive material:



<sup>5</sup> There are also elements with higher element numbers than 103 that have isotopes with long-half-lives.

	A <sub>1</sub> limit	A <sub>2</sub> limit	Specific activity	Mass limitation for special form radioactive material (g)	Mass limitation for non-special form radioactive material (g)
	Bq	Bq	Bq/g	Type A package limit	Type A package limit
U 235	unlimited	unlimited	8.014E+04	infinite	infinite
U 233	4.00E+13	6.00E+09	3.589E+08	111452	16.72
Pu 239	1.00E+13	1.00E+09	2.305E+09	4338	0.434
Pu 241	4.00E+13	6.00E+10	3.819E+12	10.5	0.0157
Am 242m	1.00E+13	1.00E+09	3.603E+11	27.8	0.0028
Np 237	2.00E+13	2.00E+09	2.613E+07	765404	76.54
Cm 244	2.00E+13	2.00E+09	3.000E+12	6.7	0.0007
Pu 238	1.00E+13	1.00E+09	6.347E+11	15.8	0.0016

**Table 2. Classification of the different types of packages**

For special form radioactive materials, we can see that transport of type A fissile excepted packages are possible (the mass limits per package due to radioprotection are greater than the mass limits for fissile excepted packages that are in the regulation (for <sup>233</sup>U, <sup>235</sup>U and <sup>239</sup>Pu)): there is no reason to have a type B package when those nuclides are transported as fissile excepted materials (for Pu239, it can be transported in a type B package, if it is a non special form radioactive material). However, the effect of the presence of a steel wrapper needs to be checked for other nuclides (e.g. for <sup>241</sup>Pu).

## 2.4 NUCLEAR PROPERTIES – CROSS SECTIONS

For criticality safety, it is important to understand the nuclear properties of various nuclides that may be involved in the operation being studied. Some of those properties are often specified by neutron cross sections ( $\sigma$ ). A neutron cross section may be seen as the probability for interaction between a nucleus and a neutron. The neutron energy together with the material structure and temperature are important. Properties specified by cross sections involve probability of nuclear absorption (fission, capture, (n, 2n) reaction), scattering (elastic, inelastic), etc. Other nuclear properties are number of neutrons per fission ( $\nu$ ), fission neutron energy spectrum, energy of scattered neutrons and probabilities for radioactive decay through spontaneous fission as well as through alpha, beta and gamma radiation.

### 2.4.1 Different types of nuclides regarding fissions

The nuclides can be divided into 3 groups:

- 1/ Nuclides that can be made to fission with neutrons. All actinide nuclides are **fissionable**. Many other nuclides are also fissionable. The fission cross-sections for <sup>208</sup>Pb and other non-actinide nuclides are discussed in reference [5] (appendix A ref.5.1). In transport and most operations where criticality safety is a potential concern, the fissionability selection should be restricted to neutron energies lower than any significant source in the system studied. The maximum significant neutron energy from fission of any fissionable nuclide could be used as a limit. This limit is probably lower than 20 MeV. In criticality safety, the <sup>208</sup>Pb fission cross section below 20 MeV is not significant. <sup>208</sup>Pb can be excepted as a fissionable material in criticality safety applications. In other applications, where much higher neutron energies are expected, <sup>208</sup>Pb could be significant as a fissionable nuclide.



- 2/ Nuclides that support fission with slow neutrons are said **fissile**. Sometimes the fissile definition is related to the capability of the nuclide to support criticality with thermal or slow neutrons. Another fissile definition is related to the product of  $\nu * \sigma_f^\dagger$ , where  $\sigma_f^\dagger$  is the thermal neutron energy fission cross-section. The preferred definition here is when direct reference to criticality is made. Most actinide nuclides with odd numbers of neutrons in their nuclei are fissile.  $^{229}\text{Th}$  appears to be an exception. Only a few actinide nuclides with even numbers of neutrons appear to be fissile, examples are  $^{232}\text{U}$ ,  $^{236}\text{Pu}$  and  $^{252}\text{Cf}$ . These are fissile but not with the same thermal energy "focus" as the nuclides with odd numbers of neutrons.
- 3/ There is no established term for nuclides that support fission or criticality with intermediate and fast neutrons. The term **fissible** will be used in this report. Some specialists want to exclude by definition the fissile nuclides from those that are fissible. In this note a fissile nuclide may also be and usually is fissible.

The difference may not be significant between using potential for criticality as a definition of fissile or fissible and using the ratios of fission cross sections multiplied by fission neutrons divided by absorption cross sections ( $\sigma_f * \nu / \sigma_a$ ). In the latter case, the cross-sections must be weighted in the correct neutron spectrum.

The absorption probability of a nuclide is obviously important to criticality. Scattering leads to reduced neutron leakage (reflection) and to slower neutrons (moderation) and is also very important.

Fissionable nuclides that are not fissile or fissible can still be important for criticality safety since they may be more efficient than typical reflector materials when surrounding a fissile or fissible material or system. Also between fissile or fissible materials in a system, these fissionable systems may increase the probability of criticality.

Radioactive decay may change the properties of a material or a mixture of materials in a way that increases the potential for criticality. This includes nuclides with shorter half-lives than 45 days. This issue was identified during the project but has not been discussed sufficiently.

#### **2.4.2 Nuclear cross sections – capability to sustain a chain reaction**

In safety applications, it is necessary to rely on well validated and quality assured cross section libraries. In this project, it seems better to take advantage of the latest information to establish best estimate data. Correction, safety or uncertainty factors can then be adjusted to fit each nuclide and each application type. The IAEA transport regulations are seen as such an application type. The time between determination of data and application can lead to modified factors. The reason is more knowledge and experience. The currently available applied cross section libraries are normally based on earlier releases of the major basic cross section data libraries. Examples of basic cross section libraries are ENDF/B, JEF and JENDL. Currently popular and widely available applied libraries are based on ENDF/B-IV, ENDF/B-V, ENDF/B-VI, JEF 2.2 and JENDL-3.2.

Information on evaluations of actinide nuclide cross sections with preliminary applied cross section libraries based on ENDF/B-VI.8, JENDL-3.3 and JEF-3 are of interest to this project. Comparisons between libraries and with experiments as well as likely causes of differences are also of interest.

##### *2.4.2.1 CROSS SECTION EVALUATIONS*

Preparation to evaluate minimum critical mass of individual actinide nuclides has highlighted some significant changes in published masses resulting from the use of the latest evaluated data. Initial proposals for this project suggested using existing product libraries available with APOLLO, SCALE, MONK, MCNP and TRIPOLI. This would yield a spread in the mass due to different methods and current processed nuclear data from JENDL3.2, JEF2.2 and ENDF/B-Vir4. The statuses of the relevant data in the three latest evaluations relative to those used in the product libraries are listed below.

The JEFF Project

In preparation for the release of JEFF3.0, review of JEF2.2 benchmarking program was firstly considered. However an overall review of the status of the data in public domain libraries was also undertaken by John Rowlands and published internally to the JEFF project as JEF/DOC-657. His document attempts to select the best evaluation for inclusion in JEFF3.0. Hence JEFF3.0 should contain the best data prior to the release of revision 8 of ENDF/B-VI and JENDL3.3.

Modern product libraries based on JEF2.2 have been validated for use in criticality studies by UK industry using the MONK code. Similar validation has been seen for SCALE and APOLLO within the JEFF project and at conferences. This validation covers the JEF\_PROJECT "major actinides"<sup>6</sup> (<sup>233</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>237</sup>Np, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>242</sup>Pu, <sup>241</sup>Am, <sup>243</sup>Am plus <sup>232</sup>Th and <sup>238</sup>U). This validation does not cover the minor actinides.

Unfortunately for this project scattering and resonance shielding are important for the JEF\_PROJECT "minor actinides". However the quality of the data and hence uncertainty in results must be influenced by the category division. Further it is probable that existing product libraries contain adequate data for the "major actinides". Indeed these are currently used for criticality assessment.

ENDF/B-VI release 8

Amongst the actinide nuclides of interest to this project, ENDF/B-VI r8 contains new evaluations for <sup>232</sup>U and <sup>236</sup>Np. These are based on JENDL evaluations with important parts of ENDF/BV retained.

During the project (November 2002) a complete ENDF/B-VI release 8 library for MCNP was obtained from KAERI, Korea for evaluation.

JENDL3.3

This library contains many new evaluations of interest. Without further review it is difficult to judge the impact on this project. However JENDL publish values of thermal and fission spectrum average cross-sections, together with resonance integrals. Once the minimum critical mass has been found to apply to a thermal solution or to a fast metallic system, it should be possible to review the pertinent data and consider the impact.

During the project (November 2002) a sub-set of the JENDL 3.3 library for MCNP was obtained from KAERI, Korea for evaluation. The sub-set included the fissionable nuclides that were significantly changed compared with the previous version, JENDL 3.2 as well as new nuclides. KAERI prepared this sub-set for the benefit of this project. JAERI has published several reports and journal articles discussing JENDL 3.3 and comparing it with other cross section libraries.

New Evaluations for Actinide nuclides

It is important to note that all three of the most modern libraries are yet to be validated for criticality. Certainly the JEFF project is asking project members and keen industrial users to test this first version noting its 18 Month expected lifetime before further development. Hence it would not yet be appropriate to use the data for "major actinides" in this project. It would of course be interesting to rerun criticality studies using the new data as part of library validation

The same is not true for the "minor actinides" where considerable development seems to have taken place particularly in JENDL.

The table in **appendix C** notes the source of the evaluation included in the three latest libraries (plus the library JEF/DOC-657). It also gives the conclusions from the JEFF review.

<sup>6</sup> For those actinides, it has been considered necessary to have good scattering cross-sections (in addition to the reaction cross-sections (capture, fission, (n,2n), (n,3n)), which are sufficient to determine the production and transmutation of the other actinides). Note that those "major actinides" don't correspond to the major actinides in criticality, that is U and Pu.

### 2.4.2.2 CAPABILITY TO SUSTAIN A CHAIN REACTION

During the second meeting of the working group it appeared necessary to define 2 types of parameters to determine if an isotope should be listed in the IAEA rules:

- the  $\eta^7$  parameter as a function of energy; it gives the capability of a given isotope to increase the number of neutron in a system for any incident neutron energy (production on absorption),
- the k infinite of the system made of one single isotope.

#### Parameter ETA

The **Table 3**, which gives the capability of sustaining a chain reaction regarding the parameter  $\eta$ , is given with some comments in **appendix A**. When  $\eta$  is above 1 for a certain energy range, then there is a possibility of criticality. This table also points out the impact of inelastic scattering (e.g. for  $^{227}\text{Ac}$ ,  $^{231}\text{Pa}$ , ...) to determine if an isotope can sustain a chain reaction ( $\eta$  can be greater than 1.0 above the fission threshold but, if inelastic scattering is more likely than fission, it will scatter the neutrons below the fission threshold).

#### Parameter k infinite

In order to determine the capability of sustaining a chain reaction, it is also of interest to calculate  $k_{\text{inf}}$ , the infinite neutron multiplication factor, for each actinide nuclide. Pure metals are calculated for all actinide nuclides (**[5]** appendix A). For actinide nuclides that are potentially fissile with slow neutrons, mixtures with water are also calculated.

The evaluated data have to be processed before use in the computer codes. The processing includes condensation over energy into various numbers of energy groups for use with codes such as SCALE and APOLLO. Even continuous energy Monte-Carlo codes (MCNP, MONK, TRIPOLI) reform some of the energy distributions of neutrons resulting from scattering and fission. They are binned to help selection of energies using random numbers.

The processed cross section libraries are expected to determine the results. The computer codes are not expected to make any significant difference for these simple systems. SCALE 4.4, MCNP4C2 and MONK were used to calculate  $k_{\text{inf}}$  for all available nuclides. Actually, some calculations were performed with the same libraries but with different codes (see **appendix B**). It shows some differences in the k infinite when using the same library but a different code. This might be due to the fact that the fission spectrum varies with the incident neutron energy and between isotopes. Most broad group codes use a fission spectrum for each isotope but do not include variation with incident neutron energies. This led here to a 10% difference in k infinite between MCNP and MONK. When MONK\_BINGO was used (it models the incident energy dependence as does MCNP), then the 10% difference disappeared. Consequently, it is important to note that this phenomenon can have large effects, for metals, and should therefore be modeled.

SCALE 4.4 was used with three different cross section libraries. The 238-group and the 44-group libraries have been processed from ENDF/B-V (using different neutron energy spectra) basic cross sections while the 27-group library has been processed from ENDF/B-IV basic cross sections. For some nuclides, more recent data have been included.

MCNP4C2 was used with nine different cross section libraries. Four of them are included in the LANL data set for MCNP4C2; the basic ENDF/B-VI.0 (.60c) library, the delayed neutron ENDF/B-VI.1 (.61c) library, the tabular unresolved resonance ENDF/B-VI.1 (.49c) library. In addition to these, an ENDF/B-VI.5 library (KAERI, Korea), a JEF 2.2 library (ENEA, Italy), a JENDL -3.2 library (JAERI, Japan), an ENDF/B-VI.8 library (KAERI, Korea) and a JENDL 3.3 library sub-set (KAERI, Korea) were used in the project.

<sup>7</sup> The parameter  $\eta(E)$  is equal to  $\sigma_f(E) \cdot v(E) / \sigma_a(E)$ .

The results show that the 238-group ENDF/B-V library in SCALE 4.4 is agreeing quite well with results with ENDF/B-VI cross-sections for MCNP4C2. Large deviations are found for  $^{236}\text{U}$ ,  $^{237}\text{Np}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$  (large differences also between different ENDF/B-VI libraries),  $^{249}\text{Bk}$  and  $^{249}\text{Cf}$ .

At the beginning of the project, the Korean ENDF/B-VI.5 library was the most recent of the available ENDF/B-VI libraries. It differs from other ENDF/B-VI libraries primarily for  $^{236}\text{Pu}$  and  $^{243}\text{Am}$ . The processed evaluation of each actinide nuclide goes back to specific releases of the basic evaluated cross sections (ENDF/B, JEF, JENDL, etc.). For example, in this Korean MCNP format library,  $^{227}\text{Ac}$  was taken from a very different source, JENDL-3.2. There was also a problem with the  $^{249}\text{Cf}$  cross sections. The three different LANL ENDF/B-VI libraries appear to be very similar. The delayed neutron library causes a slight softening of the fission spectrum. This appears to give a small but significant effect when this can be expected, for non-fissile nuclides.

The JEF 2.2 library differs from other libraries for all the Pu and for the lower Cm isotopes.

#### Other parameters - NUBAR<sup>8</sup>

The tables of critical values observed in the literature (see appendix A of [5]) suggest large changes for  $^{232}\text{U}$  and  $^{236}\text{Np}$ . These are correlated to a change in thermal NUBAR when JENDL3.2 is applied. Review of the files in the three latest evaluations indicates ENDF have retained the previous ENDF/B-V NUBAR also adopted by JEFF, whereas JENDL retained their value from JENDL3.2:

- for  $^{232}\text{U}$  the ENDF/B-Vir8 file retains a measured value of NUBAR of 3.10 from A.H. Jaffey and Al. compared by Howerton with a value from systematics of 2.37. JENDL has adopted systematics from Bois and Frehaut;
- for  $^{236}\text{Np}$  the ENDF/B-Vir8 file retains a measured value NUBAR of 3.12 from Mughabghab (84) which is again based on work by Jaffey R. JENDL has adopted data from M. Lidner and D.W. Seegmille and systematics from R.J. Howerton.

As a result of this large variation it was felt relevant to list thermal NUBAR values for all actinide nuclides of interest in **appendix E**. Some large discrepancies can be observed for  $^{232}\text{U}$  and  $^{236}\text{Np}$ ; some smaller differences are also obtained for  $^{241}\text{Am}$ ,  $^{243}\text{Am}$ ,  $^{242}\text{Cm}$ ,  $^{244}\text{Cm}$ ,  $^{247}\text{Cm}$ ,  $^{249}\text{Cf}$  and  $^{252}\text{Cf}$ .

#### Conclusion

The following table summarizes the results obtained and lists the isotopes that can sustain a chain reaction with Fast or Slow incident neutrons.

<sup>8</sup> NUBAR is the mean production of neutrons per fission.

**Table 3. Actinide nuclides capable to sustain a chain reaction**

Actinide nuclide	Slow (S) / Fast (F)	$K_{\infty} > 1$
<sup>228</sup> Th	F?	
<sup>229</sup> Th	F but see comment	Y ?
<sup>231</sup> Pa	F ?	No ?
<sup>232</sup> U	S + F*	Y
<sup>233</sup> U	S + F	Y
<sup>234</sup> U	F	Y
<sup>235</sup> U	S + F	Y
<sup>236</sup> U	F ?	No
<sup>235</sup> Np	F	Y
<sup>236</sup> Np	S + F	Y
<sup>237</sup> Np	F	Y
<sup>236</sup> Pu	S + F *	Y
<sup>237</sup> Pu	S + F	Y
<sup>238</sup> Pu	S** ? + F	Y
<sup>239</sup> Pu	S + F	Y
<sup>240</sup> Pu	F	Y
<sup>241</sup> Pu	S + F	Y
<sup>242</sup> Pu	F	Y
<sup>244</sup> Pu	F	Y
<sup>241</sup> Am	F	Y
<sup>242m</sup> Am	S + F	Y
<sup>243</sup> Am	F	Y
<sup>242</sup> Cm	S ?*** + F	Y
<sup>243</sup> Cm	S + F	Y
<sup>244</sup> Cm	F	Y
<sup>245</sup> Cm	S + F	Y
<sup>246</sup> Cm	S ?*** + F	Y
<sup>247</sup> Cm	S + F	Y
<sup>248</sup> Cm	F	Y
<sup>250</sup> Cm	F	
<sup>247</sup> Bk	S + F	
<sup>248</sup> Bk	No evaluated data available	
<sup>249</sup> Bk	F	Y
<sup>248</sup> Cf	No evaluated data available	
<sup>249</sup> Cf	S + F	Y
<sup>250</sup> Cf	F	Y
<sup>251</sup> Cf	S + F	Y
<sup>252</sup> Cf	S + F	Y
<sup>254</sup> Cf	S? + F	
<sup>252</sup> Es	No evaluated data available	
<sup>254</sup> Es	S + F	
<sup>257</sup> Fm	S + F	
<sup>258</sup> Md	No evaluated data available	

\* For those actinide nuclides, the critical mass of metal is smaller than the critical mass obtained for solution (see **appendix D** for comments), except for Pu236 for which last evaluations seems to give a higher critical mass for metal.

\*\* For <sup>238</sup>Pu,  $\eta$  is greater than one only for Fast neutrons in ENDFB/VI. In JEF2.2 and JENDL3.3 the  $\eta$  can be greater than one for Slow and Fast neutrons (see **appendix D**)

\*\*\* See comments in **appendix D**

For the JEFF\_PROJECT "major actinides", it is appropriate to use current product libraries for critical mass studies. For others actinide nuclides the three latest libraries would probably offer better data; however the data are not likely to be as accurate as those for "major actinides".

Care needs to be taken for those nuclides whose minimum critical mass comes from a system with a hard spectrum. Lack of high quality inelastic scattering data could have a considerable impact. (Note that a critical mass cannot be obtained for  $^{238}\text{U}$  due to inelastic scattering).

To derive the critical masses, we considered all available information, including that from the latest versions of the evaluated data libraries JENDL-3.3, ENDF/B-VI.8. The intention is to select limits so that a minor change in the cross section data will not require or motivate a change of the regulations. Very conservative limits may cause unnecessary problems while best estimates may lead to unacceptable safety margins. Some balance is required based on practical needs in the near future and quality of data.

More experimental data is required to confirm some selections.

## 2.5 CRITICALITY PROPERTIES

Given adequate cross sections, it is easy to evaluate the individual properties of each actinide isotope. The properties selected here are  $k_{\text{inf}}$ , the infinite neutron multiplication factor and  $k_{\text{eff}}$ , the effective neutron multiplication factor for spheres of metals and of mixtures with water. Concerning the reflectors, we decided to first focus on the bare systems and the systems reflected by water and steel. Some additional calculations have been performed by EMS with various reflectors (lead, steel, water,  $^{231}\text{Pa}$ , ...) and are presented in reference [1].

**Table 4** includes the best estimates of each property using available information. The neutron energy spectrum is included and it is supposed to be either S(low) or F(ast). Reference [5] also contains others results that are of interest, including those for other reflector materials, moderators or geometries (e.g. spherical but different radius than exactly critical).

The values of the critical masses in white have been calculated with SCALE 4.4 with sequence CSAS1X and the ENDF/B-V 238-groups library.

The values in yellow have been calculated with MCNP4C2: ENDL-92 library was used for  $^{236}\text{Np}$  and  $^{235}\text{Np}$  and LANL ENDF/B-VI library for  $^{237}\text{Np}$ ,  $^{237}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$ ,  $^{249}\text{Bk}$  [1].

EMS has obtained several cross section libraries for MCNP from different sources, in addition to libraries reported before: JENDL-3.2 from JAERI (Japan), JENDL-3.3 from KAERI (Korea) and ENDF/B-VI.8, also from KAERI have been obtained. The JENDL-3.3 library (a sub-set including actinide nuclides that were significantly changed from JENDL-3.2) was generated quickly by KAERI on EMS request for the purpose of this project. A preliminary version of the ENDF/B-VI.8 library was also made available by KAERI to EMS for this purpose. Many values of critical masses using these libraries were also obtained, in particular for those nuclides where cross sections were not available before ( $^{229}\text{Th}$ ,  $^{250}\text{Cm}$ ,  $^{247}\text{Bk}$ ,  $^{254}\text{Cf}$  and  $^{254}\text{Es}$ ).

A table with updated  $k$  infinite values for available actinide nuclides was generated. It included values from the new JENDL-3.2, JENDL-3.3 and ENDF/B-VI.8 libraries. The  $k$  infinite values are reported in appendix B.

The values in green have been calculated with JEF2.2 evaluation, APOLLO2 - Sn (CRISTAL V0.1) and 172-groups, except for Cm246 and Pu236 (thermal spectrum) where 20 energy groups were used. The values in orange have been calculated with JEF2.2 evaluation and TRIPOLI4.1 Monte Carlo (continuous) code. Results are mainly based on reference [10].

The values obtained with Monte Carlo codes (MCNP or CRISTAL) were determined with a standard deviation ( $\sigma$ ) on the  $k$  effective lower or equal to 0.001 (0.1% in  $\Delta k$ ).

