

TECHNICAL REPORT

Critical and Subcritical Mass Calculations of Curium-243 to -247 Based on JENDL-3.2 for Revision of ANSI/ANS-8.15

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(Received April 17, 2002 and accepted in revised form July 26, 2002)

Critical and subcritical masses were calculated for a sphere of five curium isotopes from ²⁴³Cm to ²⁴⁷Cm in metal and in metal-water mixtures considering three reflector conditions: bare, with a water reflector or a stainless steel reflector. The calculation were made mainly with a combination of a continuous energy Monte Carlo neutron transport calculation code, MCNP, and the Japanese Evaluated Nuclear Data Library, JENDL-3.2. Other evaluated nuclear data files, ENDF/B-VI and JEF-2.2, were also applied to find differences in calculation results of the neutron multiplication factor originated from different nuclear data files. A large dependence on the evaluated nuclear data files was found in the calculation results: more than 10% $\Delta k/k$ relative differences in the neutron multiplication factor for a homogeneous mixture of ²⁴³Cm metal and water when JENDL-3.2 was replaced with ENDF/B-VI and JEF-2.2, respectively; and a 44% reduction in the critical mass by changing from JENDL-3.2 to ENDF/B-VI for ²⁴⁶Cm metal. The present study supplied basic information to the ANSI/ANS-8.15 Working Group for revision of the standard for nuclear criticality control of special actinide elements. The new or revised values of the subcritical mass limits for curium isotopes accepted by the ANSI/ANS-8.15 Working Group were finally summarized.

KEYWORDS: critical mass, subcritical mass, curium 243, curium 244, curium 245, curium 246, curium 247, bare, water, SS-304, JENDL-3.2, MCNP, ANSI/ANS-8.15, revision

I. Introduction

The American National Standard for Nuclear Criticality Control of Special Actinide Elements, ANSI/ANS-8.15-1981,¹⁾ has been reviewed for revision by a working group chaired by Norman L. Pruvost since 1996.²⁾ The standard was intended to provide criticality safety guidance for fissionable actinides other than the so called “big three” actinides, ²³³U, ²³⁵U and ²³⁹Pu, that have been traditionally discussed by the ANSI/ANS-8.1 Working Group. The ANSI/ANS-8.15 Working Group was arranged to consist of five US members and four non-US members, who are from France, Japan, Russia, and UK. One of the authors of this paper (HO) has been the Japanese member of the group since 1997, to whom at first three curium isotopes were assigned: ²⁴⁶Cm for inclusion, and ²⁴⁵Cm and ²⁴⁷Cm for revision.

There were no critical experiments and only one replacement experiment for ²⁴⁴Cm concerning curium isotopes.³⁾ Consequently, we could almost only rely on calculation for determining their critical masses. There were several documents so far that supplied critical mass calculation of curium isotopes.^{4–11)} They gave us important information; however, none of the documents covered the whole in the aspect of nuclides, moderation and reflector conditions.

Present authors, therefore, calculated critical and subcritical masses of ²⁴⁵Cm, ²⁴⁶Cm and ²⁴⁷Cm to accomplish the task.¹²⁾ In addition to the requested three curium isotopes, they recently made calculation for ²⁴³Cm and ²⁴⁴Cm as a part of ten fissionable nuclides that had comparable critical masses to that of ²³⁵U.¹³⁾ The critical and subcritical masses of the curium isotopes were calculated as precisely as pos-

sible by using modern computational tools and the most recently published evaluated nuclear data files for standardizing these quantities in reference to the previous data. Based on the present and previously obtained critical mass values subcritical mass limits were recommended to the ANS-8.15 Working Group and the values were discussed by its members.

This paper is intended to make a report covering the critical and subcritical mass calculations of the five curium isotopes, ²⁴³Cm to ²⁴⁷Cm, and also to introduce non-members the relevant activities of ANSI/ANS-8.15 Working Group to determine the subcritical mass limits of the curium isotopes.

II. Calculation of Critical and Subcritical Masses

1. Calculation Object

The critical mass is defined as the mass corresponding to the neutron multiplication factor, $k_{\text{eff}}=1$. The subcritical masses obtained in this paper are limited to the masses corresponding to $k_{\text{eff}}=0.9$ and 0.8. Note that the critical and subcritical masses by these definitions depend on the nuclear data library, other data assumed in the calculation, such as the actinide density, reflector conditions, *etc.*, and computation methods employed.

As stated in Chap. I, five curium isotopes, ²⁴³Cm to ²⁴⁷Cm, in a metallic state are discussed in this paper. For the fissile curium isotopes, *i.e.*, ²⁴³Cm, ²⁴⁵Cm and ²⁴⁷Cm, that are capable of chain fission reactions by thermal neutrons, mixtures of metal and water are also objects of the study.

The density of each curium isotope in metal was determined based on crystallographic data compiled by R. G. Haire and calculated by J. E. Bigelow¹⁴⁾ in accordance with a consensus among the ANSI/ANS-8.15 Working Group members. For ²⁴⁴Cm metal, they suggested 244.06275 for

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atomic weight, 13.518 g/cm³ for material density of metal and 3.335 × 10²² at./cm³ for atomic density.

The shape of the fuel region was limited to a sphere and the reflector conditions assumed were three kinds: bare, surrounded with a 30-cm-thick reflector of water and a 30-cm-thick reflector of stainless steel (SS) of Type 304. The uniformity of fuel was assumed throughout this paper.

2. Method of Calculation

The neutron transport code applied for criticality calculation was a continuous energy Monte Carlo code MCNP developed at the Los Alamos National Laboratory of the USA. The versions of MCNP we used were 4B2¹⁵⁾ for ²⁴³Cm and ²⁴⁴Cm; and 4A¹⁶⁾ for ²⁴⁵Cm, ²⁴⁶Cm and ²⁴⁷Cm. The numbers of tallied source histories of the MCNP calculation were set to a million for ²⁴³Cm and ²⁴⁴Cm and a half million for ²⁴⁵Cm, ²⁴⁶Cm and ²⁴⁷Cm. The standard deviations for million-history calculations were typically 0.1% Δk.

The evaluated nuclear data file adopted as a standard was the latest released Japanese Evaluated Nuclear Data Library, JENDL-3.2.¹⁷⁾ Other evaluated nuclear data files, ENDF/B-VI¹⁸⁾ and JEF-2.2,¹⁹⁾ were also applied in addition to JENDL-3.2 to see differences in k_{eff} caused by differences in the evaluated nuclear data files.

For determining the critical/subcritical sphere radius of fuel, i.e., actinide metal or mixtures of actinide metal and water, a continuous curve having an algebraic form proposed by Rombough *et al.*²⁰⁾ was adopted for fitting the calculated results. The Rombough’s curve was already employed in “Nuclear Criticality Safety Guide”²¹⁾ to derive critical and subcritical mass/dimension curves for ²³³U-H₂O, ²³⁵U-H₂O and ²³⁹Pu-H₂O. More specifically, in order to determine the value of a sphere’s radius r for a particular value of k_{eff}, at least 4 sets of r and computationally obtained k_{eff} values were fitted to a continuous curve having the following algebraic form:

$$k_{eff}(r) = k_{\infty}(1 - e^{-r/\alpha})^{\beta r^{\gamma}}, \tag{1}$$

where k_∞ is the neutron multiplication factor of fuel in infinite media, and α, β and γ are parameters to be determined by the least square fitting of the points to the curve. The statistical errors in the Monte Carlo calculations were disregarded in fitting the curve.

The corresponding curium mass M was then obtained by the well-known formula:

$$M = \rho \cdot 4\pi r^3 / 3, \tag{2}$$

where ρ is the mass density of curium. It is worth pointing out that we took into account the differences in the density ρ by the mass numbers of curium isotopes (see **Table 1**).

For odd neutron-number curium isotopes, i.e., ²⁴³Cm, ²⁴⁵Cm and ²⁴⁷Cm, which are fissile, 10 values were selected to cover the curium concentration range, from very low to high as metal for mixtures of curium metal and water to calculate the critical/subcritical masses of curium. The critical/subcritical curium masses for these 10 values were determined, and three curves for critical/subcritical masses of curium vs. curium concentrations were drawn through these points by the spline interpolation method. The minimum critical and subcritical masses of curium were determined as the

Table 1 Critical (k_{eff}=1.0) and subcritical (k_{eff}=0.9 and 0.8) masses of curium isotopes in metal calculated with the MCNP code and JEND-3.2 library

Nuclide	Density (g/cm ³)	Reflector	Mass (kg)		
			k _{eff} =1.0	k _{eff} =0.9	k _{eff} =0.8
²⁴³ Cm	13.45	None	10.03	7.08	4.80
		Water	3.51	2.43	1.60
		SS-304	3.91	2.84	2.05
²⁴⁴ Cm	13.51	None	29.95	20.07	13.20
		Water	24.84	16.51	10.72
		SS-304	15.36	10.62	7.11
²⁴⁵ Cm	13.57	None	12.3	8.71	5.99
		Water	3.91	2.71	1.82
		SS-304	4.73	3.49	2.51
²⁴⁶ Cm	13.62	None	70.1	45.5	28.9
		Water	57.4	37.0	23.5
		SS-304	38.1	25.2	16.2
²⁴⁷ Cm	13.68	None	7.06	5.02	3.44
		Water	3.10	2.18	1.49
		SS304	3.07	2.28	1.63

minimum values of these three curves.

3. Results

An example of the relation between the radius of a fuel sphere and the calculated neutron multiplication factor k_{eff} is shown in **Fig. 1** for ²⁴⁵Cm metal without reflectors. This figure illustrates how to obtain the radii of fuel sphere, 6.00, 5.35 and 4.72 cm, corresponding to k_{eff}=1, 0.9 and 0.8, respectively, from the fitted curve. The critical mass was derived as 12.3 kgCm, and the subcritical masses corresponding to k_{eff}=0.9 and 0.8 were determined to be 8.71 and 5.99 kgCm, respectively, by multiplying the fuel density 13.57 gCm/cm³ to the corresponding volumes (see Eq. (2)).

(1) Critical and Subcritical Masses of ²⁴³Cm to ²⁴⁷Cm in Metal

The critical and subcritical masses of a sphere of the five curium isotopes, ²⁴³Cm to ²⁴⁷Cm, in metal were calculated with the three reflector conditions: bare, with a 30-cm-thick

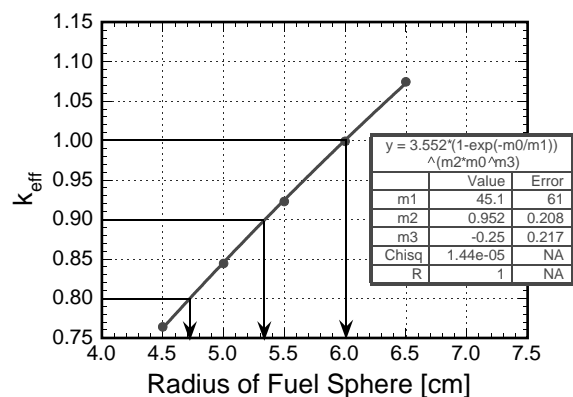


Fig. 1 Relation between spherical radius of ²⁴⁵Cm metal without reflector and calculated values of k_{eff}

water reflector and with a 30-cm-thick SS-304 reflector. The results are summarized in Table 1 and Fig. 2. It is clear from Fig. 2 that the critical/subcritical mass of a curium isotope decreases in the order of ²⁴⁶Cm, ²⁴⁴Cm, ²⁴⁵Cm, ²⁴³Cm and ²⁴⁷Cm for each reflector condition.

The relations between the critical mass of a bare metal sphere, M_{crit} , and the corresponding k_{∞} are plotted for the curium isotopes in Fig. 3. They are almost all on one curve,

$$M_{crit} = \frac{1}{c_1 + c_2 \cdot k_{\infty}^3}, \quad (3)$$

where c_1 , c_2 and c_3 are constants. Note that both fissile and non-fissile nuclides are included in the plot. Fissile curium isotopes, ^{243,245,247}Cm, have large k_{∞} values, 3.5–3.9, and small critical mass values, 7–12 kg; whereas non-fissile curium isotopes, ^{244,246}Cm, have small k_{∞} values, 2.3–2.6, and large critical mass values, 30–70 kg.

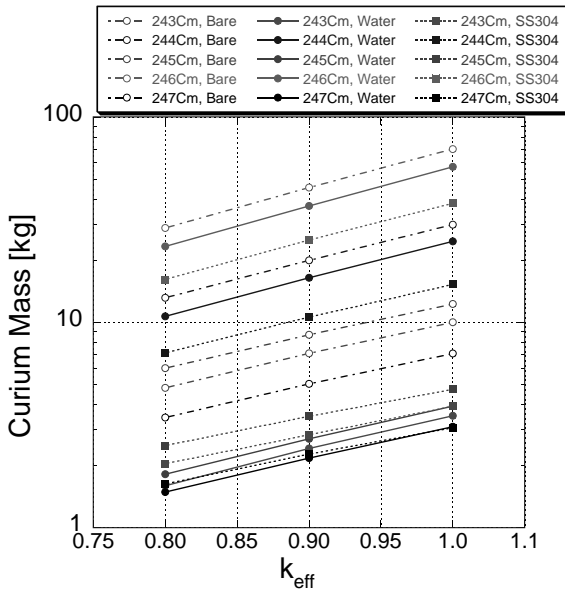


Fig. 2 Critical ($k_{eff}=1.0$) and subcritical ($k_{eff}=0.9$ and 0.8) masses of curium isotopes in metal calculated with the MCNP code and JEND-3.2 library

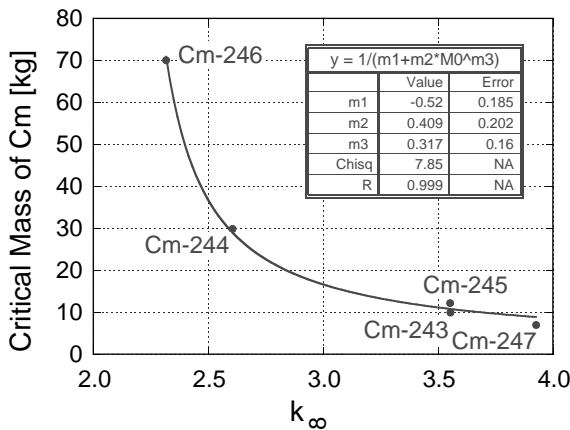


Fig. 3 Relation between the critical mass of curium isotopes in bare metal and their k_{∞}

From Fig. 2, it is observed that the lines are almost parallel, *i.e.*, the mass reduction rates are almost the same irrespective of curium isotopes and reflector conditions. Relative reduction in masses corresponding to $k_{eff}=0.9$ and 0.8 to the mass corresponding to $k_{eff}=1.0$ for curium isotopes are listed in Table 2. It shows that the subcritical masses are reduced from the critical mass to $(69\pm 5)\%$ for $k_{eff}=0.9$ and $(47\pm 6)\%$ for $k_{eff}=0.8$. The maximum deviation from the average is found to be rather small for both $k_{eff}=0.9$ and 0.8 . These relations give a rough idea that any uncertainties in obtaining k_{eff} should make differences in the critical mass of curium: An error of 0.1% in k_{eff} will produce a 0.3% error in the critical mass calculation.