**The data given in this table still need to be checked, compared and evaluated. These are preliminary results that have not been fully discussed.**

**Table 4. Values of k infinite and critical masses calculated for the project**

□ EMS  
ENDF/B-V 238gr

■ EMS  
ENDL92 or  
ENDF/B-VI

■ EMS  
New evaluations

■ IRSN  
APOLLO2 172gr

■ IRSN  
\* TRIPOLI 4.1 JEF2.2

Element	Nuclide	Density (g/cm <sup>3</sup> )	Spectrum (S, M, F)	Library	k <sub>inf</sub>	Critical mass (kg) with different reflectors		
						Bare	H <sub>2</sub> O (20cm)	Steel (30cm)
<b>Thorium</b>	<sup>229</sup> Th	11.575	F	JENDL 3.2	NA	2839	2262	994
<b>Protactinium</b>	<sup>231</sup> Pa	15.336	F	ENDF/B V 238	0.95	-	-	-
<b>Uranium</b>	<sup>232</sup> U	18.681	F	ENDF/B V 238	3.08	3.57	2.13	1.85
		18.678	F	JEF 2.2		3.697	2.178	1.965
		18.678	F	JEF 2.2		3.65	2.163	
		0.467*	S	ENDF/B V 238		9.05	5	3.05
	<sup>233</sup> U	18.762	F	ENDF/B V 238	2.56	15.8	7.32	6.11
		18.758	F	JEF 2.2		16.34	7.464	6.397
		18.758	F	JEF 2.2		17.69	4.563	
		0.056*	S	ENDF/B V 238		1.08	0.568	0.425
		0.059	S	JEF 2.2			0.5594	
	<sup>234</sup> U	18.842	F	ENDF/B V 238	1.52	145	134	83
		18.839	F	JEF 2.2		148.52	137.353	85.33
		18.839	F	JEF 2.2		145.987	135.544	
	<sup>235</sup> U	18.923	F	ENDF/B V 238	2.28	46.7	22	16.8
		18.92	F	JEF 2.2		48.23	22.09	17.159
		18.92	F	JEF 2.2		47.31		
		0.057*	S	ENDF/B V 238		1.42	0.784	0.589
		0.056	S	JEF 2.2			0.7846	
	<sup>236</sup> U	19.004	F	ENDF/B V 238	0.74	-	-	-
		19.165	F	ENDF/B V 238	0.34	-	-	-
<b>Neptunium</b>	<sup>235</sup> Np	20.303	F	ENDL 92	1.75	66.2	60	38.8
		20.303	F	JENDL 3.3	1.75	12	9.48	6.18
	<sup>236</sup> Np	20.389	F	ENDL 92	2.89	6.79	3.21	3.3
		0.012*	S	ENDL 92		0.147	0.072	0.063
	<sup>237</sup> Np	20.476	F	ENDF/B VI.2 DN	1.7	63.6	57.5	38.6
		20.45	F	JEF 2.2		81.935	75.44	49.961
		20.45	F	JEF 2.2		80.62		
<b>Plutonium</b>	<sup>236</sup> Pu	19.601	F	ENDF/B V 238	2.87	8.04	4.99	3.74
			F	ENDF/B VI.8		6.56	3.31	3.15
		19.61	F	JEF 2.2		8.418	5.04	4.01
		19.61	F	JEF 2.2		8.222	5.02	
		0.235*	S	ENDF/B V 238		16.65	10.8	6.93
			S	ENDF/B VI.8				0.97
		0.258	S	JEF 2.2			10.356	6.2
		0.258	S	JEF 2.2			3.7	2.37
	<sup>237</sup> Pu	19.685	F	ENDF/B VI.8	3.05	3.1	1.71	1.62
		0.014*	S	ENDF/B VI.8		0.257	0.136	0.11
	<sup>238</sup> Pu	19.768	F	ENDF/B V 238	2.76	9.49	7.35	4.7
		19.777	F	JEF 2.2		9.157	7.383	4.779
		19.777	F	JEF 2.2		8.946	7.285	
	<sup>239</sup> Pu	19.851	F	ENDF/B V 238	2.99	9.99	5.45	4.49
		19.86	F	JEF 2.2		10.331	6.002	4.787
		19.86	F	JEF 2.2		10.09		
		0.030*	S	ENDF/B V 238		0.885	0.494	0.353
		0.032	S	JEF 2.2			0.503	
	<sup>240</sup> Pu	19.934	F	ENDF/B V 238	2.27	35.7	32.1	19.8
			F	JENDL 3.3				18.3
		19.943	F	JEF 2.2		39.033	34.951	22.579
		19.943	F	JEF 2.2		37.547	33.605	
	<sup>241</sup> Pu	20.017	F	ENDF/B V 238	2.9	12.27	5.87	5.05
		20.027	F	JEF 2.2		13.042	6.683	5.489
		20.027	F	JEF 2.2		12.774		
		0.034*	S	ENDF/B V 238		0.511	0.246	0.205
		0.0264	S	JEF 2.2			0.269	
	<sup>242</sup> Pu	20.101	F	ENDF/B V 238	1.87	85.6	78.2	48.1
			F	JENDL 3.3				36.2

☐ EMS  
ENDF/B-V 238gr

☐ EMS  
ENDL92 or  
ENDF/B-VI

☐ EMS  
New evaluations

☐ IRSN  
APOLLO2 172gr

☐ IRSN  
TRIPOLI 4.1 JEF2.2

Element	Nuclide	Density (g/cm <sup>3</sup> )	Spectrum (S, M, F)	Library	k <sub>inf</sub>	Critical mass (kg) with different reflectors		
						Bare	H <sub>2</sub> O (20cm)	Steel (30cm)
<b>Americium</b>	<sup>241</sup> Am	13.66	F	ENDF/B VI.2 DN	2	57.6	52.5	33.8
		13.67	F	JEF 2.2		75.614	67.768	43.997
		13.67	F	JEF 2.2		72.695	65.776	
	<sup>242m</sup> Am	13.717	F	ENDF/B V 238	3.55	8.83	3.21	3
		13.727	F	JEF 2.2		14.501	6.437	4.619
		13.727	F	JEF 2.2		14.577		
		0.0034*	S	ENDF/B V 238		0.042	0.0203	0.0167
		0.0035	S	JEF 2.2			0.023	
	<sup>243</sup> Am	13.774	F	ENDF/B VI.2 DN	1.69	209	195	138
		13.774	F	ENDF/B VI.8				88.6
		13.784	F	JEF 2.2		209.639	192.839	132.352
		13.784	F	JEF 2.2		203.924	189.342	
<b>Curium</b>	<sup>242</sup> Cm	13.407	F	ENDF/B V 238	1.5	371	260	231
		13.407	F	JENDL 3.3				7.72
		13.399	F	JEF 2.2		25.771	17.603	12.225
		13.399	F	JEF 2.2		24.823	16.994	
	<sup>243</sup> Cm	13.463	F	ENDF/B V 238	3.58	8.35	2.82	3.1
		13.455	F	JEF 2.2		7.515	2.897	2.868
		13.455	F	JEF 2.2		7.415		
		0.036*	S	ENDF/B V 238		0.28	0.127	0.106
		0.0582	S	JEF 2.2			0.2689	
		0.0582	S	JEF 2.2			0.2687	
	<sup>244</sup> Cm	13.518	F	ENDF/B V 238	2.64	26.6	22.1	13.2
		13.51	F	JEF 2.2		33.051	27.072	16.811
		13.51	F	JEF 2.2		32.311		
	<sup>245</sup> Cm	13.574	F	ENDF/B V 238	3.69	9.11	3.08	3.46
		13.566	F	JEF 2.2		6.846	2.639	2.746
		13.566	F	JEF 2.2		6.739		
		0.012*	S	ENDF/B V 238		0.116	0.054	0.042
		0.0115	S	JEF 2.2			0.0473	
	<sup>246</sup> Cm	13.629	F	ENDF/B V 238	2.4	38.9	33.6	22
		13.621	F	JEF 2.2				23.2
		13.685	F	ENDF/B V 238		3.79	6.94	3.52
	<sup>247</sup> Cm	13.677	F	JEF 2.2	3.79	7.117	3.463	2.994
		13.677	F	JEF 2.2		6.982		
		0.246*	S	ENDF/B V 238		4.06	2.18	1.43
			S	JENDL 3.3				0.796
		0.2442	S	JEF 2.2			2.195	
		0.2452	S	JEF 2.2			2.207	
	<sup>248</sup> Cm	13.74	F	ENDF/B V 238	2.56	40.4	34.7	21.5
	<sup>250</sup> Cm	13.851	F	JENDL 3.3	NA	23.5	21.4	14.7
<b>Berkelium</b>	<sup>247</sup> Bk	14.671	F	JENDL 3.3	NA	75.7	41.2	35.2
			S	JENDL 3.3		124	90	52.5
	<sup>248</sup> Bk	14.731	F		NA			
	<sup>249</sup> Bk	14.79	F	ENDF/B VI.5	1.54	192	179	131
<b>Californium</b>	<sup>248</sup> Cf	15.05	F		NA			
		<sup>249</sup> Cf	15.11	F	ENDF/B V 238	4.04	5.91	2.28
		0.015*	S	ENDF/B V 238		0.129	0.06	0.051
	<sup>250</sup> Cf	15.171	F	ENDF/B V 238	3.67	6.55	5.61	3.13
	<sup>251</sup> Cf	15.232	F	ENDF/B V 238	4.32	5.46	2.45	2.27
		0.040*	S	ENDF/B V 238		0.048	0.025	0.02
	<sup>252</sup> Cf	15.292	F	ENDF/B V 238	3.63	5.87	2.91	3.32
			S	ENDF/B V 239		7.93	3.86	2.95
	<sup>254</sup> Cf	15.412	F	JENDL 3.2	NA	4.27	2.86	2.25
<b>Einsteinium</b>	<sup>252</sup> Es	8.808	F		NA	?	?	?
		<sup>254</sup> Es	8.878	F	JENDL 3.2	NA	9.89	2.26
			S	JENDL 3.2		0.077	0.033	0.032
<b>Fermium</b>	<sup>257</sup> Fm		F		NA	?	?	?
<b>Mendelevium</b>	<sup>258a</sup> Md		F		NA	?	?	?

The optimum densities for bare, slow systems are different than for reflected systems, the densities here are given for reflected systems. Reference [1] contains additional results.



It appears that the values of critical masses are the very similar when we use a continuous code or a multigroups code with JEF2.2, except for Pu236.

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

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

The differences of NUBAR can not explain alone those discrepancies.

Some very large differences on the masses have been obtained for  $^{242}\text{Cm}$  (factor 30). This should be explained by the data coming from the ENDFB/VI evaluation, which have to be revised. We can still insist on the importance on the safety factors that will be used for the definition of the exception limit depending on the confidence we have in the data. The calculations achieved with JENDL3.3 give more consistent results.

For  $^{232}\text{U}$ , the effect of the greater value of the ETA parameter with Fast neutrons explains the lower critical masses obtained for metal system than for mixture with water (see **appendix D**).

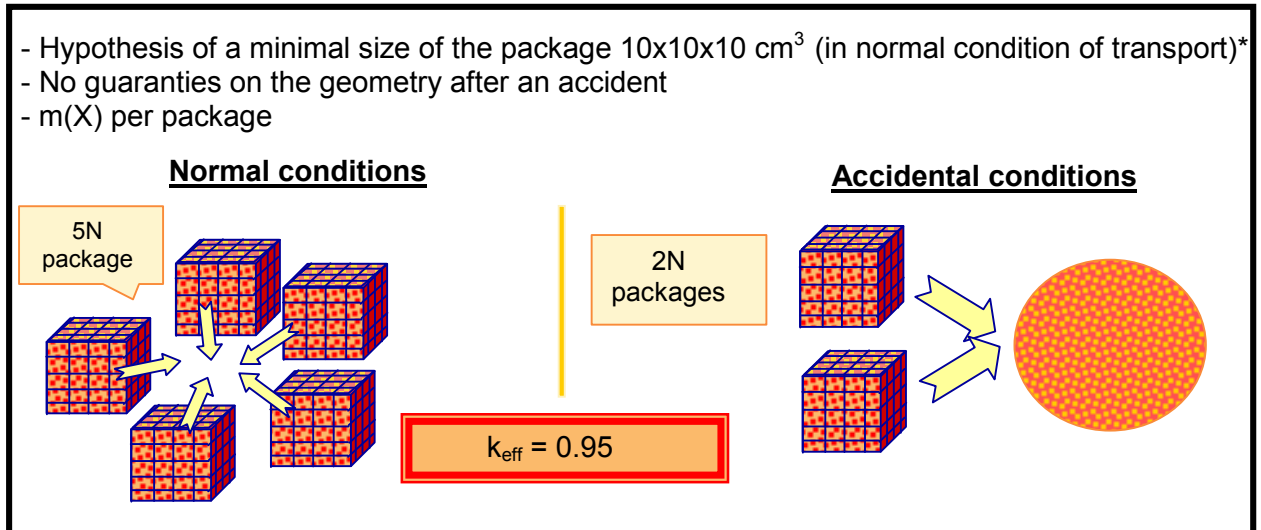
For  $^{236}\text{Pu}$ , this effect is not observed any more with the new evaluations with ENDF/B VI-8 and JEF2.2 with point-wise energy spectrum. The calculations with APOLLO2 (172 groups) gives the same results as ENDF/B VI (238 gr.), where the critical mass in fast spectrum was calculated as lower than the critical mass in thermal spectrum.

**Some work is still on going to try to compare and analyze those results. The last results given by EMS (Blue/Purple) have not been analysed.**

### 3. CALCULATIONS IN NORMAL AND ACCIDENTAL CONDITIONS

To prevent the criticality risk during a transport of excepted fissile material, we have to both study the normal conditions (array of packages containing fissile material) and the accidental conditions of transport (eventually mixing of fissile material of many packages). Therefore we performed calculations for 8 actinide nuclides (given the considerable amount of calculations needed) for the configurations given in the following graph.

#### Configurations calculated



\* The 1996 edition of the IAEA regulations changed the condition of each package in a 5N array from routine to normal conditions of transport. The 10 cm minimum dimension is obvious for the routine condition of transport but not for the normal condition of transport (normal conditions are related to the configuration after the testing). We have chosen to keep the 10 cm minimum dimension for the normal condition and also to assume a cube.

Calculations have been performed with:  $^{235}\text{U}$ ,  $^{233}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{242\text{m}}\text{Am}$  in thermal energy spectrum and  $^{237}\text{Np}$ ,  $^{244}\text{Cm}$  and  $^{238}\text{Pu}$  in fast energy spectrum. We chose the actinide nuclides already present in the regulation ( $^{235}\text{U}$ ,  $^{233}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{238}\text{Pu}$ , even if it was removed) and the ones where large discrepancies with the different calculations between CRISTAL (IRSN) and SCALE or MCNP (EMS) appeared ( $^{237}\text{Np}$ ,  $^{244}\text{Cm}$  and  $^{242\text{m}}\text{Am}$ ) (see **Table 4**).

This study will allow to check the most penalizing cases and help to set a rule and determine exception limits for the transport of excepted fissile material (§ 4).

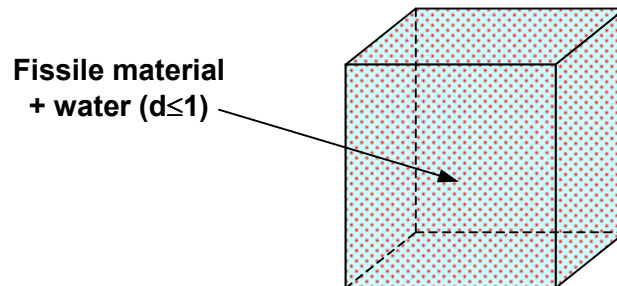
#### 3.1 CRITICALITY RISK IN NORMAL CONDITIONS OF TRANSPORT

We determine the number  $N_1$  of packages that could lead to a criticality risk in normal conditions. Each cubic package dimensions is greater<sup>9</sup> or equal to  $10 \times 10 \times 10 \text{ cm}^3$ . In order to be consistent with the regulation, it is postulated to have  $5N_1$  packages close together.

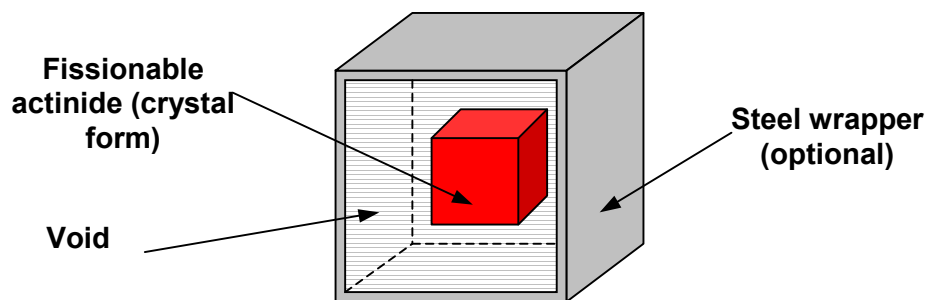
Then different calculations were performed, depending on the mass  $m(X)$  per package, to find the number of  $5N_1$  packages leading to  $k_{\text{eff}} + 3\sigma = 0.95$  (CSI equal to 50). The aim is to find the relation  $N_1 = f(m(X))$  that lead to no criticality risks.

<sup>9</sup> For every case, we checked if an increase of the dimensions of the cube is able to lead to an increase of the reactivity (see also reference [6]).

For that purpose, we defined for each case (thermal or fast spectrum) the most reactive configuration (as a function of the geometry and the moderation of the fissile material). For the thermal spectrum fissile actinide nuclides, it is a homogeneous distribution of the fissile material and water of density less or equal to 1 [6] (numerous calculations were also performed: with a sphere (or a cube) of fissile moderated material surrounded by moisture or surrounded by a water layer):



For the fast spectrum fissile isotopes, in the contrary of thermal spectrum fissile actinide nuclides, the most reactive case was found with a concentrated fissile material (metallic crystalline form) [6]:



The number of packages in array leading to  $k_{\text{eff}} = 0.95$  ( $5N_1$ ) was calculated for both cases (see details in § 3.1.1 and § 3.1.2). The calculations were first performed with 2 types of reflectors depending on the considered actinide nuclides (20 cm of water for moderated systems and 30 cm of steel for fast neutrons systems)<sup>10</sup>. Calculations were performed with and without a steel wrapper to take into account the possibility of having **Type B** packages (see § 2.3).

For the thermal fissile isotopes, the "standard moderator" first used was water. Some others moderators have been checked like  $\text{CH}_2$  and Carbon in § 3.1.1.3, in order to compare with the results obtained with water.

The calculations were performed at IRSN using APOLLO 2 (172gr) - MORET 4 programs and also TRIPOLI 4 program to check some cases with steel (CRISTAL V0 package).

Crosschecking calculations were performed at SERCO with the MONK program to check the APOLLO 2 - MORET 4 results. These crosschecking calculations used the development version of the ANSWERS Criticality code MONK 9 allowing application of the new BINGO collision processor. All calculations have been performed using a JEF2.2 based nuclear data library with a hyper-fine group structure.

Crosschecking calculations were also performed at EMS for some cases.

The calculations performed are detailed in the following paragraphs: the fissile actinide nuclides in slow energy spectrum in § 3.1.1 and the actinide nuclides only able to sustain a chain reaction in fast energy spectrum in § 3.1.2.

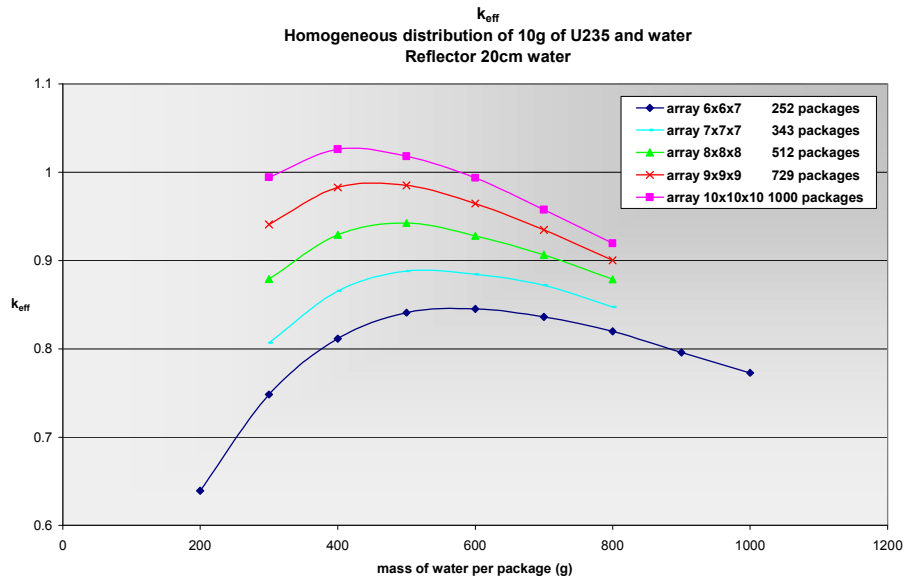
<sup>10</sup> Steel or other reflectors should also be considered for moderated systems

**3.1.1 Calculations with thermal spectrum fissile actinide nuclides**

We first studied the homogeneous distribution of fissile material and water (most reactive configuration), firstly without steel (§ 3.1.1.1), secondly with a steel wrapper (§ 3.1.1.2). The results obtained with the other moderators are presented in the last part (§ 3.1.1.3). The results have been compared with SERCO results; the comparison is given in appendix G.

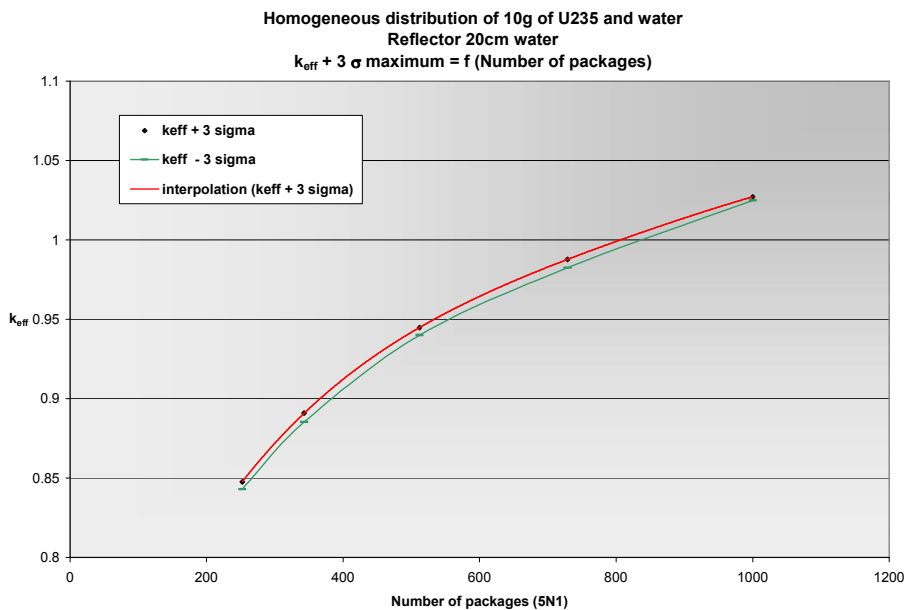
**3.1.1.1 CALCULATIONS WITHOUT A STEEL WRAPPER**

For each case, we calculated the multiplication factor for 3, 4 or 5 different arrays that give a  $k_{eff}$  around the value of 0.95. Actually, for each array, there is a maximum value of the multiplication factor. The optimum of moderation changes from a small array to a larger one due to the changes in leakages. For example, when we have 10 g of  $^{235}\text{U}$  homogeneously placed in a  $1000\text{ cm}^3$  package filled with water of density lower than 1, the results are as follows:



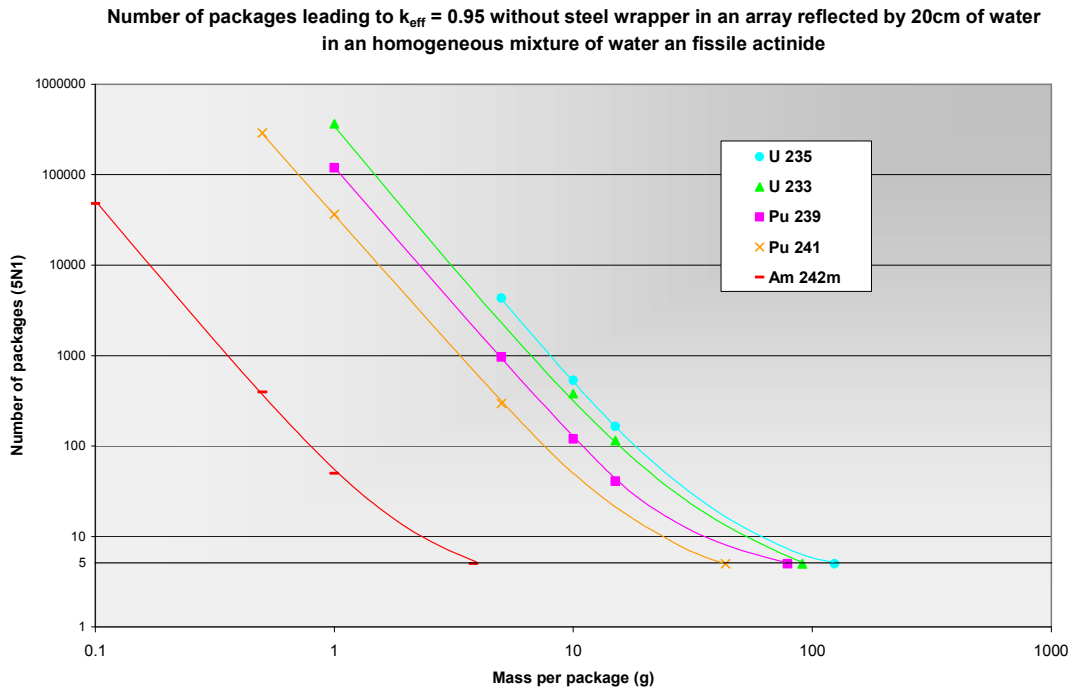
**Graph 1. Homogeneous distribution of 10 g of  $^{235}\text{U}$  and water -  $k_{eff} = f(\text{mass of water})$**

After those calculations, we plotted a graph of the maximum of the multiplication factor (+  $3\sigma$ ) for each array and did an interpolation to find out the number of packages that leads to  $k + 3\sigma = 0.95$ :



**Graph 2. Homogeneous distribution of 10 g of  $^{235}\text{U}$  and water -  $k_{eff} = f(\text{Number of packages})$**

For this example, the number of packages ( $5N_1$ ) leading to 0.95 is 533, so we can deduce that the number of transportable packages  $N_1$  is 106. We used the same method for each isotope and each mass. Finally, the results are plotted in the following graph:



**Graph 3. Number of allowable packages as a function of the mass of actinide nuclide per package\***

\*For 5 packages, we plotted the maximal allowable mass under a sphere form divided by 5 for each isotope reflected by 20 cm of water. This constitutes a conservative approach because the sphere minimizes the leakage comparing to an array of 5 cubic packages. Actually, the allowable mass per package will be greater for an array of 5 packages.