The effect of reflector on the critical mass is generally different from isotope to isotope. However, it is clearly seen from Fig. 4, which shows the critical masses of curium isotopes in the three reflector conditions in a bar graph, that non-

Table 2 Relative reduction of masses in $k_{eff}=0.9$ and 0.8 to the mass in $k_{eff}=1.0$ for curium isotopes

Nuclide	Reflector	M(0.9)/M(1)	M(0.8)/M(1)
²⁴³ Cm	None	0.71	0.48
	Water	0.69	0.46
	SS-304	0.73	0.52
²⁴⁴ Cm	None	0.67	0.44
	Water	0.66	0.43
	SS-304	0.69	0.46
²⁴⁵ Cm	None	0.71	0.49
	Water	0.69	0.47
	SS-304	0.74	0.53
²⁴⁶ Cm	None	0.65	0.41
	Water	0.64	0.41
	SS-304	0.66	0.43
²⁴⁷ Cm	None	0.71	0.49
	Water	0.70	0.48
	SS304	0.74	0.53
Average		0.693	0.468
Minimum		0.645	0.409
Maximum		0.743	0.531

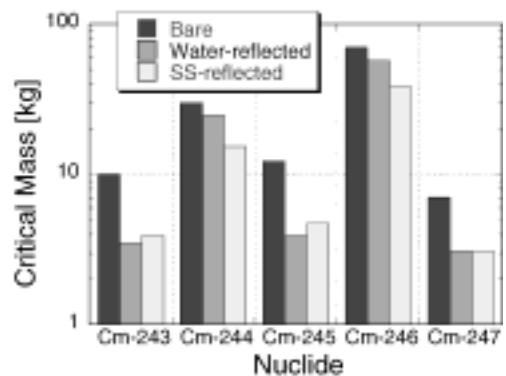


Fig. 4 Critical mass of curium isotopes in metal in bare, water-reflected and SS-reflected

fissile curium isotopes (^{244}Cm and ^{246}Cm) and fissile curium isotopes (^{243}Cm , ^{245}Cm and ^{247}Cm) each have a common tendency. Non-fissile curium isotopes have only a little water-reflector effect: the critical mass in water-reflected condition reduces less than 20% from the bare critical mass; SS-reflector effect is larger: the reduction rate is nearly 50%. Fissile curium isotopes have a large reflector effect both by a water reflector and by an SS reflector. The critical mass of a fissile curium isotope in a metal sphere reflected by either water or SS becomes less than half of that in the bare condition.

It is noted that steel is a better reflector than water for uranium and plutonium isotopes; however, this is not necessarily true for americium and curium isotopes,¹³⁾ esp. for ^{245}Cm in metal; as shown in Fig. 4 (see also Table 10). Moderation in addition to reflection by water helps to reduce the critical mass for the fissile nuclides having remarkable fissile character, like ^{245}Cm (see Sec. II-3(2)).

(2) Minimum Critical and Subcritical Masses of ^{243}Cm , ^{245}Cm and ^{247}Cm in Metal-water Mixtures

Critical and subcritical masses of a sphere of $^{243}\text{Cm-H}_2\text{O}$ in bare, with a water reflector and with an SS reflector are shown in Fig. 5. Although their values are different, their behaviors as a function of curium concentration are similar irrespective of the reflector conditions; and they are decreasing in order of magnitude, (a) bare, (b) 30-cm-thick water reflected, and (c) 30-cm-thick SS-304 reflected cases.

Quite similarly to ^{243}Cm , critical and subcritical masses of homogeneous $^{245}\text{Cm-H}_2\text{O}$ and $^{247}\text{Cm-H}_2\text{O}$ in a sphere were also calculated for various curium concentrations. Figures 6 and 7 show the homogeneous $^{245}\text{Cm-H}_2\text{O}$ and $^{247}\text{Cm-H}_2\text{O}$ results, respectively, for the same three reflector conditions as homogeneous $^{243}\text{Cm-H}_2\text{O}$. The critical and subcritical masses of curium in metal are not so different for ^{245}Cm and ^{247}Cm from ^{243}Cm (see Fig. 4). However, the decrease rate of critical/subcritical curium mass with decreased curium concentration, which is the gradient of mass slope in the figures, is quite different for these three isotopes. Curium-245 has a noticeable fissile character: the minimum critical mass is almost two orders of magnitude smaller than the critical mass in metal. For the case of homogeneous $^{247}\text{Cm-H}_2\text{O}$, on the other hand, the minimum critical mass is not so much smaller than the critical mass of curium in metal in the same reflector condition: it is less than a factor of 2. The behavior of ^{247}Cm is similar to ^{232}U and ^{236}Pu , for which nuclides the minimum critical mass in the thermal neutron energy region is only a little larger than the critical mass in metal,²²⁾ which are categorized as “quasi-fissile anomalous nuclides” by E. D. Clayton.²³⁾

The minimum values of the curium mass curves for $k_{\text{eff}}=1$, 0.9 and 0.8, and the corresponding curium concentrations for the spheres of homogeneous $^{243}\text{Cm-H}_2\text{O}$, $^{245}\text{Cm-H}_2\text{O}$ and $^{247}\text{Cm-H}_2\text{O}$ in the three reflector conditions are listed in Table 3. The relation between the minimum critical mass and the corresponding curium concentration is shown in Fig. 8. It is observed from this figure that the minimum critical masses of the three curium isotopes in each reflector condition and the corresponding curium concentrations are well described in a power relation. There seems to be an approximate power

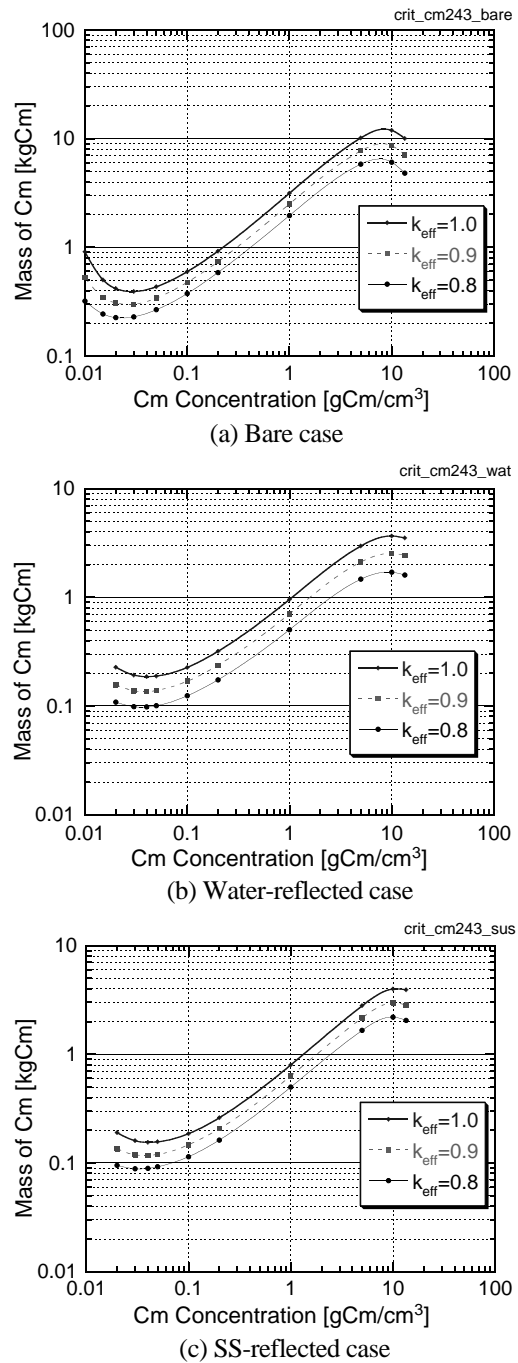


Fig. 5 Critical and subcritical masses of a sphere of homogeneous $^{243}\text{Cm-H}_2\text{O}$

relation among sets of the critical mass of a fissile nuclide and its concentration in a metal-water mixture, which was pointed out earlier by us.¹³⁾

III. Comparison with Calculation Results Based on Other Nuclear Data Files

1. Comparison of the Neutron Multiplication Factor Calculations Based on Various Evaluated Nuclear Data Files

To investigate the effect due to differences in the evaluated nuclear data files, the neutron multiplication factor was cal-

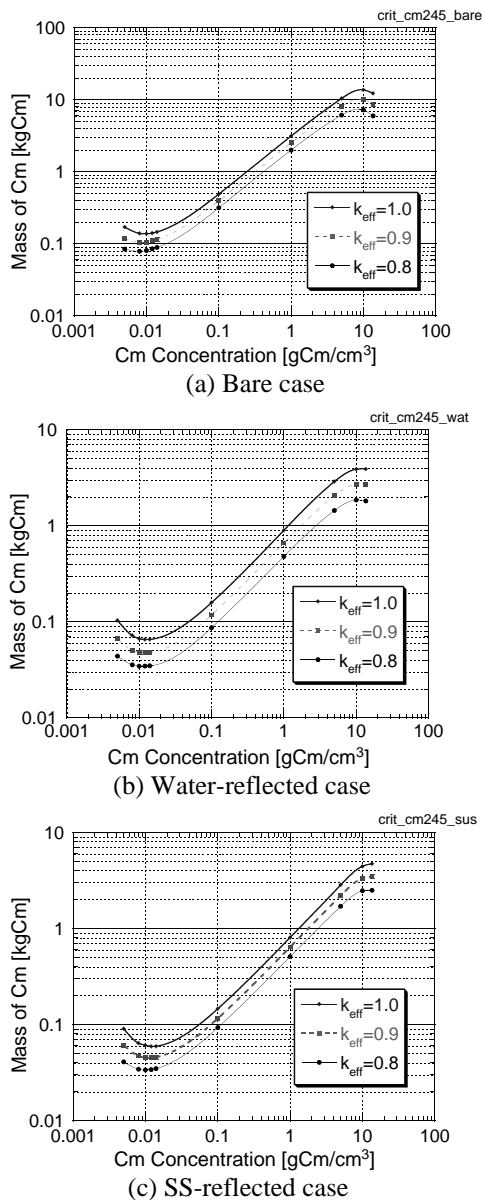


Fig. 6 Critical and subcritical masses of a sphere of homogeneous $^{245}\text{Cm-H}_2\text{O}$

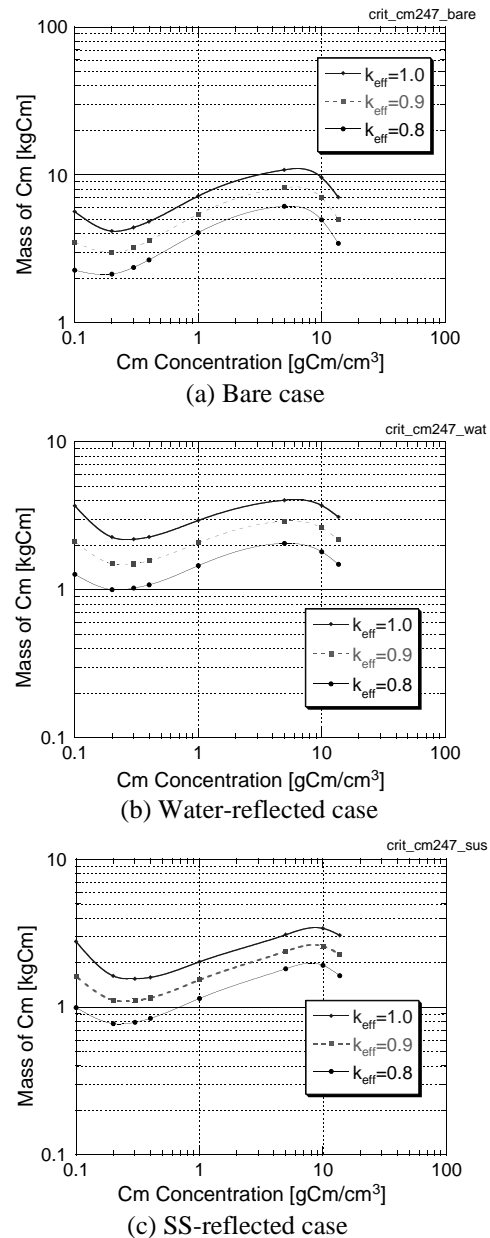


Fig. 7 Critical and subcritical masses of a sphere of homogeneous $^{247}\text{Cm-H}_2\text{O}$

culated using the ENDF/B-VI and JEF-2.2 libraries as well as the JENDL-3.2 library. The calculation was made for (1) the infinite media and (2) a finite bare sphere of curium, both (a) in metal and (b) in a mixture of metal and water (only for fissile isotopes, $^{243,245,247}\text{Cm}$). The radius of the sphere in curium metal was selected to be almost critical. The sphere radius and the curium concentration of the mixture were determined so that the sphere gave nearly the minimum critical mass. The results are shown in **Table 4**.

It is seen from this table that the results based on JENDL-3.2 are generally small compared to the results based on the other libraries with some exceptions. The results also show that the absolute values of differences are typically several percent; especially large differences are found for (1) ^{243}Cm in a metal-water mixture, (2) ^{245}Cm in metal, (3) ^{246}Cm metal in a finite sphere. In the following three sections, the reasons

for the differences are investigated by comparing microscopic reaction cross sections. Differences in the critical masses between JENDL-3.2 and ENDF/B-VI are shown in the last section for ^{245}Cm , ^{246}Cm and ^{247}Cm as examples.