### 3.1.1.2 CALCULATIONS WITH A STEEL WRAPPER

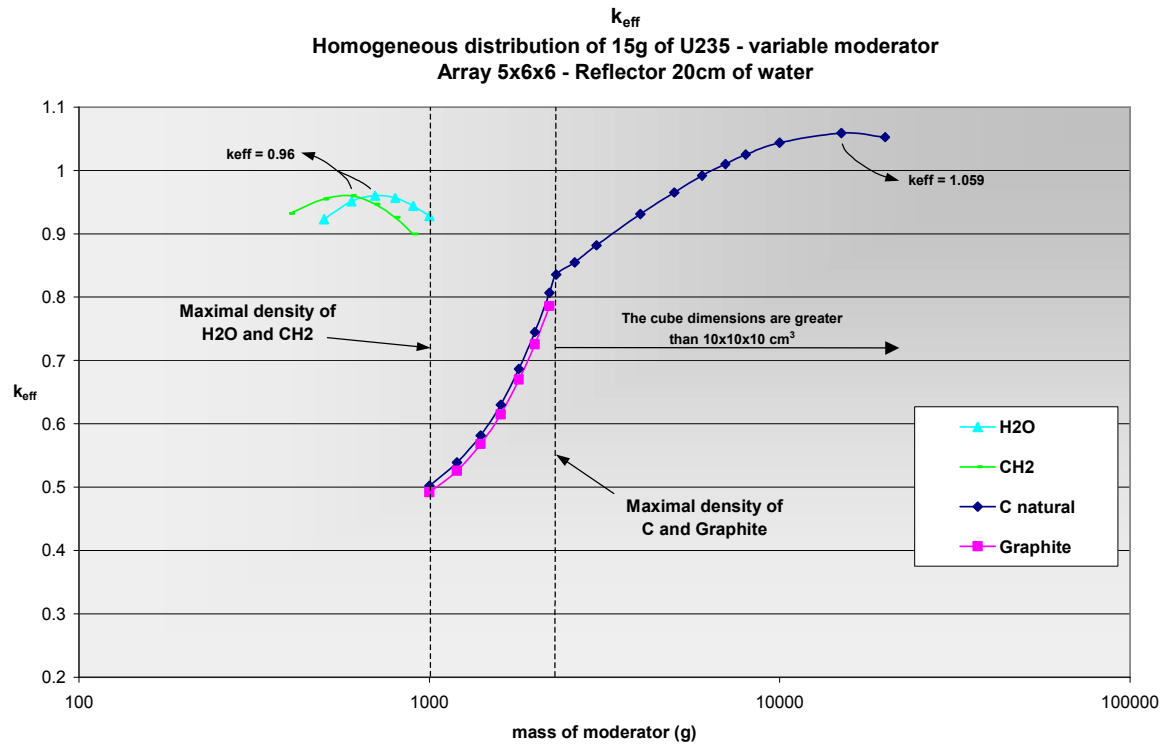
Due to the possibility to have **Type B** packages (for non-special form radioactive materials), we performed calculations for the thermal energy spectrum isotopes  $^{233}\text{U}$  and  $^{239}\text{Pu}$ . The results [6] confirms that, when packages include a wrapper of steel, it is possible to transport a higher number of packages for these two isotopes but also for the other thermal spectrum fissile isotopes, because, in this range of energy, the steel is mainly a neutron absorber. Therefore, the more the thickness of the steel wrapper increases, the higher the number of packages (that can be transported) is.

### 3.1.1.3 CALCULATIONS WITH OTHERS MODERATORS

We also checked the effect of other moderators than water that could lead to an increase of the effective multiplication factors. We studied moderators that are common materials as water:  $\text{CH}_2$ , natural carbon and graphite materials.

We only performed calculations with 15 g of <sup>235</sup>U in the package and we first checked those calculations for the 5x6x6 array; for water, this array leads to a maximum  $k_{eff}$  closer to 0.95.

We can see the results obtained in the following graph:



**Graph 4. Homogeneous distribution of 15 g of <sup>235</sup>U with different moderators**

We can deduce from those results that CH<sub>2</sub> moderator leads to the same value of maximum  $k_{eff}$  as water but with a lower mass of CH<sub>2</sub>, which is due to the higher concentration of Hydrogen per cubic centimetre. As we performed calculations with graphite and natural carbon moderators, we see that natural carbon leads to a higher  $k_{eff}$ . We first carried out calculations with a density of carbon inferior to its theoretical density (2.3 g/cm<sup>3</sup>), and, as reactivity increased, we reached this final density. So, we increased the quantity of carbon per package and the size of the package until we found the maximum  $k_{eff}$ . We found a difference of 10% in  $\Delta k$  with the water case!

An explanation is that carbon, although it has a lower scattering cross-section, has a very low capture cross-section comparing to water in thermal spectrum (see table below). As there is a large amount of carbon necessary to slow down neutrons enough, there are less absorptions in the array but more leakage of neutrons. It is the first effect that prevail and is characterized by the  $k$  infinite given for the maximum of reactivity:

	Water	Carbon
$\sigma$ scattering (b)	107	4.75
$\sigma$ capture (b)	0.644	0.0034
$k$ infinite	1.285	1.874

Of course, we can deduce from those results that packages containing graphite or carbon under any form (solid, powder or others) constitute a potential for criticality. This assertion has been taken into account when exceptions criteria have been defined (§ 4) and further calculations will have to ensure that the margins (safety factors) taken for the transport of actinide nuclides are sufficient enough to prevent such a risk (§ 4.2.1).

### 3.1.2 Calculations with fast spectrum fissile actinide nuclides

In the following paragraph, we present the calculations achieved with the CRISTAL package including the APOLLO 2 and MORET 4 programs (multigroups energy model). Then, we give the ones with steel (especially for the  $^{238}\text{Pu}$ ) with the TRIPOLI 4 program, which uses continuous energy data, as crosschecking calculations were performed by SERCO with MONK and showed discrepancies between APOLLO 2 - MORET 4 and MONK calculations in the case of  $^{238}\text{Pu}$  with a steel wrapper (see **appendix F**).

#### 3.1.2.1 *CALCULATIONS WITH APOLLO 2 – MORET 4*

We performed calculations with the 3 fast spectrum “fissile” isotopes  $^{237}\text{Np}$ ,  $^{244}\text{Cm}$  and  $^{238}\text{Pu}$ . In those cases, in the contrary of (thermal) fissile isotopes, the most reactive cases are found when the fissile material is concentrated. We have reached for each isotope the crystal density (metal density). An explanation is that, here, each package has an important quantity of actinide nuclide closer to the critical mass (approximately  $1/10^{\text{th}}$ ) comparing to the mass per package set for the (thermal) fissile actinide nuclides. This could explain the difference because each unit is very reactive. This configuration tends to reduce the leakage from each package even if it reduces the interactions between packages.

We also carried out calculations with a steel wrapper around the  $^{238}\text{Pu}$ : As we increased the steel thickness, the reactivity also increased. An explanation should be that there is a better reflection in each package when there is a steel wrapper around the package: effectively, we checked that the total leakage of neutrons outside the array is lower in this case (steel is a good reflector in fast spectrum).

When the mass in each package becomes less important, the most reactive configuration is when there is no steel wrapper (see table in **appendix F**). This phenomenon is due to two effects: When we decrease the mass per package, we go far from the allowable mass: each package is less reactive. The second effect is a spectrum effect: we checked from the listing files that the scattering is better with a lower mass of  $^{238}\text{Pu}$ , the spectrum becomes less tough and more neutrons are captured in the steel wrapper (the steel thickness is more important in this case because the volume of actinide nuclide (crystal) is lower).

We have also performed a calculation with a steel wrapper for  $^{244}\text{Cm}$  under a crystal form, but we found an opposite effect than with  $^{238}\text{Pu}$ : in this case, the reactivity is lower with a steel wrapper than the case with a “void” wrapper. An explanation comes from the fact that, in the case of  $^{244}\text{Cm}$ ,  $m(X)$  was equal to 1 kg (compared with critical mass around 15 kg) while, for  $\text{Pu}238$ ,  $m(X)=1$  kg but with a critical mass around 5 kg: in this second case the reactivity is mainly due to each unit of fissile matter and the reflection with steel increases the reactivity. Another explanation should come from the shape of the microscopic cross section of  $^{244}\text{Cm}$  (given in **appendix J**).

#### 3.1.2.2 *CALCULATIONS WITH TRIPOLI 4*

Crosschecking calculations were performed at SERCO with the MONK program and discrepancies were found with the cases treating of the fast fissile isotopes with a steel wrapper. Then, we checked our calculation with TRIPOLI 4 (continuous energy) for the  $^{238}\text{Pu}$  with a steel wrapper. We effectively found new multiplication factors of 4000 pcm less than the ones found with APOLLO 2 - MORET4 programs. An explanation comes from the fact that APOLLO 2 minimizes the steel absorption in fast spectrum because the multigroup model (172 gr) of APOLLO 2 in fast spectrum is not optimised.

So, new calculations were carried out to find the  $5N_1$  value leading to 0.95 with 1000 g and 500 g of  $^{238}\text{Pu}$  under a crystal form:

Actinides	Allowable mass g	Mass per package g	Steel thickness mm	Program used	5N1	N1
Pu 238 (crystal form)	3930	1000	31.5	CRISTAL (A2-M4)	48	9
				TRIPOLI 4	66	13
		500	35.325	CRISTAL (A2-M4)	761	152
				TRIPOLI 4	915	183

**Table 5. Calculations on  $^{238}\text{Pu}$  with a steel wrapper with TRIPOLI 4**

We can see that the number of packages that can be carried increase with the new calculations done with TRIPOLI 4 and fit with the results given by SERCO (**appendix G**).

However, the global conclusions of the calculations done with APOLLO2-MORET4 remains:

- the crystal form is the most reactive form for those actinide nuclides and approximately 1/10 of the critical mass per package,
- the steel wrapper increases the reactivity for the case of Pu238 with 1 kg of Pu238 per package.

### **3.1.3 Conclusions on Normal Conditions of Transport**

The most reactive configuration in thermal spectrum is the homogeneous distribution of fissile material and moderator (here, water<sup>11</sup>); the presence of a steel wrapper around the fissile material allows transporting more packages.

This is not the case with the fast spectrum fissible actinide nuclides were the most reactive form can be obtained when the fissible material is under a crystal form with void or steel around it. This last conclusion depends on the amount of fissile material put inside the package: If the mass of fissible material is close to the critical mass, the cases with a steel wrapper are the most reactive.

SERCO has performed crosschecking calculations of  $k_{\text{eff}}$  with MONK 9 for different configurations of fissile and fissible actinide nuclides.

The differences in  $\Delta k$  are less than 600 pcm (**appendix G**), which is consistent with other comparisons performed between the calculation tools, except for  $^{238}\text{Pu}$ , where the calculations performed by SERCO showed an over-estimation of the keff obtained in the IRSN calculations. Thus, IRSN performed calculations with a point-wise code (TRIPOLI 4) and found the same results as SERCO. The discrepancy came from the 172 groups cross-sections of the steel used in CRISTAL that under-estimate the absorption of the steel for a hard spectrum.

EMS has also performed some specific calculations for the  $k_{\text{eff}}$  values of arrays proposed by IRSN. The results are consistent with the ones obtained by IRSN and SERCO, except for fast spectrum cases. However the discrepancies are consistent with the discrepancies already obtained for the calculated critical masses of those actinide nuclides.

## **3.2 CRITICALITY RISK IN ACCIDENTAL CONDITIONS OF TRANSPORT**

We determined the number N (called  $N_2$ ) of packages that could lead to a criticality risk in accidental conditions. In order to be consistent with the regulation for fissile material, we consider that  $2N_2$  damaged packages can gather together (for accidental conditions).

Then, we calculated the number of packages  $2N_2$  leading to  $k_{\text{eff}} = 0.95$  (maximal allowable mass).

<sup>11</sup> Other mixtures than fissile material and water can be potentially more limiting



For certified packages, leakage from all packages must be collected into the most reactive form. Since we must assume that all fissionable material can escape from excepted packages, the assumption of a reflected sphere is consistent with the current IAEA Regulations.

We performed calculations of allowable masses with CRISTAL (APOLLO2 - Sn 2D (20 gr)) to determinate the  $N_2$  value [6]. For the cases with steel, we assumed that the steel wrapper is removed, in order to take the most penalizing case (when steel is homogeneously mixed with the fissile matter, the reactivity decrease mainly due to steel absorption). The fact of taking steel into account allows to carry a higher number of packages (see appendix 3 of reference [6]).

$N_2$  is calculated as follows:

$$N_2 = (1/2) * (\text{Allowable Mass}(X) / m(X)).$$

With  $m(X)$  the mass of actinide nuclide X per package.

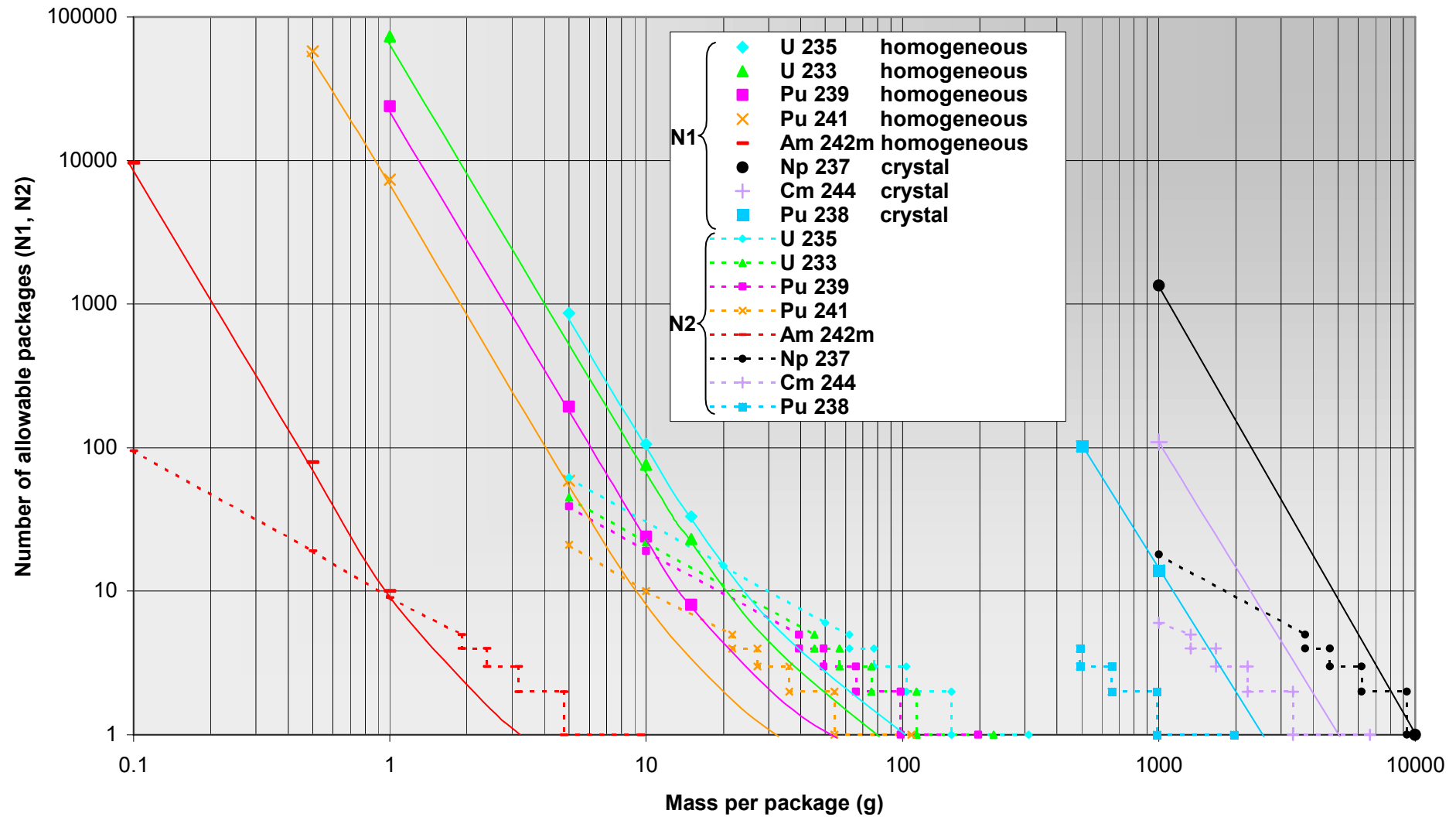
Allowable Mass is minimum mass of X in a sphere (with the reflector - water, steel or lead- that gives the lowest value) that gives a  $k$ -effective equal to 0.95

Calculations have been performed with the same 8 nuclides as for the normal conditions of transport:  $^{235}\text{U}$ ,  $^{233}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{242\text{m}}\text{Am}$  in thermal energy spectrum and  $^{237}\text{Np}$ ,  $^{244}\text{Cm}$  and  $^{238}\text{Pu}$  in fast energy spectrum.

### **3.3 COMPARISON BETWEEN THE NORMAL AND THE ACCIDENTAL CONDITIONS**

The results of the whole calculations are shown in **appendix F** which gives the values of  $N_1$ ,  $N_2$ ,  $N'_2$  (calculated for  $k_{\text{eff}} = 1$ ) and  $N$ , which is the minimal value of these 3 values and in the following graph ( $N_1$ ,  $N_2$ ):

**Number of allowable packages (N1, N2) without steel wrapper**  
**U235, U233, Pu239, Pu241 and Am242m: array reflected by 20cm of water in an homogeneous mixture of water and fissile actinide**  
**Np237, Cm244 and Pu238: array reflected by 30cm of steel with a crystal density cube in the center of each package**



**Graph 5. Number of allowable packages (N<sub>1</sub>, N<sub>2</sub>) as a function of the mass per package**

The **Graph 5**, gives the number  $N$  depending on the mass  $m(X)$  per package (for the 8 actinide nuclides studied). It shows that the accidental conditions are always more penalizing than the normal conditions up to a given amount of mass of actinide nuclide per package. For big masses of actinide nuclides per packages, the normal cases become more reactive.

When the number of packages allowed gets close to 1 package (this means we are close to the allowable mass of each actinide nuclide), the most reactive case is obtained with the normal conditions of transport (for example, with 15 g of  $^{239}\text{Pu}$  per package,  $N_1=8$  and  $N_2=13$ ).

Considering those results, exception criteria will be defined for each actinide nuclide (see § 4).

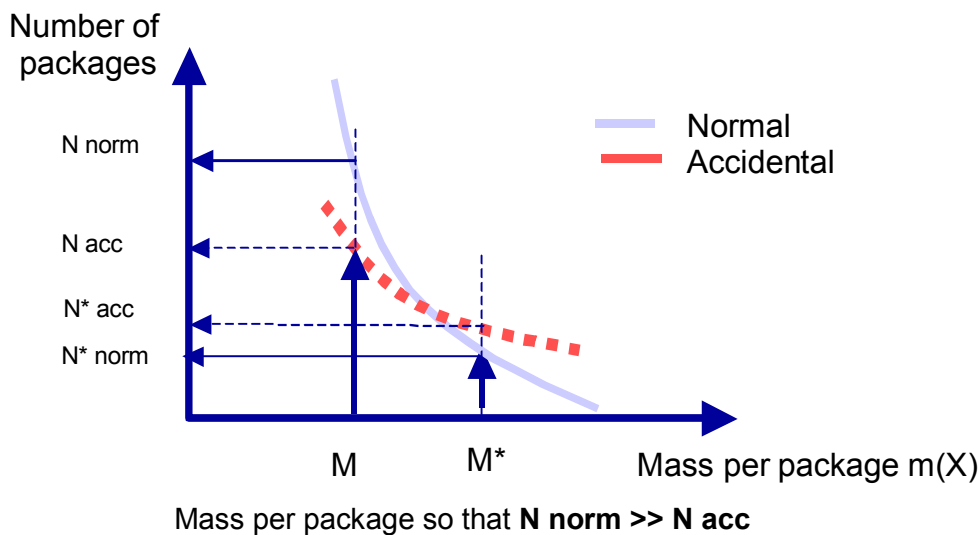
#### 4. METHOD FOR THE DEFINITION OF EXCEPTION CRITERIA

As calculations were performed to determine the most penalizing case between normal and accidental conditions of transport, it is now possible to set a rule and give mass limits for the excepted fissionable nuclides packages, with those data as starting point. The rule and the exception limits will be extended to the others actinide nuclides.