2. Comparison of Microscopic Cross Sections of ^{243}Cm

To clarify the reason behind more than 10% absolute differences in the results of the neutron multiplication factor for ^{243}Cm in infinite media and in a finite sphere of a metal-water mixture (see **Table 4**), the microscopic capture and fission cross sections, and the mean number of neutrons induced per neutron-induced fission (ν -value) are compared in **Fig. 9**. For the well-moderated system of curium concentration 0.03 gCm/cm^3 , the neutron flux has a peak around 50 meV. The ENDF/B-VI library has a 13% larger fission

Table 3 The minimum critical and subcritical masses of curium in homogeneous Cm-H₂O

Nuclide	Reflector	$k_{\text{eff}}=1.0$		$k_{\text{eff}}=0.9$		$k_{\text{eff}}=0.8$	
		Concentration (g/cm ³)	Mass (kg)	Concentration (g/cm ³)	Mass (kg)	Concentration (g/cm ³)	Mass (kg)
²⁴³ Cm	None	0.028	0.391	0.028	0.298	0.022	0.224
	Water	0.041	0.186	0.039	0.135	0.036	0.098
	SS-304	0.041	0.155	0.036	0.117	0.034	0.088
²⁴⁵ Cm	None	0.0094	0.138	0.0087	0.105	0.0081	0.079
	Water	0.0125	0.066	0.0120	0.048	0.0106	0.035
	SS-304	0.0125	0.059	0.0115	0.045	0.0098	0.034
²⁴⁷ Cm	None	0.21	4.15	0.20	2.98	0.172	2.11
	Water	0.26	2.18	0.24	1.47	0.221	1.00
	SS-304	0.27	1.55	0.24	1.09	0.226	0.77

Table 4 Comparison of k_{eff} 's of a bare sphere for five curium isotopes, ²⁴³Cm to ²⁴⁷Cm, based on three evaluated nuclear data libraries: JENDL-3.2, ENDF/B-VI and JEF-2.2

Nuclide	Concentration (g/cm ³)	Radius (cm)	k_{eff}			Relative to JENDL-3.2 $\Delta k_{\text{eff}}/k_{\text{eff}}$ (%)	
			JENDL-3.2	ENDF/B-VI	JEF-2.2	ENDF/B-VI	JEF-2.2
²⁴³ Cm	13.45	6	1.058	1.107	1.158	4.7	9.5
		∞	3.553	3.577	3.653	0.7	2.8
	0.03	15	1.029	1.163	0.895	12.9	-13.0
		∞	2.118	2.335	1.863	10.2	-12.0
²⁴⁴ Cm	13.51	9	1.084	1.113	1.064	2.7	-1.9
		∞	2.605	2.643	2.511	1.5	-3.6
²⁴⁵ Cm	13.57	6.002	0.998	1.086	1.197	8.8	19.9
		∞	3.550	3.687	3.981	3.9	12.1
	0.01	14.887	0.999	1.063	1.116	6.5	11.7
		∞	2.115	2.251	2.343	6.4	10.8
²⁴⁶ Cm	13.62	10.713	0.999	1.150	1.150	15.1	15.1
		∞	2.313	2.395	2.395	3.5	3.5
²⁴⁷ Cm	13.68	4.957	1.001	1.003	1.003	0.2	0.2
		∞	3.924	3.793	3.793	-3.3	-3.3
	0.20	17.062	1.001	1.004	1.004	0.3	0.4
		∞	1.780	1.765	1.766	-0.9	-0.8

cross section and a smaller, almost a half, capture cross section than the JENDL-3.2 library. The two libraries have almost the same ν -values. The JEF-2.2 library has a 30% smaller fission cross section and a 1.3% smaller ν -value with a little smaller capture cross section compared to the JENDL-3.2 library. (As the fission cross section has values about four times larger than the values of the capture cross sections, the difference in the capture cross section has little effect on the absorption cross sections). These comparisons quantitatively explain that the k_{eff} 's of ENDF/B-VI are larger than those of JENDL-3.2, and the k_{eff} 's of JEF-2.2 are smaller than those based on JENDL-3.2 for the homogeneous ²⁴³Cm-H₂O system.

3. Comparison of Microscopic Cross Sections of ²⁴⁵Cm

The neutron spectrum for the critical sphere of a bare ²⁴⁵Cm metal has a peak around 2 MeV.¹²⁾ **Figure 10** compares the neutron capture and fission cross-sections and the ν -value for ²⁴⁵Cm as a function of neutron energy. The ENDF/B-VI and JEF-2.2 libraries give 4.3% and 9.8% larger ν -values, respectively, which almost accounts for the relative differences in k_{∞} from JENDL-3.2, 3.9% and 12.1%, respectively. Their fission cross sections are larger than JENDL-3.2 at 2 MeV, however, their values are smaller than JENDL-3.2 below about 0.7 MeV; therefore, there might be some cancellations. The capture cross sections of ENDF/B-VI and JEF-2.2 at 2 MeV are smaller than and almost as large as that of JENDL-3.2, respectively. The comparisons account for the order of the evaluated nuclear data libraries that produce the neutron multiplication factors for ²⁴⁵Cm metal from larger to

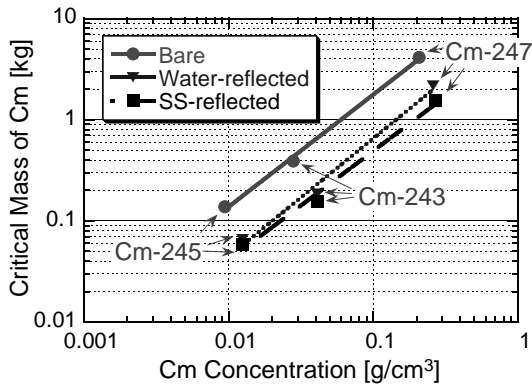


Fig. 8 Relation between the minimum critical mass of the curium isotopes ^{243}Cm , ^{245}Cm and ^{247}Cm and the corresponding curium concentration in homogeneous Cm-H₂O

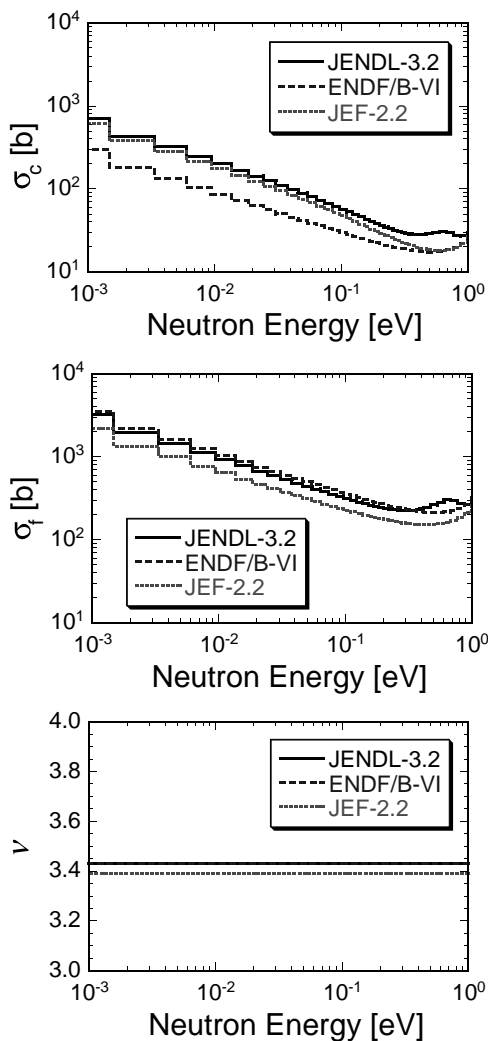


Fig. 9 Comparisons of capture and fission cross sections, and of number of neutrons released by fission for ^{243}Cm

4. Comparison of Microscopic Cross Sections of ^{246}Cm

As shown in Table 4, the neutron multiplication factor of a bare sphere of ^{246}Cm metal that was almost critical according to the calculation based on the JENDL-3.2 library was

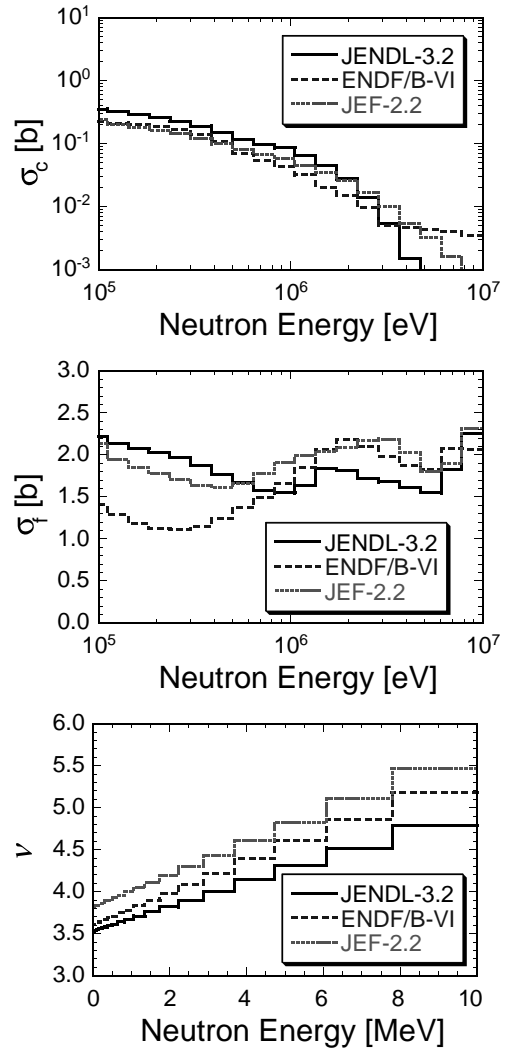


Fig. 10 Comparisons of capture and fission cross sections, and of the average number of neutrons released by fission for ^{245}Cm

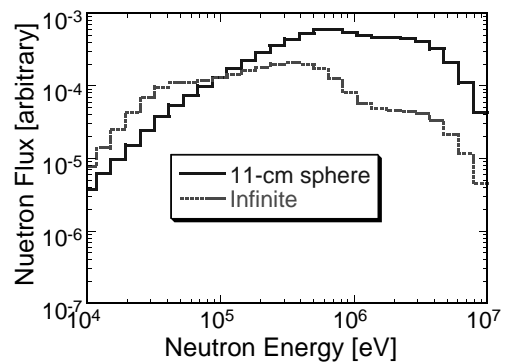


Fig. 11 Neutron spectra of ^{246}Cm metal in an 11-cm bare sphere and in the infinite medium

15% larger than unity if the calculation was based on the ENDF/B-VI library. The relative difference was reduced to 3.5% for the infinite system. (Note that the JEF-2.2 library has the same values as the ENDF/B-VI library for ^{246}Cm).

The neutron energy spectra are compared in **Fig. 11** for the critical bare sphere and for the infinite medium of ^{246}Cm

metal. The neutron energy spectrum for the critical bare sphere has values more than half of the peak value between an energy range from 0.4 to 4 MeV. The neutron energy spectrum shifts to lower energy for the infinite system: the energy having a half-maximum neutron flux ranges between 50 and 600 keV.

Figure 12 compares the neutron capture and fission cross-sections and the ν -value for ^{246}Cm as a function of neutron energy between the JENDL-3.2 and ENDF/B-VI libraries. For the neutron energy ranging from 0.4 to 4 MeV, the ENDF/B-VI library has about 10% larger ν -value and smaller capture cross sections. The fission cross section of ENDF/B-VI is larger than that of JENDL-3.2 for the same energy range. These features explain that the ENDF/B-VI library gives a larger k_{eff} value than the JENDL-3.2 library. For the energy range 50 and 600 keV, where the neutron spectrum of the infinite system has dominant values, the fission cross section of ENDF/B-VI is much smaller than JENDL-3.2, which leads to a large cancellation with the differences in the ν -value.

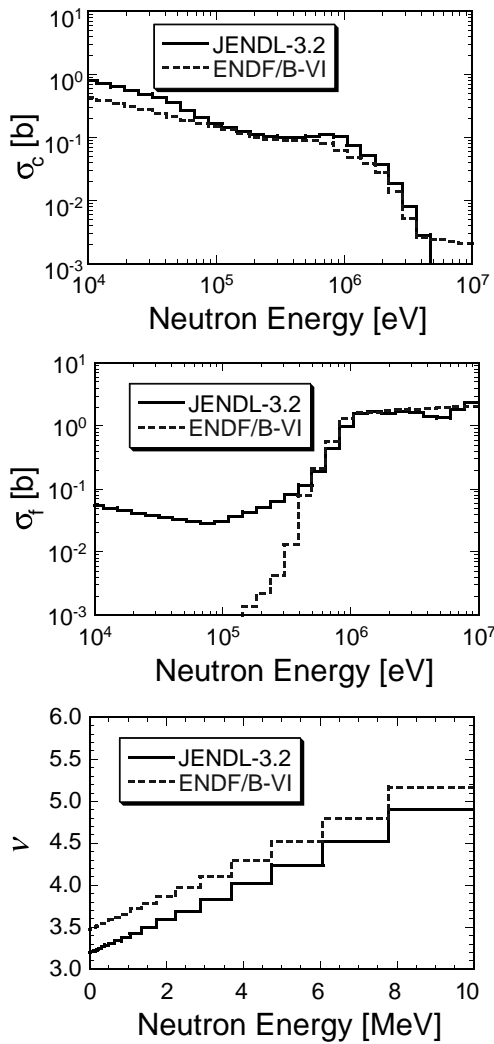


Fig. 12 Comparisons of capture and fission cross sections, and of the average number of neutrons released by fission for ^{246}Cm [Note: The JEF-2.2 library has the identical data to ENDF/B-VI library].

5. Critical Mass Differences for ^{245}Cm , ^{246}Cm and ^{247}Cm

The critical masses of curium isotopes in a metallic form were also calculated using the ENDF/B-VI library instead of the JENDL-3.2 library to see the effect of different nuclear data libraries on the critical mass. **Table 5** summarizes the results. The differences due to different nuclear data libraries are very large for ^{245}Cm and ^{246}Cm , and slight for ^{247}Cm : Replacing JENDL-3.2 with ENDF/B-VI reduces the critical masses to 76%, 56% and 98% of the corresponding masses for ^{245}Cm , ^{246}Cm and ^{247}Cm , respectively. **Figure 13** illustrates the relation between the relative difference in the effective multiplication factor based on ENDF/B-VI from that based on JENDL-3.2, and the ratio of the critical mass based on ENDF/B-VI to that based on JENDL-3.2. The relative difference in the effective multiplication factor of 10% corresponds to about 30% reduction of the critical mass, which was already mentioned in connection with Table 2.