We presented in the previous paragraph (§ 3) the work achieved to determine the number of allowable packages ( $N$ ) that can be transported in normal and accidental conditions for different masses of actinide nuclide per package. The normal conditions consist on an array of  $5 \times N$  packages of  $10 \times 10 \times 10 \text{ cm}^3$  containing  $m(X)$  g of a given actinide nuclide, with water or moisture. The accidental conditions consist of a spherical mass of  $2 \times N \times m(X)$  g of a given actinide nuclide.

Then the principles to set the rule are the following:

1. The accidental configuration should be the most penalizing<sup>12</sup>



**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 penalizing than the normal ones (graph 5)**

<sup>12</sup> EMS doesn't consider that accidental configuration should necessarily be the most penalizing.

2. Take into account every environment and possible configurations (reflectors, moderators, mixtures ...); if the package is excepted there will be no possibility to re-check the real configurations of transport.

3. Research the minimum of package leading to a criticality risk and check that this number is greater or equal to 10 ( $N > 10$ ); it will guaranty a certain dilution of the matter and reduce the consequences of human errors.

4. Keep the limit of 15 g per package for U235 that exists in the current regulation.

The following paragraphs will therefore concern the determination of the mass per package allowed in accidental conditions:

- considering that this evaluation is linked to the accidental conditions; consequently, it can be directly derived from the critical mass of each actinide nuclide,
- studying the effect of additional parameters that can lead to an increase of the reactivity (different reflectors and moderators, non uniform repartition of the moderation, mixture of actinide nuclides); this will give some "safety factors"<sup>13</sup> to apply on the critical mass to take those phenomena into account;
- using the minimum number  $N^\circ$  of packages containing 15 g of U235 that are safe in every condition of transport. This number  $N^\circ$  will be then used for every actinide nuclide as the minimum of packages needed to have a criticality risk in every condition of transport (it will be checked that  $2N^\circ$  packages in accidental condition and  $5N^\circ$  packages in normal condition are safe); It will be verified that  $N^\circ$  is greater than 10.

Finally the mass limit per package will be calculated and it will be verified that the most penalizing configurations are the accidental ones.

#### **4.1 ADDITIONAL PARAMETERS LEADING TO AN INCREASE OF REACTIVITY**

This paragraph gives the effects of parameters that may have an influence on the reactivity, such as others moderators than "standard" water moderator or others reflectors than "standard" water (thermal spectrum) or steel (fast spectrum) (see § 4.1.1) reflectors; they need to be considered in the definition of the exception criteria (§ 4.1.2). Non-uniform repartition of fissile material (§ 4.2.2), mixture of actinide nuclides (§ 4.1.3) will be checked.

##### **4.1.1 Different moderators and reflectors (F1)**

EMS has performed some calculations to study the effect of:

- different types of reflectors: water, stainless steel, lead, water plus steel or lead,
- for diluted systems, (due to the 10 cm minimum dimension), different types of neutron scatterers such as water, carbon powder (including carbonised or de-hydrated materials, lead and steel) and combinaison of these.

The calculations showed that the  $k_{\text{eff}}$  can increase by more than:

<sup>13</sup> The exception limits should preferably be based on measured data supported by critical experiments as well as reflection, moderation, and mixed configuration conditions optimised for each nuclide. In the absence of such data, experiments and evaluations for many nuclides, safety factors based on experience with "similar" nuclides and conditions may be derived in a conservative way. The safety factor method should be sufficiently conservative to encourage derivation of better data and critical experiments when a large quantity of each nuclide is available. A similar practice can be applied to derivation of mass limits for mixtures (e.g. specific isotope distributions for actinide elements, solutions, etc.) as well as to concentration and other limits.

At the moment there are too many issues that are still under discussion to motivate determination of final exception limits. The project has identified several new issues that have not been considered in existing transport regulations. During 2003 there will be discussions and meetings where most of these issues will be considered. This project as well as continued efforts in France, UK and Sweden will give important input to the discussions.

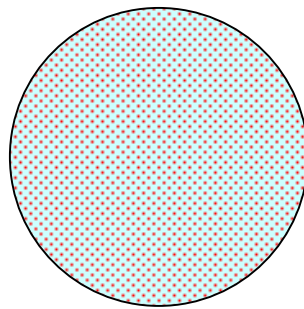
- 5% in  $\Delta k$  with a reflector different from the commonly used ones (water for water-moderated systems and steel for fast spectrum systems),
- 10% in  $\Delta k$  for diluted systems due to the added scatterers such as carbon powder in addition to water or void.

For reflector, different types of reflectors have already been checked (**appendix K**), and the critical value considered is obtained with the most penalizing reflector.

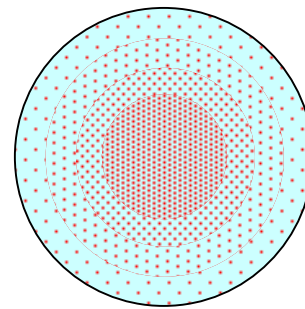
Therefore, the factor F1 mainly depends on the moderator effects. As a first step, it will be set to  $0.8^{14}$  for thermal systems and to 1 for fast spectrum ones: for fast systems, the various moderators don't play any role.

#### 4.1.2 Non uniform repartition of the fissile material (gradient of concentrations)

According to criticality general assessments, a critical fissile material with an optimum moderation ratio can become more reactive if a non uniform distribution of the fissile material and the moderator happen with an appropriate gradient of concentration: calculations demonstrated that for a critical mass, non uniform concentrations with different optimised H/X ratios could lead to an increase of reactivity. This phenomenon is mainly due to the reduction of leakages as the most part of fissile material is concentrated in the centre of the package:



Homogeneous distribution of the fissile material and water



Gradient of fissile material and water

#### 4.1.3 Mixture of actinides nuclides

EMS [1] checked the effect of mixing two actinide nuclides in different ways. We wanted to see if the presence of two nuclides X and Y in a package could lead to increase the reactivity, as we put in the package a given ratio of the critical mass  $M_c(X)$  and  $M_c(Y)$  of each nuclide (indeed  $m(X)/M_c(X) + m(Y)/M_c(Y) = 1$ ). The equation given in the Introduction and used in the transport regulations is a typical application of the Rule of Fractions.

Homogeneous and heterogeneous configurations were studied with different ratios. MCNP with continuous energy cross-sections has been used rather than SCALE with multigroup cross-sections to reduce the potential for errors due to incorrect resonance treatment.

A useful study on the commonly used method to assume under-criticality by the Rule Of Fraction (ROF) when there are two or more nuclides per package has been done [5]. For a specific nuclide  $i$  with a mass  $m_i$  and a minimum critical mass  $M_i$ , the Fraction of Critical Mass  $FCM_i$  is defined as  $m_i/M_i$ . The Rule Of Fractions is based on the following assumption:

$$\sum_i FCM_i \leq 1.0$$

<sup>14</sup> Actually, when they exist, the comparisons of the results of the calculations ( $k_{eff}$ ) with various moderators or reflectors will be used to determine the value of F1.

If the mass  $M_i$  is not exactly critical, a corresponding value of  $k_{\text{eff}}$  can be calculated. The more general Rule Of Fractions will be:

$$\sum_i FRM_i \leq 1.0$$

where  $FRM_i$  is a reference value. For a mixture of two nuclides the ROF means that the resulting  $k_{\text{eff}}$  for the mixture is equal to or lower than the interpolated value between the two reference values for the individual nuclides.

A reason for not using the exact critical mass  $FCM_i$  is that different cross sections will give variations of the critical mass. The general ROF will give the same information without the need for calculating exact minimum critical masses for each cross section set.

There are ways to conservatively cover cases where the ROF does not apply directly. One way is to shift the interpolated line so that all mixtures will give  $k_{\text{eff}}$  below the line. This requires studies of each potential mixture.

The studies showed that the ROF is not a conservative method both in homogeneous and heterogeneous cases.  $k$  effective of mixtures of actinide nuclides can be 10% in  $\Delta k$  higher than the  $k$  effective calculated with the ROF. As we define in § 4.3 a new CSI rule, we should consider the potential risk for the mixtures of actinide nuclides<sup>15</sup>.

## 4.2 SAFETY MARGINS DETERMINATION AND NEW RULE (CSI) DEFINITION

The different actinide nuclides have been defined as Fissile "S" (that can sustain a chain reaction with thermal neutrons – "S"low neutrons) or as "F" actinide nuclides (that can sustain a chain reaction only with hard spectrum – "F"ast neutrons). The list is given in **appendix H**. The actinide nuclides in pink are neither fissile nor fissionable alone.

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 penalizing 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 un-known phenomenon (mixture, repartition of the fissile matter); 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 condition must be greater than 10 (a human error leading to a 2x10 package array should be safe).

**The consignment limit** will be introduced and should be less than half of the critical mass.

The excepted mass per package could therefore be determined as follows:

<sup>15</sup> Natural uranium is a good start for evaluation of mixing effects. Natural uranium is completely excepted from criticality control in the transport regulations. Consideration is of course required if it is credible in the contents of certified packages, but for excepted packages there is no such control. Natural uranium is fissionable material that is not fissile or fissile in water (it is fissile in mixtures with heavy water or graphite). It will significantly reduce the critical mass for many nuclides. EMS is studying this mixing configuration for all nuclides out of the scope of this project.

- 1- define the minimum of the **critical mass  $M_c(\min)$**  obtained for commons moderators (water or mist of water) and reflectors (water for thermal energy spectrum or steel for fast energy spectrum) obtained with the different codes (see **Table 4 § 2.5**),
- 2- set a **reflector/moderator correction factor  $F_1$**  for the critical mass to take into account the possibility of other reflectors (**see appendix K**), other moderators (for fissile actinide nuclide only), heterogeneity of the moderator (for fissile actinide nuclides only) (see **§ 4.2.1 and 4.2.2**); as a first step, this factor has been set to 0.8 for fissile actinide nuclides and set to 1 for the others (this factor could be revised),
- 3- set a **margin correction factor  $F_2$** , to take into account a safety margin of 5% in  $\Delta k$ ; this factor has been derived from calculations when available, otherwise it has been set to 0.8 (we can note here that the value obtained with this factor applied to the critical mass ( $k_{\text{eff}}=1$ ) is very close to the values of allowable masses ( $k_{\text{eff}}=0.95$ ) calculated), (**Appendix H**)
- 4- set a **library correction factor  $F_3$** , due to the libraries uncertainties; this factor will be determined by the discrepancies observed between the codes used for the calculations of the critical masses; if no comparison exists the factor will be set, as a first approach to 0.2)
- 5- determine **safe mass  $M_{\text{safe}} = F_1.F_2.F_3.M_c(\min)$** ,
- 6- determine the **mass allowed per group of packages  $M_{\text{group}} = M_{\text{safe}}/2$** ;  
1/2 is the same safety factor as in the 2N array to be assessed in accidental situations. This safety factor is also justified in section 8 below.
- 7- determine the **mass allowed per package  $m^* = M_{\text{group}}/12$** ;  
this number of 12 has been set to find a value of 15 g per package for  $^{235}\text{U}$ . It allows to keep the same safety margins for the transport of fissile excepted  $^{235}\text{U}$  as those derived from the requirements of the current regulations. It comes  **$m^* = F_1.F_2.F_3.M_c(\min)/24$**
- 8- Check on the **Graph 5 § 3.3**, that for the mass  $m^*$ , the number of allowable packages in accidental condition ( $N_2(m^*)$ ) is less or equal to the number of allowable packages in normal conditions ( $N_1(m^*)$ ).

Then the Criticality Safety Index for the package will be set as:

$$\text{CSI} = 50 \sum_X \frac{m(X)}{M_{\text{group}}(X)}, \text{ with } m(X) \leq m^*(X)$$

Where  $m(X)$  is the actual transported amount of the isotope X per package and  $M_{\text{group}}(X)$  is half the Safe Mass of the isotope X..

An additional rule is set considering that "if the total mass of a group of actinide nuclides is less than 10 mg, they do not need to be taken into account"; indeed the participants think that a mass of less than 10 mg of fissile isotopes cannot have a significant impact on the reactivity as it is less than 1/30 of the minimum  $m^*(X)$  obtained for any of the actinide nuclides X.

Finally the rule is the following:

$$\text{If } \sum_{X_j} m(X_j) < 10 \text{ mg then } m(X_j) = 0$$

$$\text{And CSI} = 50 \sum_{X \neq X_j} \frac{m(X)}{M_{\text{group}}(X)}, \text{ with } m(X) \leq m^*(X)$$

To determine the mass allowed per package, we took in consideration safety margins including the effect of different reflectors and moderators, the libraries uncertainties and a safety margin of 5% in k-effective to get the safe mass. We also allowed a very low mass per package as we divided by  $2 \cdot 12$  this safe mass.

The effect of mixing nuclides has not been evaluated properly. Without such evaluations it would be against criticality safety principles to neglect the issue. At least for nuclides and mixtures that are credible today or in the near future, some evaluation is required. Heterogeneous mixtures of fissile or fissionable nuclides with natural uranium metal are such a case.

However, as the correction factors give very conservative mass limits (leading to safe masses far from the critical mass of each nuclide, both in normal and accidental cases), **we didn't use a safety margin** to take into account the **effect of the mixtures of actinide nuclides (§ 4.2.3)**, even if the ROF is not acceptable, **because those precautions seem sufficient, at a first stage, to prevent the criticality risk.**

### 4.3 EXCEPTION MASS LIMITS

The table 5 gives the results obtained for the different actinide nuclides.

The values are not the definitive values as they still have to be crosschecked and internationally discussed (critical mass and, mainly, correction factors).

In appendix H, an additional value, in the last column is given as "ANS". This is the value of  $m^*$  (maximum mass per package) obtained with the method described above while considering that the safe mass is equal to the minimum of the critical mass obtained multiplied by the factor proposed by the ANS 8.15 group (noted  $F_{ANS}$ ); then the maximum mass allowed per package  $m^*(X)_{ANS}$  is:

$$m^*(X)_{ANS} = \frac{M_{crit}(X) \times F_{ANS}}{2 \times 12}$$



**Table 5. Calculation example of Exception Limits and Corrections factors**

Nuclide	Critical Mass Min	Factors to consider on Mc			Safe Mc (g)	Mass per group of package (g)	Mass per package (g)
		heterogeneity reflectors moderator	Margin of 5% in $\Delta k$	Libraries			
<sup>227</sup> Ac							
<sup>228</sup> Th							
<sup>229</sup> Th	<b>994</b>	-	-	-	-	-	-
<sup>230</sup> Th							
<sup>232</sup> Th							
<sup>231</sup> Pa							
<sup>232</sup> U	<b>1850</b>	1	0.8	0.95	1406	703	58
<sup>233</sup> U	<b>425</b>	0.8	0.8	0.95	258	129	10
<sup>234</sup> U	<b>83000</b>	1	0.8	0.95	63080	31540	2628
<sup>235</sup> U	<b>589</b>	0.8	0.8	1	376	188	15
<sup>236</sup> U							
<sup>238</sup> U							
<sup>235</sup> Np	<b>6180</b>	1	0.8	0.2	6208	3104	258
<sup>236</sup> Np	<b>63</b>	0.8	0.8	0.2	8	4	0.3
<sup>237</sup> Np	<b>38600</b>	1	0.75	0.7	20265	10132	844
<sup>236</sup> Pu	<b>970</b>	1	0.8	0.9	2692	1346	112
<sup>237</sup> Pu	<b>110</b>	0.8	0.8	0.2	14	7	0.5
<sup>238</sup> Pu	<b>4700</b>	1	0.8	1	3760	1880	156
<sup>239</sup> Pu	<b>353</b>	0.8	0.8	1	225	112	9.3
<sup>240</sup> Pu	<b>18300</b>	1	0.8	0.85	13464	6732	561
<sup>241</sup> Pu	<b>205</b>	0.8	0.8	0.9	118	59	4.9
<sup>242</sup> Pu	<b>36200</b>	1	0.75	0.9	29862	14931	1244
<sup>244</sup> Pu	<b>60300</b>	1	0.8	0.2	21280	10640	886
<sup>241</sup> Am	<b>33800</b>	1	0.7	0.75	17745	8872	739
<sup>242m</sup> Am	<b>17</b>	0.8	0.8	0.85	9	4.5	0.37
<sup>243</sup> Am	<b>88600</b>	1	0.7	0.95	88012	44006	3667
<sup>242</sup> Cm	<b>7720</b>	1	0.8	0.2	1956	978	81
<sup>243</sup> Cm	<b>106</b>	0.8	0.8	0.5	33	16.5	1.37
<sup>244</sup> Cm	<b>13200</b>	1	0.85	0.75	8415	4207	350
<sup>245</sup> Cm	<b>42</b>	0.8	0.8	0.85	22	11	0.91
<sup>246</sup> Cm	<b>22000</b>	1	0.8	0.2	3520	1760	146
<sup>247</sup> Cm	<b>796</b>	0.8	0.8	0.2	183	91	7
<sup>248</sup> Cm	<b>21500</b>	1	0.8	0.2	3440	1720	143
<sup>250</sup> Cm	<b>14700</b>	1	0.8	0.2	2240	1120	93
<sup>247</sup> Bk	<b>35200</b>	1	0.8	0.2	5568	2784	232
<sup>248</sup> Bk	-	1	1	0.2	-	-	-
<sup>249</sup> Bk	<b>131000</b>	1	0.8	0.2	20960	10480	873
<sup>248</sup> Cf	-	1	0.8	0.2	-	-	-
<sup>249</sup> Cf	<b>51</b>	0.8	0.8	0.2	6	3	0.25
<sup>250</sup> Cf	<b>3130</b>	1	0.8	0.2	500	250	20
<sup>251</sup> Cf	<b>20</b>	0.8	0.8	0.2	2	1	0.083
<sup>252</sup> Cf	<b>2910</b>	1	0.8	0.2	465	232	19
<sup>254</sup> Cf	<b>2250</b>	1	0.8	0.2	353	176	14
<sup>252</sup> Es	-	1	0.8	0.2	-	-	-
<sup>254</sup> Es	<b>32</b>	1	0.8	0.2	5.1	2.5	0.20
<sup>257</sup> Fm	-	1	0.8	0.2	-	-	-
<sup>258a</sup> Md	-	1	0.8	0.2	-	-	-

The complete table is given in appendix H.

For those limits, it was checked, for U235, that a carbon moderator would not lead to a unacceptable situation. For that purpose, calculations were performed at IRSN with 15 g of  $^{235}\text{U}$  moderated with carbon (see § 3.1.1.3) in normal conditions (for accidental conditions, a water moderator is more penalizing). Results showed that, for normal conditions, carbon leads to a higher  $k_{\text{eff}}$  than water, but the safety factors proposed give a safety margin of 20% in  $\Delta k$  (**appendix I**). For the accidental conditions of transport, we checked that the minimal allowable/critical mass is always smaller with water. Even if  $k$  infinite is smaller with water, leakages are higher with carbon; then, in order to get the allowable/critical mass, the sphere of fissile material ( $^{235}\text{U}$ ) and moderator must be larger with carbon than with water. This leads to a higher mass of fissile material to obtain criticality with carbon. This consideration allows us to keep the safety rule set with water, for U235 (see § 4.3).

Moreover, for those limits for U235, the following results were obtained (with water moderator and water reflector only):

- 5N (N=12) un-damaged packages containing each 15 g of U235 gives a maximum keffective of  $k_{\text{eff}} + 3\sigma = 0.82235$  ( $\sigma = 0.1\%$ ),
- 2N packages in spherical form (total of 360 g of U235), gives a maximum k effective of 0.827.

For those special calculations, the accidental case is slightly more conservative than the normal one and we can observe the margins in terms of reactivity when we apply a safety factor on the mass (a factor 50% on the critical mass of water-moderated systems gives for U235 a reduction of 20% in  $\Delta k$ ).

## 5. CONCLUSION

The work planned for the Phase I, Phase II, Phase III and Phase IV of the DG TREN project consisted in:

- agreeing the work plan between the participants (phase 1); the inception report related to this first phase was sent to the European commission the 11<sup>th</sup> of April 2002 (IRSN/DPEA/SEC/A/02.112),
- defining the broader list of radionuclides that should be include in the regulation, review the existing work and experiences (phase 2),
- performing calculations for, at least, the first 15 selected actinide nuclides defined in the inception report (phase 3),
- determining exception limits per package and per consignment (and possibly per conveyance) for all fissionable nuclides that we will propose for inclusion in the regulation. The main objective of this programme is to adapt the existing rule (derived from the woodcock & Paxton calculations) for the special actinide nuclides or to define a new methodology.

The bibliographic survey has been achieved; the result is given in the interim report [5]. For the minor actinides, it shows some great differences on the criticality values (k-infinite and critical masses) depending on the evaluation used for the cross sections and on the codes used.

To explain those discrepancies, some studies were realized based on a comparison of the different evaluations, especially on the NUBAR values. It pointed out some great differences on this parameter for  $^{236}\text{Np}$  and  $^{232}\text{U}$  depending on the evaluation used; however it also showed that the current libraries are satisfying for the "major actinides". For the minor actinides, the differences are somehow covered by the uncertainties on the cross-section evaluations themselves...

The second step involves definition of nuclides that can sustain a chain reaction. K-inf is the parameter that determines this possibility. Eta as a function of energy can be used to guess this property but is not sufficient in many cases. Eta can be used to include fissionable nuclides that are not fissile or fissible but has eta larger than 1.0 in some energy range. Such fissionable nuclides may increase the potential for criticality in mixtures with fissile or fissible nuclides.

The list of actinide nuclide able of sustaining a chain reaction is given in the next table.

Element	Nuclide	Spectrum	Density (g/cm <sup>3</sup> )	T <sub>1/2</sub> (y)
Actinium	<sup>227</sup> Ac	NF	10.043	21.77
Thorium	<sup>228</sup> Th	NF?	11.524	1.913
	<sup>229</sup> Th	F	11.575	7900 (7340)
	<sup>230</sup> Th	NF	11.626	75400
	<sup>232</sup> Th	NF	11.727	1.4 x 10 <sup>10</sup>
Protactinium	<sup>231</sup> Pa	NF?	15.336	32500
Uranium	<sup>232</sup> U	S	18.681	69.8
	<sup>233</sup> U	S	18.762	1.59 x 10 <sup>5</sup>
	<sup>234</sup> U	F	18.842	2.45 x 10 <sup>5</sup>
	<sup>235</sup> U	S	18.923	7.04 x 10 <sup>8</sup>
	<sup>236</sup> U	NF	19.004	2.34 x 10 <sup>7</sup>
	<sup>238</sup> U	NF	19.165	4.46 x 10 <sup>9</sup>
Neptunium	<sup>235</sup> Np	F	20.303	1.058
	<sup>236</sup> Np	S	20.389	155000
	<sup>237</sup> Np	F	20.476	2.14 x 10 <sup>6</sup>
Plutonium	<sup>236</sup> Pu	S	19.601	2.87
	<sup>237</sup> Pu	S	19.685	45.7 d
	<sup>238</sup> Pu	F	19.768	87.74
	<sup>239</sup> Pu	S	19.851	24110
	<sup>240</sup> Pu	F	19.934	6537
	<sup>241</sup> Pu	S	20.017	14.4
	<sup>242</sup> Pu	F	20.101	3.76 x 10 <sup>5</sup>
	<sup>244</sup> Pu	F	20.267	8.2 x 10 <sup>7</sup>
Americium	<sup>241</sup> Am	F	13.66	432.2
	<sup>242m</sup> Am	S	13.717	141
	<sup>243</sup> Am	F	13.774	7370
Curium	<sup>242</sup> Cm	F	13.407	162.8 d
	<sup>243</sup> Cm	S	13.463	28.5
	<sup>244</sup> Cm	F	13.518	18,11
	<sup>245</sup> Cm	S	13.574	8500
	<sup>246</sup> Cm	F	13.629	4780
	<sup>247</sup> Cm	S	13.685	1.56 x 10 <sup>7</sup>
	<sup>248</sup> Cm	F	13.74	3.4 x 10 <sup>5</sup>
	<sup>250</sup> Cm	F	13.851	9700 (8000)
Berkelium	<sup>247</sup> Bk	S	14.671	1400
	<sup>248</sup> Bk	S?	14.731	9
	<sup>249</sup> Bk	F	14.79	320 d
Californium	<sup>248</sup> Cf	F	15.05	333.5 d
	<sup>249</sup> Cf	S	15.11	351
	<sup>250</sup> Cf	F	15.171	13.1
	<sup>251</sup> Cf	S	15.232	900
	<sup>252</sup> Cf	S	15.292	2.64
	<sup>254</sup> Cf	F	15.412	60.5 d
Einsteinium	<sup>252</sup> Es	S?	8.808	1.29
	<sup>254</sup> Es	S	8.878	276 d
Fermium	<sup>257</sup> Fm	S?		100.5 d
Mendelevium	<sup>258</sup> Md	S?		51.5 d

NF

Non-Fissile non-fissile isotope

S

Slow spectrum fissile isotope

F

Fast spectrum fissile isotope

The third step consisted in calculations of the critical masses of the 15 first isotopes of the list with the different codes available among the participants (i) ENDF/B-5 (and other libraries) and MCNP for EMS (ii) JEF2.2 with APOLLO2 (172 groups) or TRIPOLI (continuous in energy).

In step 4, the participants performed calculations to determine the maximum number of packages for a given mass per package leading to an acceptable value of the k-effective; this was realized for arrays of packages in normal and accidental conditions.

Then the principle to define the exception criteria were the following:

1. the accidental conditions must be more penalizing than the normal conditions; therefore the calculations of the mass limit per package for a given number of package will be derived from the calculations of the critical masses (for the accidental conditions, the fissile matter forms a sphere); for the maximum mass per package and associated maximum of packages, it will be checked that the accidental conditions are more penalizing than the normal ones,
2. the different parameters that may increase the reactivity must be considered,
3. the current limit of 15 g/package for U235 will be kept,
4. the minimum number of packages that should lead to a criticality hazard is greater than 10.

Finally the rule is the following:

$$\text{If } \sum_{X_j} m(X_j) < 10 \text{ mg then } m(X_j)=0$$

$$\text{And CSI} = 50 \sum_{X \neq X_j} \frac{m(X)}{M_{\text{group}}(X)}, \text{ with } m(X) \leq m^*(X)$$

With  $M_{\text{group}}(X)$  and  $m^*(X)$ , for each actinide nuclide X, given (as preliminary results) in the following chart:

This proposition is consistent with the UK proposition for change of the IAEA regulation number UK/02-07.

**The values in the next table are not proposed limits but only calculation examples.**

Table 6 Calculation Example of Exception Limits

Nuclide	Mass per group of package (g) M group	Mass per package (g) m*(X)
<sup>227</sup> Ac		
<sup>228</sup> Th		
<sup>229</sup> Th	x	x
<sup>230</sup> Th		
<sup>232</sup> Th		
<sup>231</sup> Pa		
<sup>232</sup> U	703	58
<sup>233</sup> U	129	10
<sup>234</sup> U	31540	2628
<sup>235</sup> U	188	15
<sup>236</sup> U		
<sup>238</sup> U		
<sup>235</sup> Np	494	41
<sup>236</sup> Np	4	0.3
<sup>237</sup> Np	10132	844
<sup>236</sup> Pu	349	29
<sup>237</sup> Pu	7	0.5
<sup>238</sup> Pu	1880	156
<sup>239</sup> Pu	112	9.3
<sup>240</sup> Pu	6222	518
<sup>241</sup> Pu	59	4.9
<sup>242</sup> Pu	12217	1018
<sup>244</sup> Pu	4824	402
<sup>241</sup> Am	8872	739
<sup>242m</sup> Am	4.5	0.37
<sup>243</sup> Am	29459	2454
<sup>242</sup> Cm	617	51
<sup>243</sup> Cm	16.5	1.37
<sup>244</sup> Cm	4207	350
<sup>245</sup> Cm	11	0.91
<sup>246</sup> Cm	1760	146
<sup>247</sup> Cm	50	4
<sup>248</sup> Cm	1720	143
<sup>250</sup> Cm	1176	98
<sup>247</sup> Bk	2816	234
<sup>248</sup> Bk	x	x
<sup>249</sup> Bk	10480	873
<sup>248</sup> Cf	x	x
<sup>249</sup> Cf	3	0.25
<sup>250</sup> Cf	250	20
<sup>251</sup> Cf	1	0.083
<sup>252</sup> Cf	232	19
<sup>254</sup> Cf	180	15
<sup>252</sup> Es	x	x
<sup>254</sup> Es	2.5	0.20
<sup>257</sup> Fm	x	x
<sup>258a</sup> Md	x	x

Possible additional work

During the work achieved many questions were raised and will need additional work to be answered. This is the case of:

- **mixture of fissionable actinide nuclides and fissile/fissible ones;** the note [1] gives preliminary results on that subject;
- **influence of the radioactive decay on the criticality risk.** Actually, the reactivity of a package could increase during a certain period with the radioactive decay of actinides nuclides. A preliminary study [8] has been done with the production/decay chains of 6 actinide nuclides ( $^{232}\text{U}$ ,  $^{233}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ,  $^{240}\text{Pu}$ ,  $^{242}\text{Pu}$ ) including almost every fissile (or fissionable) nuclide that have been selected. The goal of this study was to find the way the actinide nuclides are formed by  $\alpha$ ,  $\beta^+$  or  $\beta^-$  disintegrations and try to see if a non-fissile actinide nuclide or a "low reactive" actinide nuclide can decay into a "high reactive" one. This means that if an isotope X decays to a more reactive isotope Y, the limit mass for X will be the one used for the isotope Y<sup>16</sup>. For each isotope transported, the study gives which limit of mass should be applied, according to the exception criteria (§ 4.4) set in **appendix H**;
- **use and determination of the safety factors.**

Those questions will be debated during the IAEA technical Committee Meeting that will be held in Autumn 2003 on the subject of fissile excepted actinide nuclides.

However, some additional work will need to be achieved on the other exception rules currently existing in the IAEA regulation [7] para 672; it concerns the fissile material excepted from the fissile requirements due to:

- para. 672 a) ii), a ratio of fissile nuclides on hydrogen,
- para. 672 a) iii), a concentration limit of fissile nuclide.

Both of those exception rules need to consider a limit per "group of packages" to take into account accidental conditions (precipitation, etc.).

Moreover, the current paragraph 672 d) may need to be revised due to the results obtained during this project.

<sup>16</sup> Increase of reactivity due to radioactive decay must be considered both in derivation of exception limits and in safety assessments for package designs requiring authority approval. Fissionable nuclides such as Pa-233 with quite short half-lives can clearly increase reactivity if shipped not too long after production. Within a year, essentially all of it will decay into the fissile nuclide U-233. For nuclides with longer half-lives, the maximum length of the transport needs to be considered. If somewhere up to between a one year and a hundred years period of time is considered, radioactive decay of nuclides such as Am-243 with very long half-lives do not need to be considered. The combined effect of nuclides gaining reactivity and of nuclides losing reactivity can be considered.

**REFERENCES**

- [1] EMS/NC/2002-01 - "Basic criticality data and exception limits for actinide nuclides"  
Draft report, 31 July 2002, D.MENNERDAHL
- [2] R.M. WESTFALL densities of actinides Metals and Oxides Based on Crystallographic Data,  
Obtained through the ANS 8.15 group
- [3] N.J. BARTON, C.K. WILSON, "Review of Fissile Exception Criteria in IAEA Regulations",  
pp. 9.15-9.22, ICNC '95, The Fifth International Conference on Nuclear Criticality Safety,  
September 17-21, 1995, Albuquerque
- [4] "The criticality aspects of transportation of fissile materials",  
by E.R. WOODCOCK and H.C. PAXTON  
Technology Engineering and Safety – volume 4, p401– Pergamon Press 1961
- [5] Interim report - "Evaluation of nuclear criticality safety data and limits for actinides in transport"  
C4/TMR2001/200-1 - C. DEAN, D. MENNERDAHL, C. LAVARENNE
- [6] "Determination of the number of allowable packages in array for the transport of actinides  
U235, U233, Pu239, Pu241, Am242m, Np237, Cm244 and Pu238 in normal and accidental  
conditions."  
Note SEC/T/03.144 5th May 2003 - F. JEAN
- [7] IAEA SAFETY STANDARDS SERIES No. ST-1  
Regulations for the Safe Transport of Radioactive Material – 1996 Edition  
REQUIREMENTS
- [8] "Identification of the production chains of fissile and fissionable actinides to consider the  
criticality risk involved by radioactive decays of high nucleon number nuclides"  
Note SEC/T/03.143 - 5th May 2003 - F. JEAN
- [9] IAEA SAFETY STANDARDS SERIES No. TS-G-1.1 (ST-2)  
Advisory Material for the IAEA Regulations for the Safe Transport of Radioactive Material
- [10] « Valeurs minimales critiques sphériques des actinides métalliques et en solution, nues ou  
réfléchies. Etude complémentaire et comparative »  
Note SEC/T/02.322 du 18 octobre 2002 – M. DULUC et J ANNO



## APPENDIX A

Table 6. Capability to sustain a chain reaction

Actinide	Slow (S) / Fast (F)	Library	Comments
<sup>227</sup> Ac	Neither see comments	JENDL-3.2=3.3	$\eta > 1.0$ above the fission threshold but inelastic scattering is at least 10 times more likely than fission. It will scatter the neutrons below the fission threshold.
<sup>228</sup> Th	F	JENDL-3.2=3.3	JANIS graphs show this is highly likely but a K-infinity calculation is required to confirm this.
<sup>229</sup> Th	F but see comment	JENDL-3.2=3.3	Classified fissible at high energy but there is a significant region between 1 and 10eV where $\eta > 1.0$ .
<sup>230</sup> Th	Neither see comments	JEF-2.2	$\eta > 1.0$ above the fission threshold but inelastic scattering is much more likely than fission. It will scatter the neutrons below the fission threshold.
<sup>232</sup> Th	Neither see comments	JENDL-3.3 but 3.2 is adequate	$\eta > 1.0$ above the fission threshold but inelastic scattering is much more likely than fission. It will scatter the neutrons below the fission threshold
<sup>231</sup> Pa	F	JENDL3.2	JANIS $\eta$ graphs of JEF-2.2 =ENDF/B-V =ENDF/B-VI are very similar to JENDL-3.3=JENDL3.2. Yet Ganesan, in Physor2000, is quoting K-infinite=0.9727 (JENDL-3.2), and 0.9410 ENDF/B-VI.5. This difference is probably due to incorporation of anisotropic angular data into JENDL-3.2 from model codes.(ENDF/B-V assumes isotropic distributions)
<sup>232</sup> U	F + S	ENDF/B-VI r8	The high NUBAR retained in ENDF/B-VI r8 is attributed to Howerton's review. However this only lists an experimental value by Jaffey and Lerner. Howerton contacted the experimentalists noting the discrepancy with his theory. This discrepancy had also been noted by the experimentalists who wanted to repeat the measurement but the experiment was terminated. R Q Wright should be contacted about this. In the mean time we must be pessimistic and take the evaluation giving the lowest critical mass. ENDF/B-VI r8 is suggested because it retains the high experimental nu plus the anisotropic inelastic scatter.
<sup>233</sup> U	S + F	JEF-2.2, JENDL-3.2 ENDF/B-V and beyond.	
<sup>234</sup> U	F	JENDL-3.3	The new JENDL file uses model codes to represent anisotropy in inelastic levels. For Pa231 this seemed to increase K-infinity significantly.
<sup>235</sup> U	S + F	JEF-2.2, JENDL-3.2 ENDF/B-VI	Results will alter with versions of ENDF/B-VI. For soft spectra JEF benchmarking indicates the effect is not large.
<sup>236</sup> U	F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>238</sup> U	Neither see comments.	JEF-2.2, JENDL-3.2 ENDF/B-VI	$\eta > 1.0$ above the fission threshold but inelastic scattering will scatter the neutrons below the fission threshold.
<sup>235</sup> Np	F	JENDL-3.3	New evaluation for JENDL-3.3. ENDL-84 also has data.
<sup>236</sup> Np	S + F	ENDF/B-VI r8	JENDL-3.3 is a conversion of JENDL-3.2. ENDF/B-VI r8 retains an experimental nubar of 3.12 compared with JENDL-3.3's more likely value of 2.4 from systematics. However for this work it is important to use the higher value. As nubar is constant at energies important to solutions one could scale JENDL-3.2 results.
<sup>237</sup> Np	F	JEF-2.2, JENDL-3.2 ENDF/B-VI	

Actinide	Slow (S) / Fast (F)	Library	Comments
<sup>236</sup> Pu	S + F	JENDL-3.3 or ENDF/B-VI7	ENDF/B-VI7 –1995 and JENDL3.3 use the same resonance data. $\eta$ is 2.37 compared with 1.43 in earlier files. (including earlier release of ENDF/B-VI!) This is a significant data difference (noted in JEF/DOC-657) and needs review because JEFF3.0 has retained the earlier file.
<sup>237</sup> Pu	S + F	JENDL-3.3	New evaluation for JENDL-3.3 gives thermal $\eta$ 2.375 compared with 2.25 from earlier data.
<sup>238</sup> Pu	Possible solution!	JEF-2.2, JENDL-3.2 ENDF/B-VI	$\eta > 1$ above 175eV Important to know how Pu238 energy sources are transported-could they be cut into fine particles in a solution? Metallic critical mass is molten (for this mass the specific heat is very important)!
<sup>239</sup> Pu	S + F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>240</sup> Pu	F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>241</sup> Pu	S + F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>242</sup> Pu	F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>244</sup> Pu	F	JEF-2.2 JENDL-3.3	JEFF retained JEF-2.2 in JEFF-3.0. $\eta$ is higher than in JENDL-3.3 but the new JENDL file uses model codes to represent anisotropy in inelastic levels. This may result in higher K values.
<sup>241</sup> Am	F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>242m</sup> Am	S + F	JENDL-3.3	Thermal $\eta$ significantly higher than JEF-2.2. JENDL-3.3 significantly different from JENDL-3.2.
<sup>243</sup> Am	F	JEF-2.2, JENDL-3.2 ENDF/B-VI	
<sup>242</sup> Cm	S ? + F	JENDL-3.2 or 3.3	The fission cross section is larger in the JENDL file and has resonance structure not present in the other file. This gives the possibility of a fissile solution.
<sup>243</sup> Cm	S + F	ENDF/B-VI post 1995 or JENDL-3.3	Data are very similar – MAZLOV adopted.
<sup>244</sup> Cm	F	ENDF/B-VI or JENDL-3.3	New JENDL-3.3 evaluation has updated inelastic angular data.
<sup>245</sup> Cm	S + F	ENDF/B-VI7 or JENDL-3.3	Both ENDF and JENDL have adopted MASLOV data
<sup>246</sup> Cm	S ? + F	ENDF/B-VI7	$\eta$ is above 1.0 for the cross section between resonances from 6.5 –400eV in ENDF/B-VI7 only. JENDL-3.3 and ENDF/B-VI7 seem to have the same inelastic scatter angular representation.
<sup>247</sup> Cm	S + F	JENDL-3.3	$\eta$ larger in JENDL-3.3. Fission increased significantly from JENDL-3.2
<sup>248</sup> Cm	F	JEF-2.2 ENDF/B-VI, JENDL-3.2 or JENDL-3.3	JEF-2.2 and ENDF/B-VI are the same; nubar is higher than JENDL-3.2. JENDL-3.2 has anisotropic inelastic secondary data which are improved in JENDL-3.3
<sup>250</sup> Cm	F	JENDL-3.3	Only in JENDL and 3.3 modified (JENDL-3.2 adopted for JEFF3.0)
<sup>247</sup> Bk	S + F	JENDL-3.3	Only in JENDL-3.3 Completely theoretical.

Actinide	Slow (S) / Fast (F)	Library	Comments
<sup>248</sup> Bk	?		NO DATA in EXFOR or CINDA
<sup>249</sup> Bk	F	ENDF/B-VI or JENDL-3.3	JEF adopted ENDF/B-VI conversion of late addition to ENDF/B-V. The files contain different anisotropic inelastic angular distributions.
<sup>248</sup> Cf	?		NO relevant DATA in EXFOR or CINDA Yet a K-infinity reported from Russia in TRANSSACIII
<sup>249</sup> Cf	S + F	ENDF/B-VI or JENDL-3.3	ENDF/B-VI is a conversion of a late addition to ENDF/B-V $\eta$ is similar.
<sup>250</sup> Cf	F	ENDF/B-VI or JENDL-3.3	ENDF/B-VI adopted late ENDF/B-V evaluation as did JEF-2.2 = JEFF3.0. $\eta$ is above 1.0 at lower energies in ENDF/B-VI (above ~37KeV) compared with 321KeV in JENDL-3.3 but angular distribution shapes may make significant effects.
<sup>251</sup> Cf	S + F	ENDF/B-VI	ENDF/B-VI and JEF-2.2=JEFF3.0 adopted ENDF/B-V with $\eta$ above JENDL-3.3=JENDL-3.2.
<sup>252</sup> Cf	S + F	ENDF/B-VI	The ENDF/B-VI = ENDF/B-V =JEF-2.2=JEFF3 evaluation yields a higher $\eta$ up to 52.1KeV. ie at energies important to solutions.
<sup>254</sup> Cf	S + F	JENDL-3.2	JENDL-3.3 and JEFF-3.0 have adopted JENDL-3.2. The file is VERY crude being 1/V fits to Mughabghab's thermal data joined to a model calculation above 120eV.
<sup>252</sup> Es	?		NO DATA in EXFOR. CINDA contains fission data 2-4MeV only.
<sup>254</sup> Es	S + F	JENDL-3.2	JENDL-3.3 and JEFF-3.0 have adopted JENDL-3.2. The file is VERY crude being 1/V fits to Mughabghab's thermal data, then a shape adjustment to fit his resonance integral. The curve is joined to a model calculation above 5eV.
<sup>257</sup> Fm	S + F	No evaluation	There is some data in EXFOR that indicate probable critical mass in a solution. A crude evaluation could be constructed in a similar manner to Es-254.
<sup>258</sup> Md	?		No relevant DATA in EXFOR or CINDA.



Some calculations have been performed at SERCO showing some differences for the k infinite when using the same library but a different code. The results are given below:

Actinide	MCNP (EMS)		MONK DICE (SERCO)		MONK BINGO (SERCO)
	ENDF/B-Vlr5	JEF2.2	ENDF/B-Vlr4	JEF2.2	JEF2.2
<sup>230</sup> Th	0.1497	1	0.1456	1	1
<sup>232</sup> Th	0.0798	0.0681	0.0736	0.0643	0.0682
<sup>236</sup> U	0.7353	0.6538	0.6736	0.5928	0.6544
<sup>238</sup> U	0.3445	0.3438	0.3389	0.3377	0.3433
<sup>237</sup> Np	1.705	1.6308	1.6768	1.6064	1.6355
<sup>238</sup> Pu	2.7612	2.8682	2.7457	2.8526	2.869
<sup>240</sup> Pu	2.2653	2.1188	2.2187	2.097	2.1226
<sup>242</sup> Pu	1.8669	1.9194	1.8316	1.894	1.9229
<sup>241</sup> Am	1.998	1.9628	1.806	1.9457	1.9705
<sup>243</sup> Am	1.6878	1.5917	1.4925	1.5723	1.5997
<sup>244</sup> Cm	2.6412	2.5117	2.6158	2.4699	2.511

### Appendix C - available evaluations for the actinide nuclides

Actinide	JEFF-3.0 Source	JEF/DOC-657	JENDL3.3	ENDF/B-VI rto8
<sup>227</sup> Ac	Not in JEFF-3.0	Not reviewed	JENDL-3.2	Not in ENDF/B-VI
<sup>228</sup> Th	JENDL-3.2	JENDL-3.2	JENDL-3.2	Not in ENDF/B-VI
<sup>229</sup> Th	JENDL-3.2 Minor change to NUBAR representation.	JENDL-3.2	JENDL-3.2	Not in ENDF/B-VI
<sup>230</sup> Th	JEF-2.2	JEF-2.2	JENDL-3.2	ENDF/B-V-mat 9034
<sup>232</sup> Th	JENDL-3.2	JENDL-3.2	New Evaluation	ENDF/B-V –mat 9040
<sup>231</sup> Pa	JEF-2.2 + ENDFBVIr4 NUBAR	JENDL-3.2+delayed neutron data from ENDF/B-VI.	JENDL-3.2	ENDF/B-V-mat 9131
<sup>232</sup> U	JEF-2.2 with ENDF/BVIr4 NUBAR	JEF-2.2 delayed neutron data from ENDF/BVI.	JENDL-3.2	New evaluation r8 mat 9219 Mughabghab resonance, JENDL3.2 high E, New fission, ENDF/B-V NU preserved
<sup>233</sup> U	JENDL-3.2 with ENDFBVIr4 NUBAR	JENDL-3.2 with ENDF/B-VI delayed neutron spectrum.	New Evaluation	ENDF/B-V-mat 9222
<sup>234</sup> U	JEF-2.2	JENDL-3.2	New Evaluation	ENDF/B-V-mat 9225
<sup>235</sup> U	ENDF/BVIr 5	ENDF/B-VI release 4 or later plus updated unresolved resonance data.	New Evaluation but same resonance parameters as ENDF/B-VI r5.	New Evaluation r5 mat 9228
<sup>236</sup> U	JENDL-3.2	JENDL-3.2	New Evaluation	New Evaluation in 1989 mat 9231
<sup>238</sup> U	JEF-2.2 with revised inelastic, from JENDL-3.2. (n,xn) from ENDF/B-VI r7. Delayed neutron fraction and delayed fission spectrum from JEF/DOC-920. NUBAR was recalculated.	JEF-2.2 with revised inelastic, (n,3n) and fission spectrum from JENDL-3.2 or as advised by WPEC sub-group, possibly incorporating new IRMM Geel, Lowell and Obninsk measurements of inelastic	New Evaluation	ENDF/BVIr3 in 1997 mat 9237
<sup>235</sup> Np	Not in JEFF-3.0	ENDL-84	New Evaluation	Not in ENDF/B-VI
<sup>236</sup> Np	JENDL-3.2	JENDL-3.2	JENDL-3.2	New Evaluation R8 mat 9343. New resonance parameters NUBAR 3.12 from Mughabghab.