The critical curium masses of a sphere of homogeneous $^{245}\text{Cm-H}_2\text{O}$ without and with a water reflector were also calculated as a function of ^{245}Cm concentration to be seen in **Figs. 14(a)** and **(b)**, respectively, comparing the results based on ENDF/B-VI with the JENDL-3.2 results. These figures show that the ENDF/B-VI results are smaller than the JENDL-3.2 results throughout all the curium concentration ranges. **Table 6** summarizes the calculated results of the minimum critical masses and the corresponding concentrations of curium, which shows that the calculated minimum critical masses based on ENDF/B-VI are about 15% smaller in both conditions with and without a water reflector than those based on JENDL-3.2.

IV. Subcritical Mass Limits

The subcritical limit is defined by the ANSI/ANS-8.1⁽²⁴⁾ standard as follows:

The limiting value assigned to a controlled parameter that results in a subcritical system under specified conditions. The parameter limit allows for uncertainties in the calculations and experimental data used in its derivation but not for contingencies; e.g., double batching or failure of analytical techniques to yield accurate values.

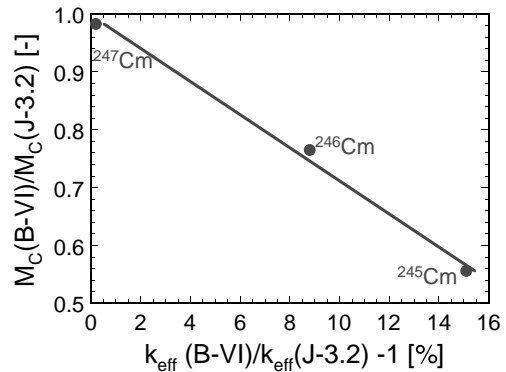


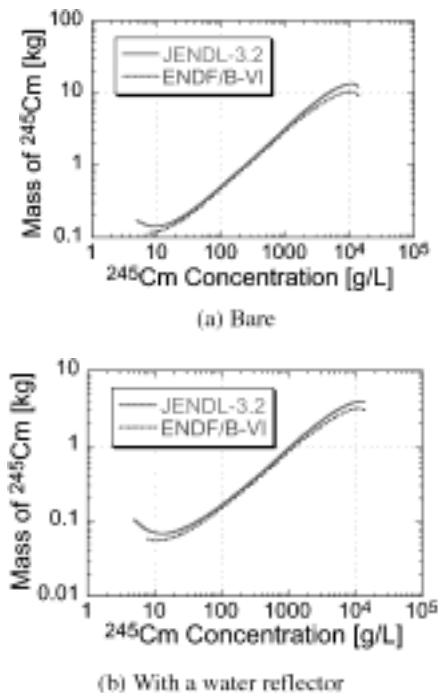
Fig. 13 The relation between the relative difference in the effective multiplication factor based on ENDF/B-VI from that based on JENDL-3.2, and the ratio of the critical mass based on ENDF/B-VI to that based on JENDL-3.2

Table 5 Critical masses of Cm isotopes calculated with different nuclear data libraries

Cm isotopes	Reflector	Critical mass ($k_{\text{eff}}=1$) (kgCm)		M_E/M_J
		M_J (JENDL-3.2)	M_E (ENDF/B-VI)	
^{245}Cm	None	12.3	9.41	0.765
	Water	3.91	3.03	0.774
^{246}Cm	None	70.1	39.0	0.556
^{247}Cm	None	7.06	6.94	0.983

Table 6 Calculated minimum critical masses of homogeneous ^{245}Cm -H₂O based on ENDF/B-VI compared with those with based on JENDL-3.2

Reflector	Nuclear data library	Fuel concentration (gCm/l)	Minimum critical mass (gCm)	Ratio to JENDL-3.2
None	JENDL-3.2	10.0	138	1
	ENDF/B-VI	8.1	117	0.85
Water	JENDL-3.2	12.1	65.6	1
	ENDF/B-VI	11.6	54.9	0.84

**Fig. 14** Calculated critical masses based on the ENDF/B-VI library compared with those based on the JENDL-3.2 library for a sphere of homogeneous ^{245}Cm -H₂O

The standard states the subcritical limit should be based on experiments:

Where applicable data are available, subcritical limits shall be established on bases derived from experiments, with adequate allowance for uncertainties in the data. In the absence of directly applicable experimental measurements, the limits may be derived from calculations made by a method shown by comparison with experimental data to be valid in accordance with 4.3

As already stated in Chap. I, there is no experimental data except one replacement experiment for ^{244}Cm involving curium isotopes. Consequently, we, the ANSI/ANS-8.15 Working Group, made a technical judgment based on calculational studies to decide the subcritical mass limits for the curium isotopes. Like other actinides without experiments, we followed a criterion to decide subcritical mass limit: 50% of the minimum of the critical masses calculationally obtained with various methods. In so doing, we decided the latest version of nuclear library would take priority over the older versions in the same series. Therefore, e.g., if we have both results based on ENDF/B-V and -VI, we selected only the ENDF/B-VI result to find the minimum.

Note that through the comparisons below, half of the critical mass corresponding to $k_{\text{eff}}=1$ was found to be nearly equal to the subcritical mass corresponding to $k_{\text{eff}}=0.8$. This fact might help in discussing about and establishing a margin for subcriticality of the system having no critical experiments.

1. Cm-243

Table 7 gives a comparison of the critical and subcritical masses of ^{243}Cm metal in bare, water-reflected and steel-reflected systems. The present calculation results based on the JENDL-3.2 for bare and water-reflected ^{243}Cm metal spheres, 10.0 and 3.51 kg, are close to Komuro's values,⁷⁾ 9.72 and 3.35 kg, respectively, based on the JENDL-3.0 library. Anno's results⁹⁾ based on JEF-2.2 gave much smaller values, which is consistent with the fact that JEF-2.2 gave larger k_{eff} and k_{∞} values compared to JENDL-3.2 (see Table 4). Although there are no ENDF's results, they are expected to be between these two (see again Table 4). The subcritical mass limits were decided essentially based on the Anno's results that gave the minimum of the calculated critical masses. These subcritical mass limits are new, as the previous ANSI/ANS-8.15 standard did not supply subcritical mass limits for ^{243}Cm metal.