Actinide	JEFF-3.0 Source	JEF/DOC-657	JENDL3.3	ENDF/B-VI rto8
<sup>237</sup> Np	ENDF/B-VI r4, (n,xn) data from JEF-2.2.	ENDF/B-VI with (n,2n) replaced by JEF-2.2 and unresolved parameters added from JENDL-3.2 but await results from integral studies before a final choice is made.	New Evaluation	ENDF/B-VI r1 –mat 9346
<sup>236</sup> Pu	JEF-2.2	JEF-2.2	New Evaluation	New evaluation Sept 1995 mat 9428
<sup>237</sup> Pu	JEF-2.2	JEF-2.2	New Evaluation	ENDF/B-V mat 9431
<sup>238</sup> Pu	JENDL-3.2 with unresolved resonance parameters from BROND-2.2 and fission energy from ENDFB-VI.	JENDL-3.2 with continuum energy distribution and unresolved parameters but not infinite dilution cross section from BROND2; but await results from integral studies before a final choice is made.	New Evaluation but much the same as JEFF3 comments.	ENDF/B-V mat 9434
<sup>239</sup> Pu	New Evaluation	New Evaluation	New Evaluation	ENDF/B-VI r2 –1997 mat 9437
<sup>240</sup> Pu	New Evaluation	New Evaluation	New Evaluation comments similar to JEFF3.0	ENDF/B-VI r2 mat 9440
<sup>241</sup> Pu	New Evaluation	New Evaluation	New Evaluation	ENDF/B-VI r2 mat 9443
<sup>242</sup> Pu	New Evaluation	New Evaluation	New Evaluation	ENDF/B-V mat 9446
<sup>244</sup> Pu	JEF-2.2	JEF-2.2	New Evaluation	ENDF/B-V mat 9452
<sup>241</sup> Am	JEF-2.2	New unresolved data from Frohner otherwise a strong hint to use ENDF/B-VI subject to effects from integral trends	New Evaluation	ENDF/B-VI r2 Aug 1994 mat 9543
<sup>242m</sup> Am	JEF-2.2 + ENDFBVI NUBAR	JEF-2.2 with delayed neutron data from ENDF/B-VI	New Evaluation	ENDF/B-V mat 9547
<sup>243</sup> Am	JEF-2.2.	Review ENDF/B-VI release 5 and integral results. ENDF/B-VI r5, JENDL-3.2 and BROND all improve on JEF-2.2.	New Evaluation	ENDF/B-VI r1 mat 9549
<sup>242</sup> Cm	JENDL-3.2 data were adopted below 40 keV (limit of the unresolved resonance region) and JEF-2.2 adopted at higher energies.	ENEA to review; noting photon production and fission energy data in ENDF/B-VI.	New Evaluation	ENDF/B-V mat 9631

Actinide	JEFF-3.0 Source	JEF/DOC-657	JENDL3.3	ENDF/B-VI rto8
<sup>243</sup> Cm	JENDL-3.2 data were adopted below 40 keV (limit of the unresolved resonance region) and JEF-2.2 adopted at higher energies.	ENEA to review; noting delayed neutron data in JENDL-3.2 or BROND.	New Evaluation	ENDF/B-VI July 1995 MAZLOV Mat 9634
<sup>244</sup> Cm	JEF-2.2	ENEA to review	New Evaluation	ENDF/B-V mat 9637
<sup>245</sup> Cm	JENDL-3.2.	ENEA to review	New Evaluation - mainly Maslov	ENDF/B-VI says r2 but distributed in 2000! Mat 9640.
<sup>246</sup> Cm	MASLOV – MINSK (DIST-FEB96)	BROND/MASLOV	New Evaluation – mainly Maslov	ENDF/B-VI says r2 but distributed in 2000! Mat 9643.
<sup>247</sup> Cm	JENDL-3.2	JENDL-3.2	New Evaluation	ENDF/B-V mat 9646
<sup>248</sup> Cm	JENDL-3.2	JENDL-3.2.	New evaluation but JENDL2 resolved parameters	ENDF/B-V mat 9649
<sup>250</sup> Cm	JENDL-3.2	JENDL-3.2	New Evaluation	Not in ENDF/B-VI
<sup>247</sup> Bk	Not in JEFF-3.0	Not reviewed	New Evaluation	Not in ENDF/B-VI
<sup>248</sup> Bk	Not in JEFF-3.0	Not reviewed	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>249</sup> Bk	ENDF/B-VI r4	Adopt ENDF/B-VI/CENDL2	JENDL-3.2	ENDF/B-VI r1 Possibly very late addition to ENDF/BV then converted. mat 9752
<sup>248</sup> Cf	Not in JEFF-3.0		Not in JENDL-3.3	Not in ENDF/B-VI
<sup>249</sup> Cf	ENDF/B-VI r4	Adopt ENDF/BVI = CENDL data.	JENDL-3.3 with new evaluation of angular and energy data.	ENDF/B-VI r1 Possibly very late addition to ENDF/BV then converted. mat 9852
<sup>250</sup> Cf	JEF-2.2	JEF-2.2	JENDL-3.2	ENDF/B-V mat 9855
<sup>251</sup> Cf	JEF-2.2 with ENDFBVI r4 NUBAR.	JEF-2.2 with delayed neutron data from ENDF/B-VI.	JENDL-3.2	ENDF/B-V mat 9858
<sup>252</sup> Cf	JEF-2.2.	JEF-2.2 with delayed neutron data from ENDF/B-VI.	JENDL-3.2	ENDF/B-V mat 9861
<sup>254</sup> Cf	JENDL-3.2	JENDL-3.2	JENDL-3.2	Not in ENDF/B-VI
<sup>252</sup> Es	Not in JEFF-3.0	Not reviewed	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>254</sup> Es	JENDL-3.2	JENDL-3.2	JENDL-3.2	Not in ENDF/B-VI
<sup>257</sup> Fm	Not in JEFF-3.0	Not reviewed	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>258</sup> Md	Not in JEFF-3.0	Not reviewed	Not in JENDL-3.3	Not in ENDF/B-VI



## Appendix D - Graphs of ETA ( $\eta$ )

### $^{232}\text{U}/^{236}\text{Pu}$

Reference [5] (appendix A) indicates a smaller critical mass in metal than in solution for  $^{232}\text{U}$  and  $^{236}\text{Pu}$ . **Graph 1** below shows ETA for both nuclides plus  $^{233}\text{U}$  where the critical mass in solution is much smaller than in metal. **Graph 2** covers a smaller energy range and significant dips in eta for  $^{232}\text{U}$  and  $^{236}\text{Pu}$  can be seen between 1 and 10 eV whereas the dip in  $^{233}\text{U}$  is much narrower. This may explain why a smaller mass is achieved in metal.

However, for  $^{236}\text{Pu}$  it appears that, with both ENDF/B-VI.8 and JENDL-3.3 cross sections, the moderated minimum critical mass is smaller than the metal case. The figures 1 and 2 clearly show that this is credible.

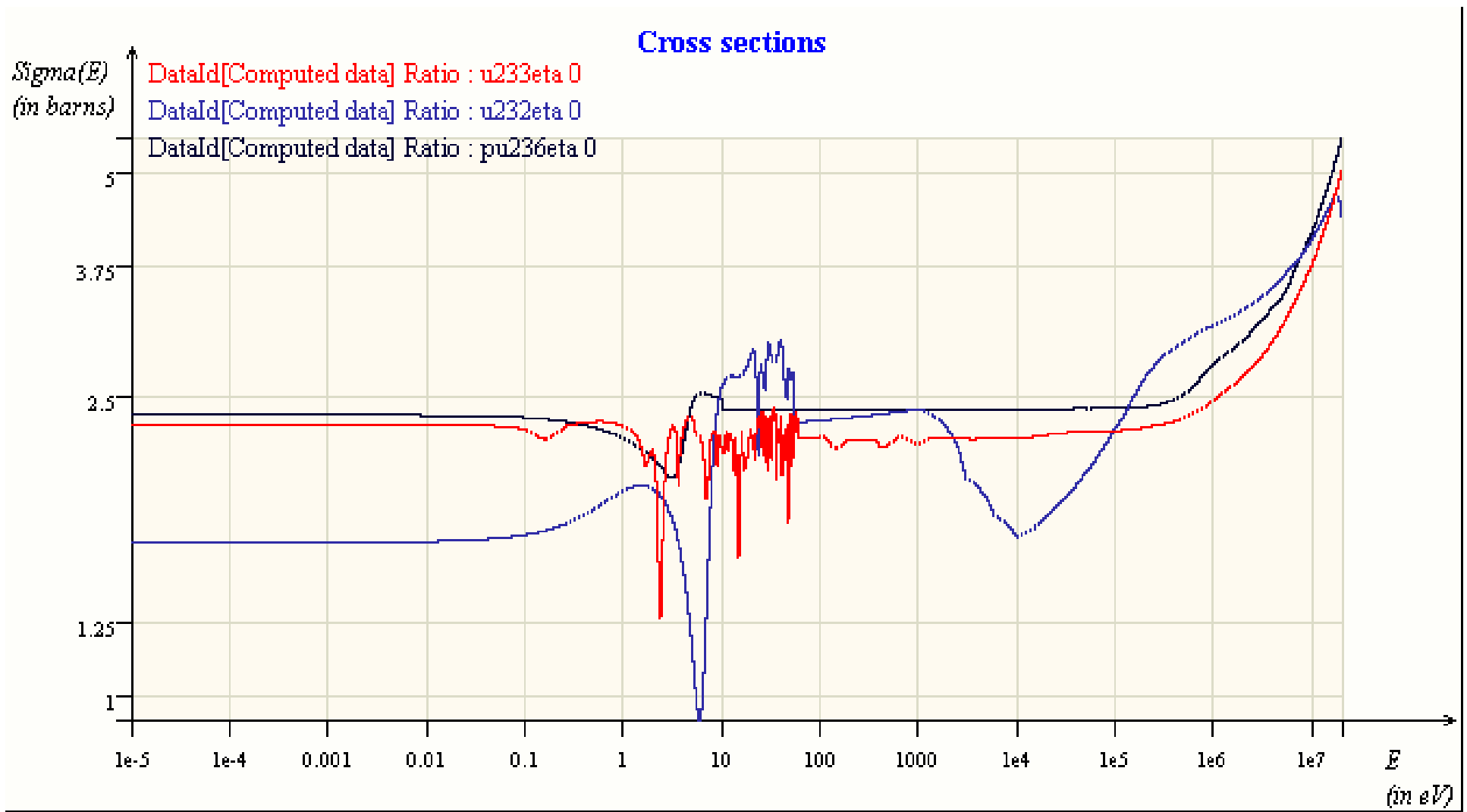
### $^{238}\text{Pu}$

**Table 3** indicates the possibility of criticality in solution yet all results in reference [5] are fast systems. **Appendix A** notes ETA is more than 1 above 175 eV. This is in JEF2.2 and JENDL3.3 but not in ENDF/B-VI.7. **Graphs 3** and **4** show this.

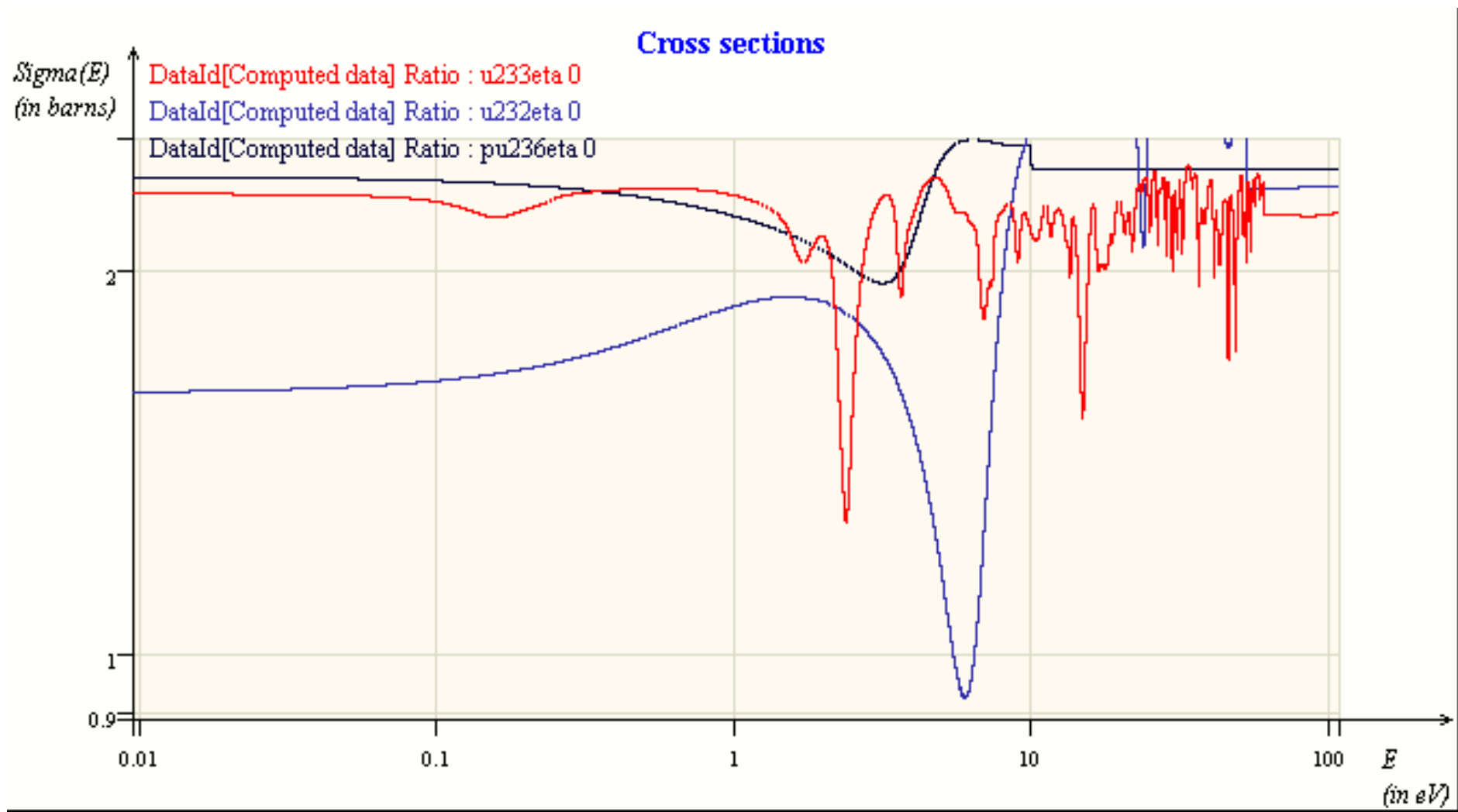
### $^{242}\text{CM}, ^{246}\text{CM}$ AND $^{254}\text{CF}$

**Table 3** again indicates the possibility of criticality in solution yet all results in reference [5] are fast systems. **Appendix A** notes ETA is more than 1 in JENDL3.3 at various energies (all for  $^{254}\text{Cf}$ ). **Graphs 5, 6** and **7** show this.

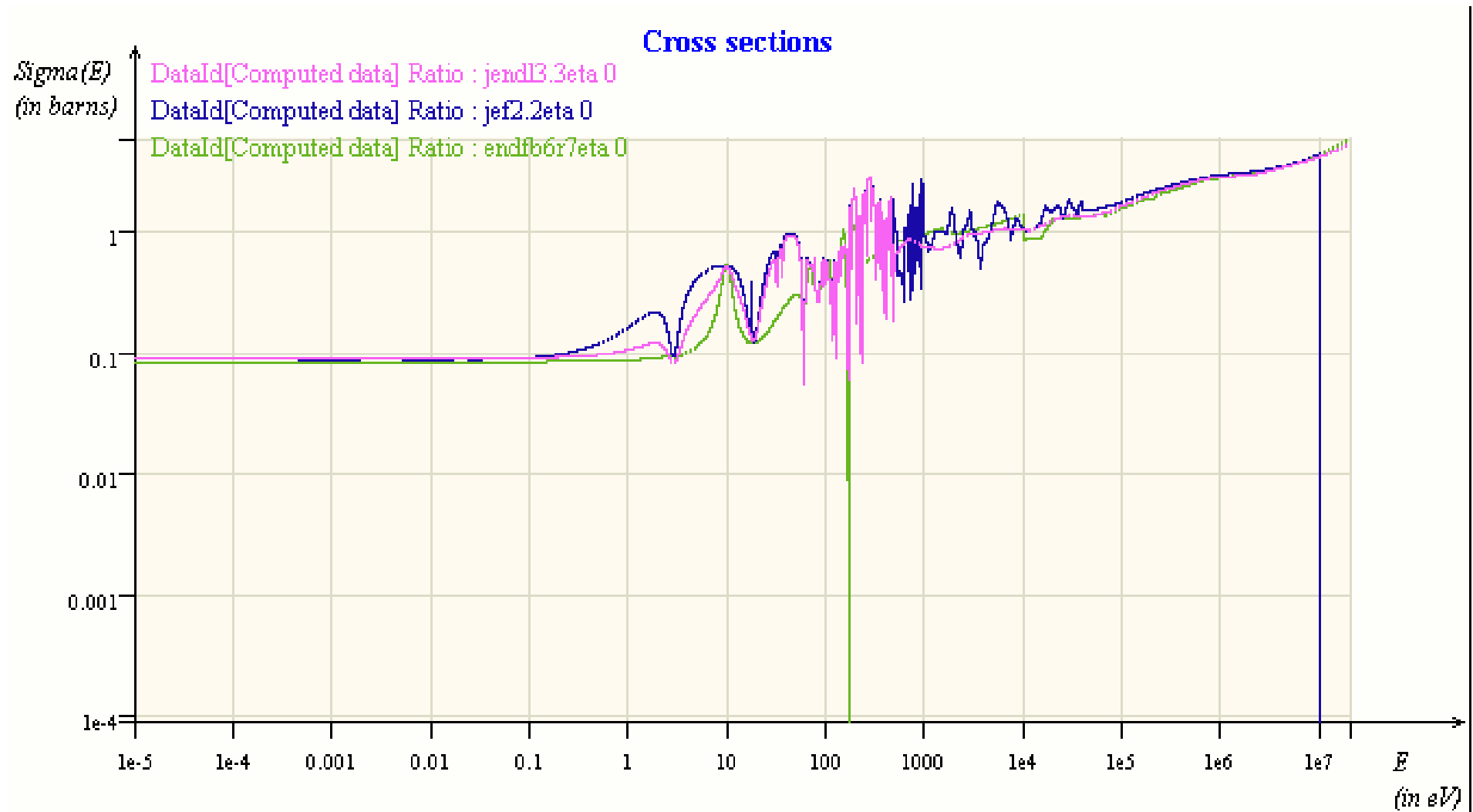
Graph 1 - ETA in  $^{232}\text{U}$ ,  $^{233}\text{U}$  and  $^{236}\text{Pu}$



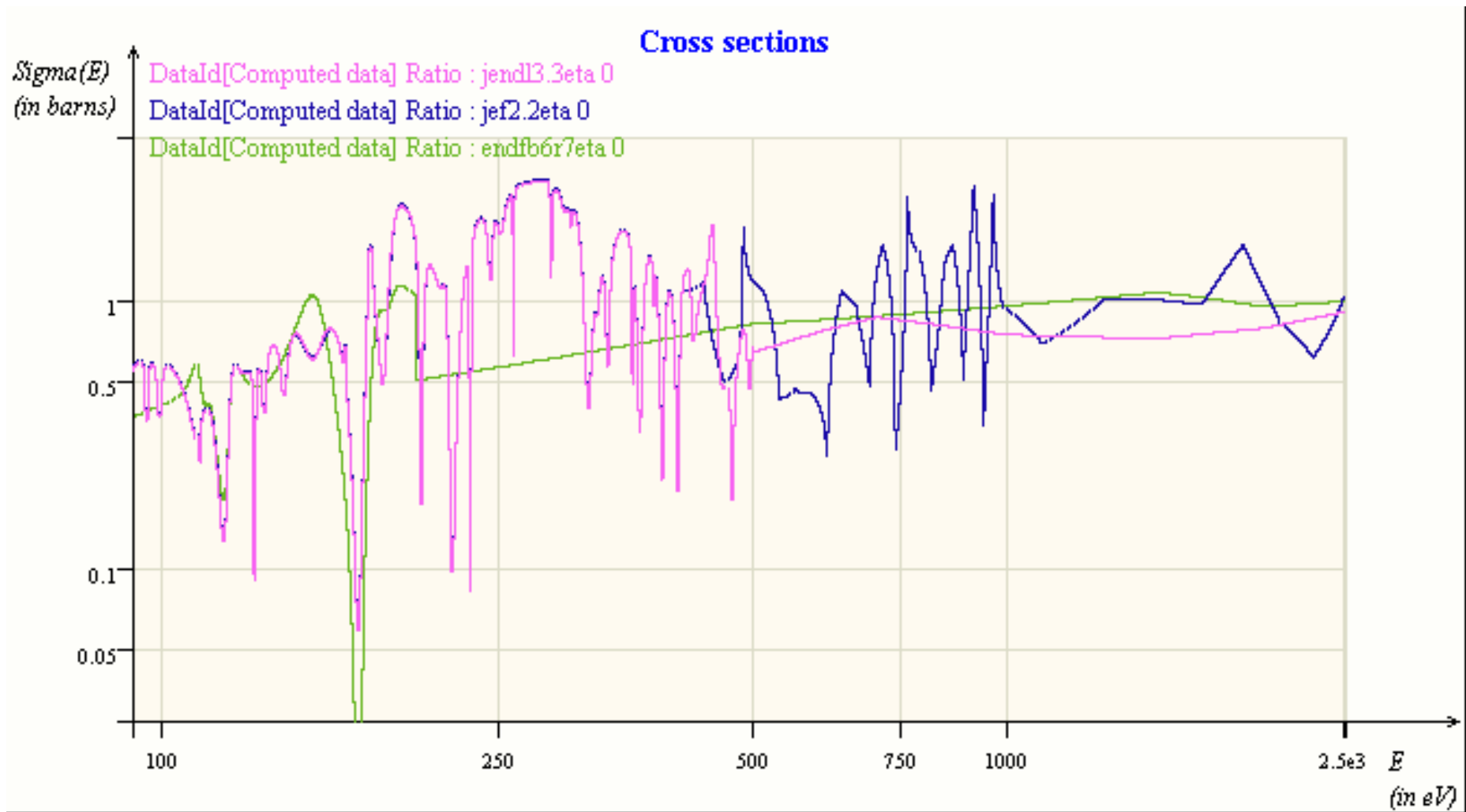
Graph 2 - ETA in  $^{232}\text{U}$ ,  $^{233}\text{U}$  and  $^{236}\text{Pu}$  (Zoom)



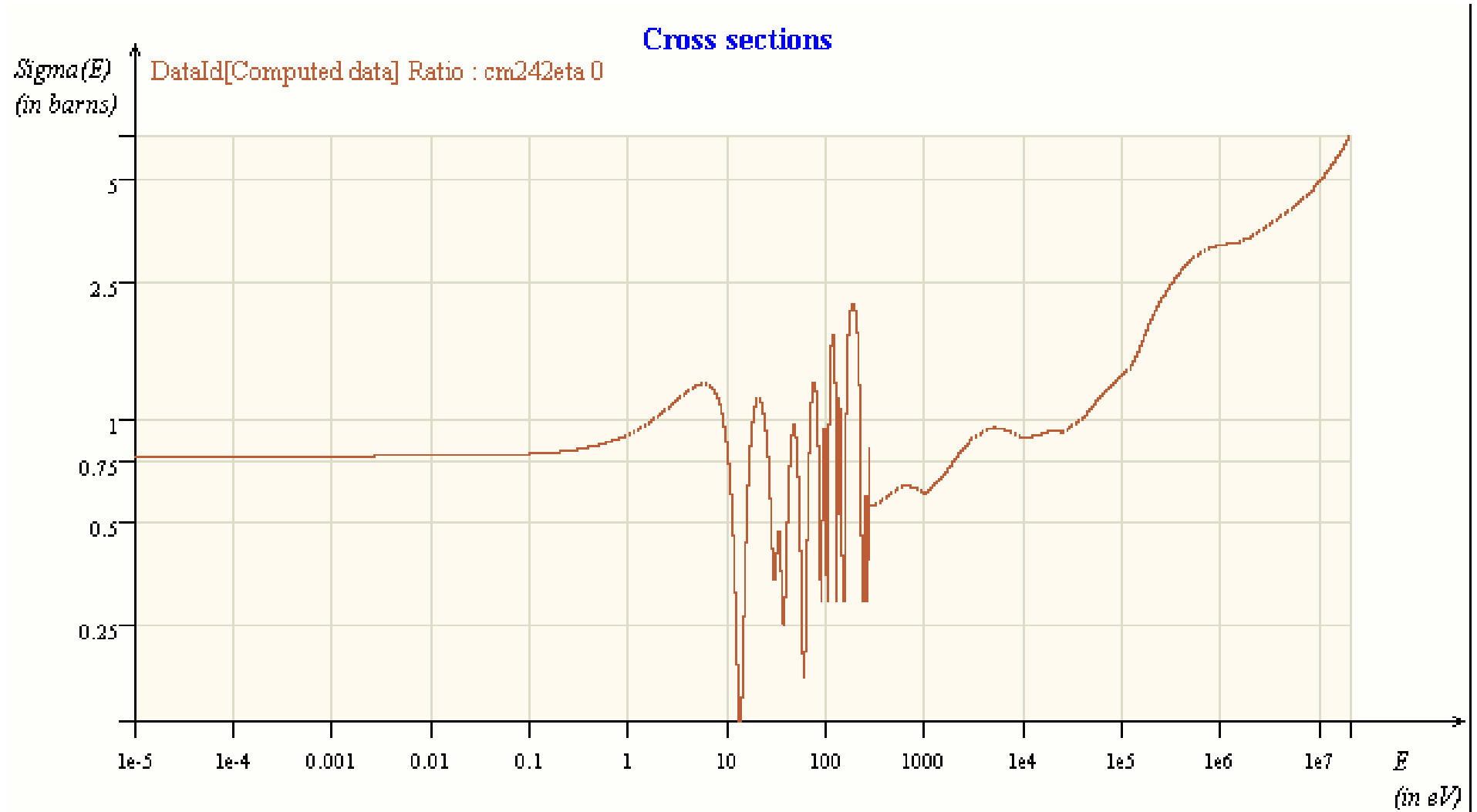
Graph 3 - <sup>238</sup>Pu Eta from Various Evaluations



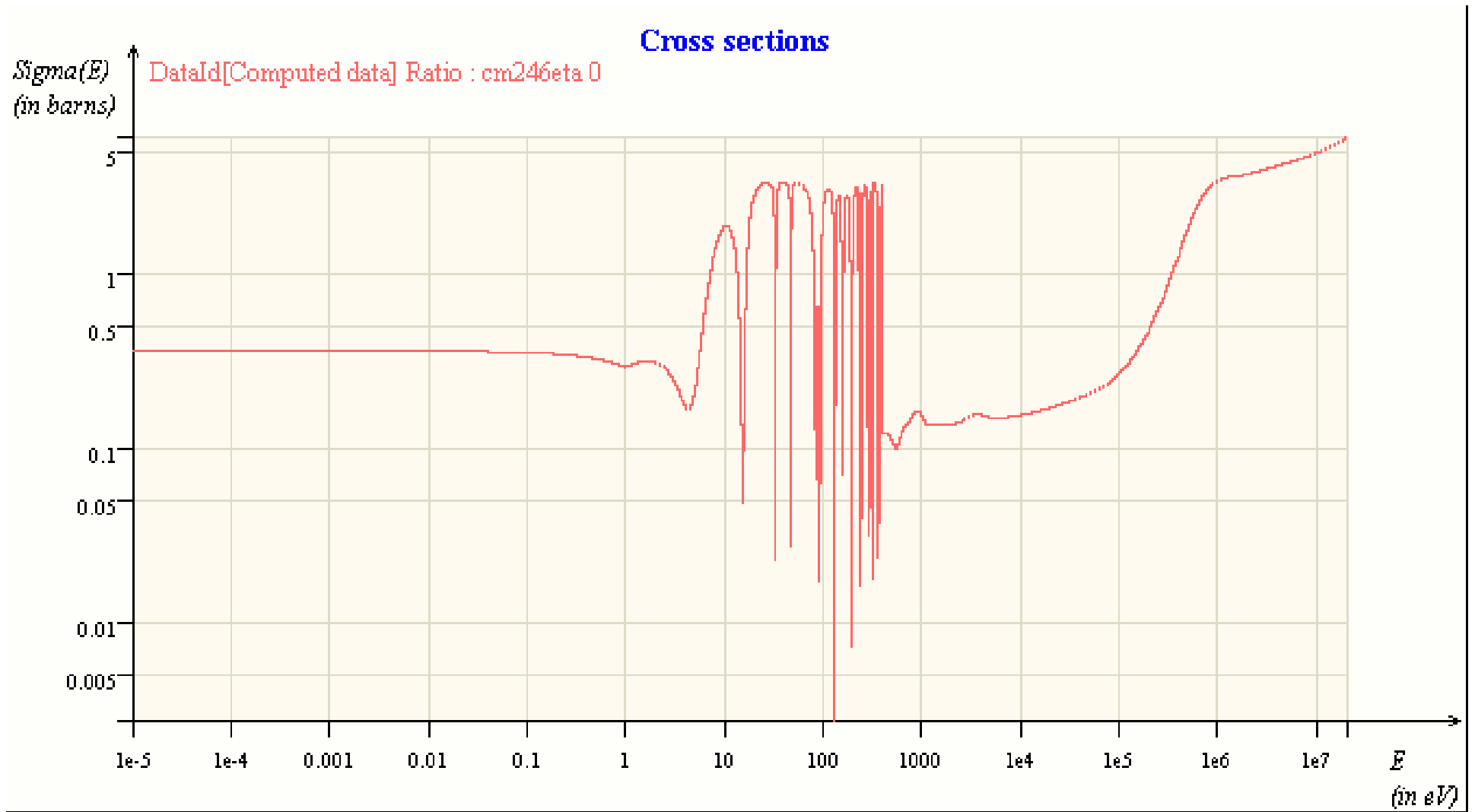
**Graph 4 -  $^{238}\text{Pu}$  Eta from Various Evaluations (Zoom)**



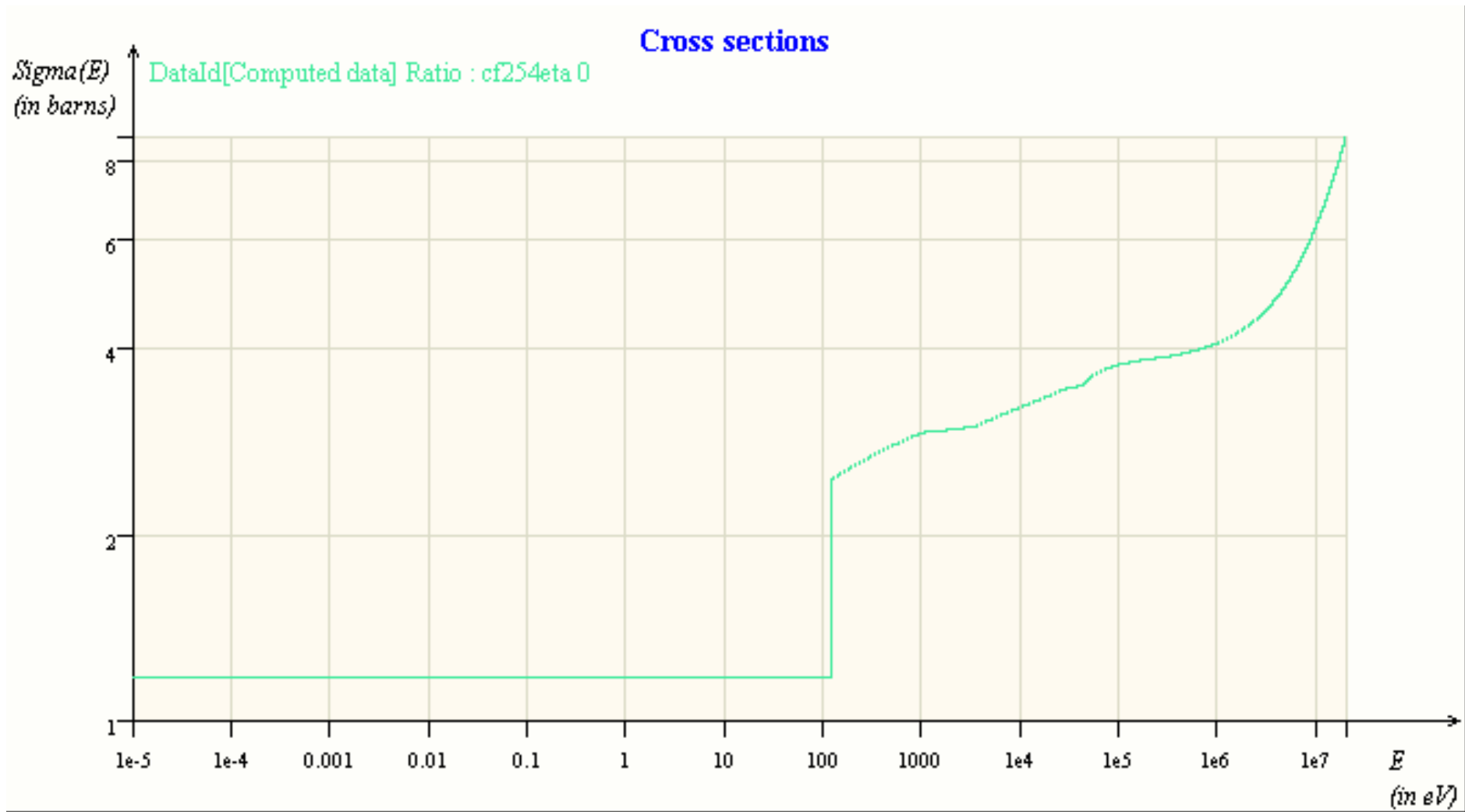
Graph 5 - Eta in <sup>242</sup>Cm from JENDL3.3



**Graph 6** - Eta in <sup>246</sup>Cm from JENDL3.3



Graph 7 - Eta in <sup>254</sup>Cf from JENDL3.3





**APPENDIX E - Thermal NUBAR for the new evaluations**

<b>Actinide</b>	<b>JEFF-3.0 Source</b>	<b>JENDL3.3</b>	<b>ENDF/B-Virto8</b>
<sup>227</sup> Ac	Not in JEFF-3.0	1.8321	Not in ENDF/B-VI
<sup>228</sup> Th	2.021	2.021	Not in ENDF/B-VI
<sup>229</sup> Th	2.0872	2.0872	Not in ENDF/B-VI
<sup>230</sup> Th	2.01	2.074	2.01
<sup>232</sup> Th	0.0 (threshold 400KeV)	0.0 (threshold 400KeV)	1.9487
<sup>231</sup> Pa	2.29	2.1816	2.29
<sup>232</sup> U	3.13	2.456	3.13
<sup>233</sup> U	2.493	2.48771	2.4947
<sup>234</sup> U	2.352	2.36447	2.352
<sup>235</sup> U	2.4367	2.43663	2.4367
<sup>236</sup> U	2.3829	2.3829	2.317
<sup>238</sup> U	2.489	2.48754	2.492088
<sup>235</sup> Np	Not in JEFF-3.0	2.66385	Not in ENDF/B-VI
<sup>236</sup> Np	2.40705	2.40705	3.12
<sup>237</sup> Np	2.63581	2.6014	2.63581
<sup>236</sup> Pu	2.86	2.814	2.814
<sup>237</sup> Pu	2.824	2.863	2.824
<sup>238</sup> Pu	2.89471	2.8447	2.895
<sup>239</sup> Pu	2.87777 but varying	2.88432 but varying	2.8789 but varying
<sup>240</sup> Pu	2.78391	2.78391	2.803
<sup>241</sup> Pu	2.9313	2.9313	2.9453
<sup>242</sup> Pu	2.8595	2.8594	2.81
<sup>244</sup> Pu	2.884	2.82	2.884
<sup>241</sup> Am	3.33	3.0599	3.238788
<sup>242m</sup> Am	3.264	3.2705	3.264
<sup>243</sup> Am	3.0616	3.2085	3.272833
<sup>242</sup> Cm	3.44	3.2509	3.44
<sup>243</sup> Cm	3.43301	3.43201	3.43201
<sup>244</sup> Cm	3.24	3.24435	3.46
<sup>245</sup> Cm	3.5313	3.5964	3.5964
<sup>246</sup> Cm	3.61416	3.61416	3.61416
<sup>247</sup> Cm	3.8034	3.8034	3.58
<sup>248</sup> Cm	3.1296	3.1296	3.49
<sup>250</sup> Cm	3.39994	3.3999	Not in ENDF/B-VI
<sup>247</sup> Bk	Not in JEFF-3.0	3.5067	Not in ENDF/B-VI
<sup>248</sup> Bk	Not in JEFF-3.0	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>249</sup> Bk	3.3464	3.4189	3.3464
<sup>248</sup> Cf	Not in JEFF-3.0	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>249</sup> Cf	3.8869	4.0628	3.8869
<sup>250</sup> Cf	3.63	3.63406	3.63
<sup>251</sup> Cf	4.14	4.1059	4.14
<sup>252</sup> Cf	4.06	3.8923	4.06
<sup>254</sup> Cf	3.8508	3.8508	Not in ENDF/B-VI
<sup>252</sup> Es	Not in JEFF-3.0	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>254</sup> Es	4.0832	4.0832	Not in ENDF/B-VI
<sup>257</sup> Fm	Not in JEFF-3.0	Not in JENDL-3.3	Not in ENDF/B-VI
<sup>258</sup> Md	Not in JEFF-3.0	Not in JENDL-3.3	Not in ENDF/B-VI

**Appendix F - Recapitulative table on the transport of actinide nuclides in normal and accidental conditions**

Reflector	Actinides	Critical mass (k = 1) g	Allowable mass (k = 0.95) g	Mass per package g	Steel thickness mm	Void thickness mm	5N1	N1	N2 (allowable)	N'2 (critical)	N
WATER (20 cm)	U 235	784.6	620.3	15	-	-	165	33	20	26	20
				10	-	-	533	106	31	39	31
				5	-	-	4327	865	62	78	62
	U 233	559.4	454	15	0	-	115	23	15	18	15
					2	-	9695	1939			
					5	-	infinite	infinite			
				10	0	-	380	76			
	Pu 239	503	394.3		2	-	604500	120900	22	27	22
					5	-	infinite	infinite			
				1	-	-	366400	73280	227	279	227
				15	0	-	41	8			
					2	-	247	49	13	16	8
					5	-	2685	537			
				10	0	-	120	24			
	Pu 241	268.9	216.9		2	-	5365	1073	19	25	19
					5	-	infinite	infinite			
				5	-	-	965	193	39	50	39
				1	-	-	119270	23854	197	251	197
	Am 242m	23	19.1		0	-	295	59	21	26	21
					5	-	infinite	infinite			
1				-	-	36640	7328	108	134	108	
STEEL (30 cm)	Np 237 (crystal form)	49961	37213	0.5	-	-	289000	57800	216	268	216
				0.1	-	-	48130	9626	95	115	95
	Cm 244 (crystal form)	16811	13336	1	0	-	50	10	9	11	9
					5	-	infinite	infinite			
				0.5	-	-	395	79	19	23	19
Pu 238 (crystal form)	4779	3930	0.1	-	-	48130	9626	95	115	95	
				0.5	-	-	48130	9626	95	115	95
STEEL (30 cm)	Np 237 (crystal form)	49961	37213	10000	-	10.6	9	1	1	2	1
				1000	31.76	-	infinite	infinite	18	24	18
	Cm 244 (crystal form)	16811	13336	10000	-	4.77	1	0	0	0	0
				1000	29	-	infinite	infinite	6	8	6
	Pu 238 (crystal form)	4779	3930	1000	31.5	-	66	13	1	2	1
				500	35.325	-	915	183	3	4	3
				-	35.325	512	102				

### Appendix G - Crosschecking calculations by SERCO with MONK

Case	Description	MONK9 - BINGO		CRISTAL SD ≤ 100pcm	(M-C)/C	Δk (pcm)
		k effective	SD (1σ)			
9f8001	15g of U235, array 5x6x6 (180 packages), 700g Water, No wrapper	0.9541	±0.0005	0.96033	-0.65%	623
9f8002	As 9f8001 but 600g Water	0.9449	±0.0005	0.95137	-0.68%	647
9f8003	As 9f8001 but 800g Water	0.9517	±0.0005	0.95662	-0.51%	492
9f8004	15g of U233, array 5x5x5 (125 packages), 700g Water, No wrapper	0.9589	±0.0005	0.95971	-0.08%	81
9f8005	As 9f8004 but 600g Water	0.9447	±0.0005	0.94622	-0.16%	152
9f8006	As 9f8004 but 800g Water	0.957	±0.0005	0.95631	0.07%	-69
9f8007	15g of Pu239, array 3x4x4 (48 packages), 1000g Water, No wrapper	0.9656	±0.0005	0.97062	-0.52%	502
9f8008	As 9f8007 but 900g Water	0.9522	±0.0005	0.95795	-0.60%	575
9f8009	As 9f8007 but 1100g Water	0.9719	±0.0005	0.95317	1.97%	-1873
9f8010	1g of Pu241, array 35x35x35 (42875 packages), 100g Water, No wrapper	0.9694	±0.0005	0.97299	-0.37%	359
9f8011	As 9f8010 but 50g Water	0.8387	±0.0005	0.84297	-0.51%	427
9f8012	As 9f8010 but 75g Water	0.9391	±0.0005	0.94065	-0.16%	155
9f8013	As 9f8010 but 150g Water	0.9377	±0.0005	0.94107	-0.36%	337
9f8014	1000g of Np237, array 20x20x20 (8000 packages), 0g Water, Air wrapper 31.76mm thick	0.9664	±0.0005	0.97078	-0.45%	438
9f8015	1000g of Pu238, array 4x4x4 (64 packages), 0g Water, Steel wrapper	0.9418	±0.0005	0.98534	-4.42%	4354
				<b>0.94099</b> <b>(TRIPOLI 4)</b>	<b>0.09%</b>	<b>-81</b>
9f8016	As 9f8015 but 3x3x3 (27 packages)	0.8298	±0.0005	0.86427	-3.99%	3447
				<b>0.83109</b> <b>(TRIPOLI 4)</b>	<b>-0.16%</b>	<b>129</b>
9f8017	As 9f8015 but 5x5x5 (125 packages)	1.039	±0.0005	1.0825	-4.02%	4350
				<b>1.04073</b> <b>(TRIPOLI 4)</b>	<b>-0.17%</b>	<b>173</b>
9f8018	As 9f8015 but actinide and steel smeared across 10x10x10cm cube	0.6442	±0.0005	undone	-	-
9f8019	As 9f8018 but 5x5x5 (125 packages)	0.7788	±0.0005	undone	-	-

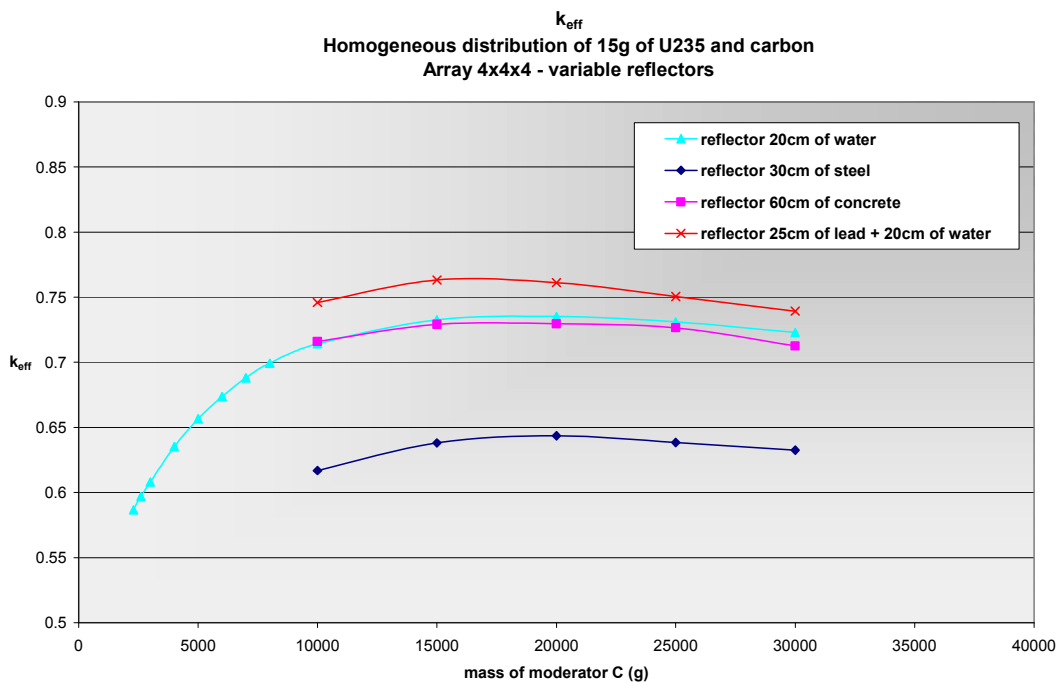
## Appendix H – Exception criteria and limits of mass

		Critical mass (g) (k=1)				Allowable mass (k=0.95)		Factors to consider on Mc				Safe Mc (g)	Mass per group of package (g)	Mass per package (g)	From ANS safe mass per package	
Nuclide	Spectrum	20 cm water		30 cm steel/lead		Min	20 cm water (IRSN)	30 cm steel (IRSN)	heterogeneity reflectors moderator F1	Margin of 5% in Δk F2	Libraries F3					ANS 8.15
		EMS	IRSN	EMS	IRSN											
<sup>227</sup> Ac	NF															
<sup>228</sup> Th	NF?															
<sup>229</sup> Th	F	2262	x	994	x	<b>994</b>	x	x								
<sup>230</sup> Th	NF															
<sup>232</sup> Th	NF															
<sup>231</sup> Pa	NF?															
<sup>232</sup> U	F	2130	2178	1850	1965	<b>1850</b>	x	1641	1	0.8	0.95	0.5	1406	703	58	38
<sup>233</sup> U	S	568	560	425	x	<b>425</b>	454	x	0.8	0.8	0.95	0.7	258	129	10	12
<sup>234</sup> U	F	134000	137350	83000	85330	<b>83000</b>	x	63138	1	0.8	0.95	0.5	63080	31540	2628	1729
<sup>235</sup> U	S	784	785	589	x	<b>589</b>	620	x	0.8	0.8	1	0.7	376	188	15	17
<sup>236</sup> U	NF															0
<sup>238</sup> U	NF															0
<sup>235</sup> Np	F	9480	x	6180	x	<b>6180</b>	x	x	1	0.8	0.2		988	494	41	
<sup>236</sup> Np	S	72	x	63	x	<b>63</b>	x	x	0.8	0.8	0.2		8	4	0.3	
<sup>237</sup> Np	F	57500	75440	38600	49960	<b>38600</b>	x	37213	1	0.75	0.7	0.7	20265	10132	844	1125
<sup>236</sup> Pu	F	3310	5040	970	4000	<b>970</b>	x	3287	1	0.8	0.9	0.5	698	349	29	20
<sup>237</sup> Pu	S	136	x	110	x	<b>110</b>	x	x	0.8	0.8	0.2		14	7	0.5	
<sup>238</sup> Pu	F	7350	7380	4700	4780	<b>4700</b>	x	3930	1	0.8	1	0.7	3760	1880	156	137
<sup>239</sup> Pu	S	494	503	353	x	<b>353</b>	394	x	0.8	0.8	1	0.7	225	112	9.3	10.2
<sup>240</sup> Pu	F	32100	34950	18300	22580	<b>18300</b>	x	17790	1	0.8	0.85		12444	6222	518	
<sup>241</sup> Pu	S	246	269	205	x	<b>205</b>	217	x	0.8	0.8	0.9	0.7	118	59	4.9	5.9
<sup>242</sup> Pu	F	78200	69350	36200	44240	<b>36200</b>	x	34127	1	0.75	0.9	0.7	24435	12217	1018	1055
<sup>244</sup> Pu	F	222000	x	60300	x	<b>60300</b>	x	x	1	0.8	0.2		9648	4824	402	

Nuclide	Spectrum	Critical mass (g) (k=1)					Allowable mass (k=0.95)		Factors to consider on Mc				Safe Mc (g)	Mass per group of package (g)	Mass per package (g)	From ANS safe mass per package
		20 cm water		30 cm steel/lead		Min	20 cm water (IRSN)	30 cm steel (IRSN)	heterogeneity reflectors moderator F1	Margin of 5% in Δk F2	Libraries F3	ANS 8.15				
		EMS	IRSN	EMS	IRSN											
<sup>241</sup> Am	F	52500	67770	33800	44000	<b>33800</b>	x	32207	1	0.7	0.75	0.7	17745	8872	739	985
<sup>242m</sup> Am	S	20	23	17	x	<b>17</b>	19	x	0.8	0.8	0.85		9	4.5	0.37	
<sup>243</sup> Am	F	195000	192800	88600	132350	<b>88600</b>	x	92506	1	0.7	0.95		58919	29459	2454	
<sup>242</sup> Cm	F	260000	17600	7720	12225	<b>7720</b>	x	9934	1	0.8	0.2		1235	617	51	
<sup>243</sup> Cm	S	127	269	106	x	<b>106</b>	227	x	0.8	0.8	0.5	0.5	33	16.5	1.37	2.20
<sup>244</sup> Cm	F	22100	27070	13200	16810	<b>13200</b>	x	13336	1	0.85	0.75	0.5	8415	4207	350	275
<sup>245</sup> Cm	S	54	47	42	x	<b>42</b>	40	x	0.8	0.8	0.85	0.5	22	11	0.91	0.87
<sup>246</sup> Cm	F	33600	x	22000	23200	<b>22000</b>	x	19239	1	0.8	0.2	0.5	3520	1760	146	458
<sup>247</sup> Cm	S	2180	2200	796	x	<b>796</b>	1773	x	0.8	0.8	0.2	0.5	101	50	4	16
<sup>248</sup> Cm	F	34700	x	21500	x	<b>21500</b>	x	x	1	0.8	0.2		3440	1720	143	
<sup>250</sup> Cm	F	21400	x	14700	x	<b>14700</b>	x	x	1	0.8	0.2		2352	1176	98	
<sup>247</sup> Bk	F	41200	x	35200	x	<b>35200</b>	x	x	1	0.8	0.2		5632	2816	234	
<sup>248</sup> Bk	F	x	x	x	x	<b>x</b>	x	x	1	1	0.2		x	x	x	
<sup>249</sup> Bk	F	179000	x	131000	x	<b>131000</b>	x	x	1	0.8	0.2		20960	10480	873	
<sup>248</sup> Cf	F	x	x	x	x	<b>x</b>	x	x	1	0.8	0.2		x	x	x	
<sup>249</sup> Cf	S	60	x	51	x	<b>51</b>	x	x	0.8	0.8	0.2	0.2	6	3	0.25	
<sup>250</sup> Cf	F	5610	x	3130	x	<b>3130</b>	x	x	1	0.8	0.2	0.2	500	250	20	26
<sup>251</sup> Cf	S	25	x	20	x	<b>20</b>	x	x	0.8	0.8	0.2	0.2	2	1	0.083	0.167
<sup>252</sup> Cf	F	2910	x	2950	x	<b>2910</b>	x	x	1	0.8	0.2	0.2	465	232	19	24
<sup>254</sup> Cf	F	2860	x	2250	x	<b>2250</b>	x	x	1	0.8	0.2	0.2	360	180	15	18
<sup>252</sup> Es	F	x	x	x	x	<b>x</b>	x	x	1	0.8	0.2		x	x		
<sup>254</sup> Es	F	33	x	32	x	<b>32</b>	x	x	1	0.8	0.2	0.2	5.1	2.5	0.20	
<sup>257</sup> Fm	F	x	x	x	x	<b>x</b>	x	x	1	0.8	0.2		x	x		
<sup>258a</sup> Md	F	x	x	x	x	<b>x</b>	x	x	1	0.8	0.2		x	x		

## APPENDIX I – Effect of a Carbon moderator for U235

We tried to see if the safety factors used to define the exception criteria (see § 4.3 and appendix H) cover the cases of a carbon or a graphite moderator. For  $^{235}\text{U}$ , the maximum mass per package is 15 g and the maximum mass per group of packages has been set to 190 g (appendix H – considering the safety factor F1 and a safety margin of 5%): This means that we can only transport 12 packages if they contain 15 g of  $^{235}\text{U}$ . So, we have to check for the normal conditions of transport if 5x12 (60) packages are leading to a  $k_{\text{eff}}$  equal or higher to 0.95. Consequently, we performed calculations with an array of 4x4x4 (64) packages with taking into account different “standard” reflectors: 20 cm of water, 30 cm of steel, 60 cm of concrete and 25 cm of lead adding to 20 cm of water. Results are plotted in the graph below:



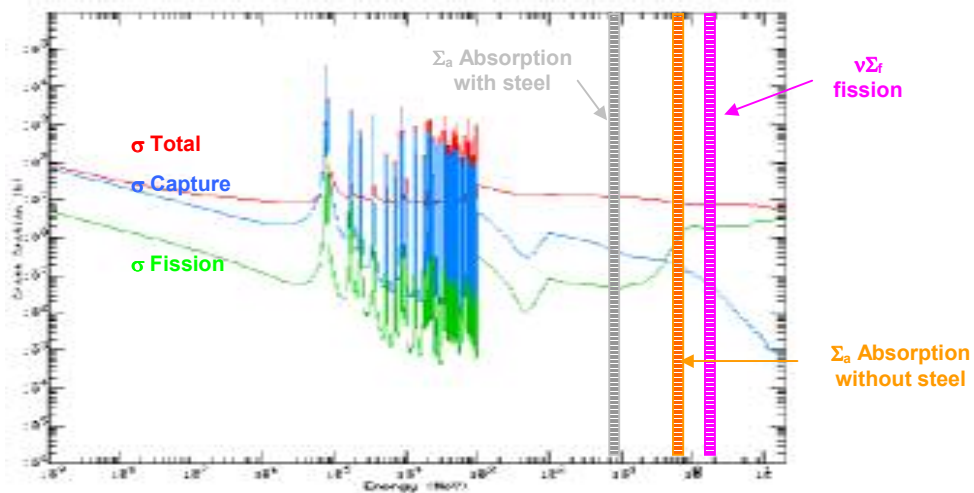
**Graph 1.**  $k_{\text{eff}}$  in normal conditions of transport (array) for the  $^{235}\text{U}$  moderated by natural carbon with taking into account the limitations of transportable mass.

We see from the graph that, even with the better reflector (lead with water), the maximum  $k_{\text{eff}}$  obtained is 0.763, which is inferior to 0.95 (-18,7% in  $\Delta k$ ). We can consider that, even if carbon constitute a potential risk, the safety margins (appendix H) taken in the study are sufficient to prevent a criticality incident during a transport in normal conditions.

EMS indicates that mixtures such as carbon and water need to be evaluated when normal conditions are studied. Mixtures of wet carbon powder and fissile nuclides will give higher values of  $k_{\text{eff}}$  for 10x10x10 cm<sup>3</sup> sized packages.

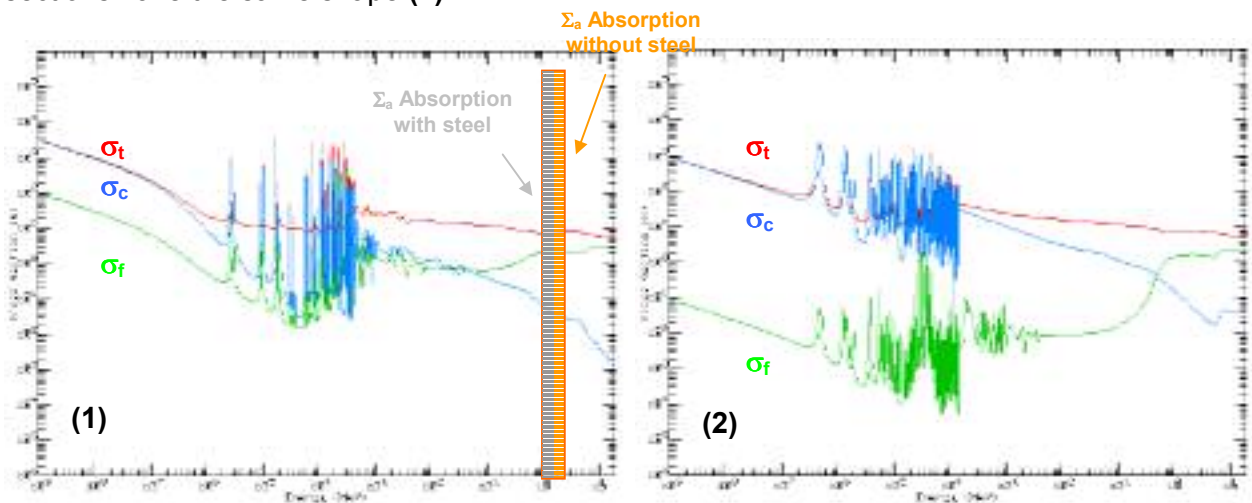
## APPENDIX J – Most penalizing cases for arrays of packages containing 1 kg of Cm244 or 1 kg of Pu238

We can see that under 0.4 MeV, the fission cross section is lower than the capture cross-section; above 0.4 MeV, it is the opposite. As the average value of the absorption without a steel wrapper (void wrapper) is above 0.4 MeV (~ 0.5 MeV) and the average value of the absorption with a steel wrapper is under 0.4 MeV (~ 0.06 MeV) it can explain the shape of the curves. As we can see, steel scatters a little bit neutrons.



**Graph 1. Microscopic cross section of <sup>244</sup>Cm (JEF 2.2)**

There is no such phenomenon with <sup>238</sup>Pu because the fission cross-section is above the capture cross section with and without a steel wrapper (see **Graph 2 (1)**). We also performed a calculation with a steel wrapper for <sup>237</sup>Np and found the same effect as for <sup>244</sup>Cm because the microscopic cross sections have the same shape **(2)**:



**Graph 2. Microscopic cross section of <sup>238</sup>Pu (1) and <sup>237</sup>Np (2) (JEF 2.2)**

## APPENDIX K – Effect of reflectors on critical masses (EMS)

The values in Orange corresponds to best reflector and the values in purple corresponds to the lowest values (for the different libraries)

After completing the project for the European Commission, EMS is continuing the study on actinide nuclides and exception limits for the transport regulations. The following tables contain preliminary data from this continued study. In addition, results involving natural uranium will be made available for all nuclides. Validation by comparison with critical experiments is being carried out. Evaluated specifications and results from critical experiments are publicly available for  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  in almost pure forms. Concerning  $^{241}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{244}\text{Cm}$  and  $^{237}\text{Np}$ , there are experiments giving reactivity influences. They are not reliable for determining the critical mass with each nuclide on its own. For other actinide nuclides, there is even less experimental support. Concerning reflectors, there are several experiments with water, steel, lead and other reflectors. There are also many experiments with natural uranium surrounding other fissionable materials. The preliminary JENDL-3.3 and ENDF/B-VI.8 libraries are giving quite good and consistent results so far. Exceptions are the nuclides for which problems are indicated for in the table of  $k_{\text{inf}}$ .

All results are preliminary. A safety factor of 10 or 20 may be motivated for nuclides without experimental support, unless other conservative measures have been applied. Even when experimental results are available from a single source, care must be taken since the specified data may be incorrect.

### Metals at theoretical density

Actinide nuclide	Cross-section library	Critical mass (kg)				
		Bare	H <sub>2</sub> O 20 cm	SS 30 cm	Pb 25 cm	1.5 cm H <sub>2</sub> O + 30 cm SS
Th-229	JENDL-3.2	2839	2262	994	1082	
U-232	ENDF/B-V-238	3.57	2.13	1.85	2.11	1.66
	ENDF/B-VI.8					2.92
	JENDL-3.2					5.91
	ENDF/B-VI.5					1.80
U-233	ENDF/B-V-238	15.7 9	7.32	6.11	7.62	5.02
	ENDF/B-VI.8					5.52
	JENDL-3.3					5.45
U-234	ENDF/B-V-238	145	134	83.0	79.6	
	ENDF/B-VI.8				86.0	
	JENDL-3.3			86.1	87.4	
	JEF-2.2			85.1	84.8	
U-235	ENDF/B-V-238	46.7	22.0	16.8	21.2	14.3
	ENDF/B-VI.8					15.7
	JENDL-3.3					15.4
	JEF-2.2					15.2
Np-235	ENDL-92	66.2	60.0	38.8	38.4	
	JENDL-3.3	12.0	9.48	6.18	6.93	
Np-236	ENDL-92	6.79	3.21	3.30	3.89	2.52
	ENDF/B-VI.8	6.86	3.10	3.35	3.94	2.58
	JENDL-3.2	15.8	8.31	7.18	8.67	6.22
Np-237	ENDF/B-VI.2 DN	63.6	57.5	38.6	36.6	
	ENDF/B-VI.8			38.1	37.2	
	JENDL-3.3			37.9	39.8	
	JEF-2.2			47.6	47.3	



Actinide nuclide	Cross-section library	Critical mass (kg)				
		Bare	H <sub>2</sub> O 20 cm	SS 30 cm	Pb 25 cm	1.5 cm H <sub>2</sub> O + 30 cm SS
Pu-236	ENDF/B-V-238	8.04	4.99	3.74	4.26	3.84
	ENDF/B-VI.8	6.56	3.31	3.15	3.74	2.60
	JENDL-3.3	7.40	3.58	3.52	4.31	2.81
Pu-237	ENDF/B-VI.8	3.10	1.71	1.62	1.87	1.40
	JENDL-3.3	3.58	1.99	1.91	2.23	1.64
Pu-238	ENDF/B-V-238	9.49	7.35	4.70	5.25	5.11
	ENDF/B-VI.8			5.15		
	JENDL-3.3			4.76		
	JEF-2.2			4.69		
Pu-239	ENDF/B-V-238	9.99	5.45	4.49	5.30	4.10
	ENDF/B-VI.8					4.33
	JENDL-3.3					4.33
	JEF-2.2					4.33
Pu-240	ENDF/B-V-238	35.7	32.1	19.8	19.8	25.5 (1 cm H <sub>2</sub> O)
	ENDF/B-VI.8			20.9	21.06	
	JENDL-3.3			18.3	19.4	
	JEF-2.2			21.3	21.9	
Pu-241	ENDF/B-V-238	12.3	5.87	5.05	6.23	4.08
	ENDF/B-VI.8					4.38
	JENDL-3.3					4.16
	JEF-2.2					4.46
Pu-242	ENDF/B-V-238	85.6	78.2	48.1	45.8	
	ENDF/B-VI.8			50.7	49.2	
	JENDL-3.3			36.2	37.3	
	JEF-2.2			43.3	42.9	
Pu-244	ENDF/B-V-238	241	222	133	123	
	ENDF/B-VI.8			140	130	
	JENDL-3.3			60.3	61.0	
Am-241	ENDF/B-VI.2 DN	57.6	52.5	33.8	30.7	
	ENDF/B-VI.8			33.9	31.4	
	JENDL-3.2			42.5	43.7	
	JEF-2.2			40.5	37.6	
Am-242m	ENDF/B-V-238	8.83	3.21	3.00	3.87	2.15
	ENDF/B-VI.8					2.37
	JENDL-3.3					3.20
Am-243	ENDF/B-VI.2 DN	209	195	138	121	
	ENDF/B-VI.8			88.5		
	JENDL-3.3			133	133	
	JEF-2.2			125		
Cm-242	ENDF/B-V-238	371	260	231	197	
	ENDF/B-VI.8			239	205	
	JENDL-3.3	15.4	11.2	7.72	8.52	
	JEF-2.2			11.6	12.7	
Cm-243	ENDF/B-V-238	8.35	2.82	3.10	3.82	2.19
	ENDF/B-VI.5					2.21
	JENDL-3.3					2.07
	JEF-2.2					2.14
Cm-244	ENDF/B-V-238	26.6	22.1	13.2	13.0	
	ENDF/B-VI.8			13.9	13.8	
	JENDL-3.3			15.6	15.8	
	JEF-2.2			15.9	16.4	

Actinide nuclide	Cross-section library	Critical mass (kg)				
		Bare	H <sub>2</sub> O 20 cm	SS 30 cm	Pb 25 cm	1.5 cm H <sub>2</sub> O + 30 cm SS
Cm-245	ENDF/B-V-238	9.11	3.08	3.46	4.31	2.15
	ENDF/B-VI.5					2.36
	JENDL-3.3					3.00 (SS?)
	JEF-2.2					1.87
Cm-246	ENDF/B-V-238	38.9	33.6	22.0	20.4	
	ENDF/B-VI.8			25.9	25.0	
	JENDL-3.3			23.1 (SS?)	24.5	
	JEF-2.2			22.0	21.1	
Cm-247	ENDF/B-V-238	6.94	3.52	2.84	3.37	2.73
	ENDF/B-VI.8					2.80
	JENDL-3.3					2.36
	JEF-2.2					2.80
Cm-248	ENDF/B-V-238	40.4	34.7	21.5	20.5	
	ENDF/B-VI.8			22.6	21.5	
	JENDL-3.3			33.1 (SS?)	34.6	
	JEF-2.2			22.1	21.5	
Cm-250	JENDL-3.3	23.5	21.4	14.7	15.2	
Bk-247	JENDL-3.3	75.7	41.2	35.2	42.6	30.4
Bk-249	ENDF/B-VI.5	192	179	131	117	
	ENDF/B-VI.8			132	117	
	JENDL-3.2			150		
Bk-250	JENDL-3.2-J	6.17	2.83	2.76	3.36	2.44
Cf-249	ENDF/B-V-238	5.91	2.28	2.39	2.95	1.65
	ENDF/B-VI.8					2.09
	JENDL-3.3					1.91
Cf-250	ENDF/B-V-238	6.55	5.61	3.13	3.45	
	ENDF/B-VI.8			3.34		
	JENDL-3.2			5.19		
Cf-251	ENDF/B-V-238	5.46	2.45	2.27	2.77	1.85
	ENDF/B-VI.8					1.90
	JENDL-3.2					1.12
Cf-252	ENDF/B-V-238	5.87	2.91	3.32	3.51	2.72
	ENDF/B-VI.8					2.65
	JENDL-3.2					1.94
Cf-254	JENDL-3.2	4.27	2.86	2.25	2.56	2.43
Es-254	JENDL-3.2	9.89	2.26	2.90	4.22	1.85
Fm-255	JENDL-3.2	6.73	1.77	2.11	3.07	1.57

### Uniform homogeneous water-moderated critical masses

Actinide nuclide	Cross-section library	Bare mass	Approximate minimum critical mass (g) with reflectors						
			H/X	Bare	H/X	H <sub>2</sub> O 20 cm	H/X	SS 30 cm	Pb 25 cm
U-232	ENDF/B-V-238	67.5	9050	53.7	5000	29.2*	2910*	3260*	3250*
	ENDF/B-VI.8					29.2	3450		
	JENDL-3.2					29.2	12000		
	ENDF/B-VI.5					29.2	3070		
U-233	ENDF/B-V-238	550	1080	458	568	392*	424*	440*	420*
	ENDF/B-VI.8					392			432
	JENDL-3.3					392			432

Actinide nuclide	Cross-section library	Bare mass H/X	Approximate minimum critical mass (g) with reflectors						
			Bare	H/X	H <sub>2</sub> O 20 cm	H/X	SS 30 cm	Pb 25 cm	1.5 cm H <sub>2</sub> O + 30 cm SS
U-235	ENDF/B-V-238	550	1420	458	784	458	589	595	588
	ENDF/B-VI.8					458			615
	JENDL-3.3					458			615
	JEF 2.2					458			609
Np-236	ENDL-92	3208	147	2138	72.0	2566	59.3	62.0	55.2
	ENDF/B-VI.8					2566			34.1
	JENDL-3.2					2566			163
Pu-236	ENDF/B-V-238	132	16600	110	10800	110	6930	6920	7770
	ENDF/B-VI.2					110	11200		
	ENDF/B-VI.8					132			834
	JENDL-3.3					132			841
Pu-237	ENDF/B-VI.5	2220	257	1910	136	1910	110	111	105
	ENDF/B-VI.8					1910			105
	JENDL-3.3					1910			83.3
Pu-239	ENDF/B-V-238	988	877	889	494	889	353		
	ENDF/B-VI.8					889	372		
	JENDL-3.3					889	369		
	JEF 2.2					889	363		
Pu-241	ENDF/B-V-238	1212	511	784	246	784	205	219	204
	ENDF/B-VI.8					889			208
	JENDL-3.3					889			209
	JEF 2.2					889			207
Am-242m	ENDF/B-V-238	10300	42	7820	20	7820	17	18	15.7
	ENDF/B-VI.8					7820			16.5
	JENDL-3.3					7820			18.1
Cm-243	ENDF/B-V-238	1052	280	740	127	665*	105*	115*	97*
	ENDF/B-VI.5					665			103
	JENDL-3.3					665			153
	JEF 2.2					665			214
Cm-245	ENDF/B-V-238	3330	116	2220	54	2220	42	45	39
	ENDF/B-VI.5					2220			44.2
	JENDL-3.3					2220			49.1
	JEF 2.2					2220			38.5
Cm-247	ENDF/B-V-238	141	4060	109	2180	109	1430	1580	1510
	ENDF/B-VI.8					109	1530		1560
	JENDL-3.3					141	796		760
	JEF 2.2					109	1500		1540
Bk-247	JENDL-3.3	44.8	12400 0	29.3	89900	24800	52400	57500	59700
Bk-250	JENDL-3.2	1240	384	932	191	932	164	171	152
Cf-249	ENDF/B-V-238	3040	129	1830	60	2280*	50*	54*	46*
	ENDF/B-VI.2					2280			54.2
	JENDL-3.3					2280			49.4
Cf-251	ENDF/B-V-238	8310	48	7030	25	7030	20	21	19
	ENDF/B-VI.8					7030			19.5
	JENDL-3.2					7030			26.1
Cf-252	ENDF/B-V-238	50.4	7930	34.7	3860	34.7	2950	3210	2840
	ENDF/B-VI.8					34.7			2982
	JENDL-3.2					34.7			3020
Es-254	JENDL-3.2	3170	76.6	2110	33.3	2110	32.1	35.5	27.4
Fm-255	JENDL-3.2	6340	39.4	3960	16.5	3960	16.8	18.1	13.3