Table 7 Comparison of critical and subcritical masses of ^{243}Cm in metal system, and the subcritical mass limits accepted by ANSI/ANS-8.15 Working Group (kg)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
10.0	3.51	3.91	1	JENDL-3.2	MCNP4B2	13)
7.08	2.43	2.84	0.9	JENDL-3.2	MCNP4B2	13)
4.80	1.60	2.05	0.8	JENDL-3.2	MCNP4B2	13)
9.72	3.35	—	1	JENDL-3.0	MULTI-KENO-3.0	7)
8.10	—	3.28	1	ENDL-82	DTF-IV	4)
7.34	2.83	2.76	1	JEF-2.2	APOLLO-II	9)
—	—	—	ANSI/ANS8.15-1981			1)
3.7	1.4	1.4	Accepted subcritical mass limits			

Table 8 Comparison of the minimum critical and subcritical masses of ^{243}Cm in water mixture systems, and the subcritical mass limits accepted by ANSI/ANS-8.15 Working Group (g)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
391	186	155	1	JENDL-3.2	MCNP4B2	13)
298	135	117	0.9	JENDL-3.2	MCNP4B2	13)
224	98	88	0.8	JENDL-3.2	MCNP4B2	13)
739	382	—	1	ENDL-82	DTF-IV	4)
—	116	—	1	CEA86 ^{a)}	DTF-IV	5)
—	265	—	1	JEF-2.2	APOLLO-II	25)
—	90	—	ANSI/ANS8.15-1981			1)
190	90	80	Accepted subcritical mass limits			

^{a)} The cross sections were first thought to be based on the JEF-1 library, however, it was recently revealed that they were from ENDF/B-V.²⁵⁾

The minimum critical masses of homogeneous $^{243}\text{Cm-H}_2\text{O}$ from various information sources are shown in **Table 8**. French results are widely ranged: from 116 to 382 g. We adopted the latest value 265 g based on JEF-2.2 as the representative one of the three French results.^{4,5,25)} This value is larger than the JENDL's result, as expected from Table 4. The subcritical mass limits for the homogeneous $^{243}\text{Cm-H}_2\text{O}$ in bare, water-reflected and steel-reflected systems, were decided as a half of the JENDL's results: 190, 90 and 80 g, respectively. [Note: As shown in the footnote of Table 8, the cross sections of CEA86 were considered to be originated from the JEF-1 library at the ANSI/ANS-8.15 Working Group meeting in November 2001. However, it was recently revealed that they were from ENDF/B-V. The authors would propose the members of the Group to make a calculation based on the ENDF/B-VI file. The subcritical mass limits for homogeneous $^{243}\text{Cm-H}_2\text{O}$ should be smaller if there were results based on ENDF/B-VI. See Table 4].

2. Cm-244

The critical and subcritical masses of ^{244}Cm metal calculated in this study are shown in **Table 9** in comparison with those obtained according to other studies. A wide range of values was found at a first look: for example, 13.2 to 33.0 kg for the critical curium mass in the bare condition. However, it can be pointed out that the old values are smaller; the recently obtained values are comparatively larger and in a small range:

27–33 kg. The ANSI/ANS-8.15 Working Group decided the subcritical mass limit as 14 kg for a bare metal of ^{244}Cm based on the minimum of these values, 27 kg. Considering the reflector effects, the revised values of 11 and 7 kg, respectively for water- and steel-reflected metal of ^{244}Cm were decided, which became more than twice of the previously recommended values, 5 and 3 kg, respectively.

3. Cm-245

Table 10 lists the critical and subcritical masses of ^{245}Cm metal in bare, water-reflected and steel reflected systems. The present calculation result based on the JENDL-3.2 library for a bare ^{245}Cm metal sphere, 12.3 kg, is very close to Nojiri's result based on the same code-library combination, 12.4 kg;⁸⁾ it agrees well with the results based on JENDL-3, ENDL-82 and -85. However, Anno's result based on JEF-2.2, 6.81 kg,⁹⁾ was much smaller. Another result based on the ENDF/B-V has a comparable critical mass value, 9.33 kg,⁸⁾ to our ENDF/B-VI result, 9.41 kg. Similar tendencies were found for the results with a water reflector and with a steel reflector. The subcritical mass limits were decided based on Anno's results that gave the minimum. These subcritical mass limits are new in this revision.

The minimum critical masses of homogeneous $^{245}\text{Cm-H}_2\text{O}$ from various information sources are shown in **Table 11**. The present result for the bare case based on the JENDL-3.2 library, 138 g, is almost equal to that of Srinivasan's, 136 g;⁴⁾

Table 9 Comparison of critical and subcritical masses of ^{244}Cm in metal systems, and the subcritical mass limits accepted by the ANSI/ANS-8.15 Working Group (kg)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
30.0	24.8	15.4	1	JENDL-3.2	MCNP4B2	13)
20.1	16.5	10.6	0.9	JENDL-3.2	MCNP4B2	13)
13.2	10.7	7.1	0.8	JENDL-3.2	MCNP4B2	13)
27.6	22.8	—	1	JENDL-3.0	MULTI-KENO-3.0	7)
21.2	10.6	—	1	ENDL-82	DTF-IV	4)
33.0	26.9	16.0	1	JEF-2.2	APOLLO-II	9)
13.5	11.5	7.6	1	ENDF/B-IV	XSDRNPM	11)
27	—	—	1	ENDF/B-VI		14)
—	5	3	ANSI/ANS8.15-1981			1)
14	11	7	Accepted subcritical mass limits			

Table 10 Comparison of critical and subcritical masses of ^{245}Cm in metal systems, and the subcritical mass limits accepted by the ANSI/ANS-8.15 Working Group (kg)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
12.3	3.91	4.73	1	JENDL-3.2	MCNP4A	12)
8.71	2.71	3.49	0.9	JENDL-3.2	MCNP4A	12)
5.99	1.82	2.51	0.8	JENDL-3.2	MCNP4A	12)
9.41	3.03	—	1	ENDF/B-VI	MCNP4A	12)
12.4	3.78	—	1	JENDL-3.0	MULTI-KENO-3.0	7)
9.33	3.14	3.59	1	ENDF/B-V	SCALE4.3	8)
12.5	3.86	4.80	1	ENDL-85	MCNP4A	8)
12.4	3.59	4.77	1	JENDL-3.2	MCNP4A	8)
12.3	—	5.80	1	ENDL-82	DTF-IV	4)
6.81	2.61	2.66	1	JEF-2.2	APOLLO-II	9)
—	—	—	ANSI/ANS8.15-1981			1)
3.4	1.3	1.3	Accepted subcritical mass limits			

Table 11 Comparison of the minimum critical and subcritical masses of ^{245}Cm in water mixture systems, and the subcritical mass limits accepted by the ANSI/ANS-8.15 Working Group (g)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
138	66	59	1	JENDL-3.2	MCNP4A	12)
105	48	45	0.9	JENDL-3.2	MCNP4A	12)
79	35	34	0.8	JENDL-3.2	MCNP4A	12)
117	54.9	—	1	ENDF/B-VI	MCNP4A	12)
89.2	40.5	—	0.9	ENDF/B-VI	MCNP4A	12)
136	62	—	1	ENDL-82	DTF-IV	4)
—	42.7	—	1	JEF-1	DTF-IV	5)
—	46.5	—	1	JEF-2.2	APOLLO-II	25)
—	30	—	ANSI/ANS8.15-1981			1)
58	23	—	Accepted subcritical mass limits			

however, the present value for the water-reflected case, 66 g, is a little larger than the corresponding value, 62 g.⁴⁾ Rossignol gave a considerably smaller value, 42.7 g.⁵⁾ Recently, IPSN supplied 46.5 g²⁵⁾ as the minimum critical mass based on JEF-2.2, on which the subcritical mass limit of $^{245}\text{Cm-H}_2\text{O}$ with a water reflector, 23 g, was decided. The subcritical mass limit for the bare $^{245}\text{Cm-H}_2\text{O}$ system, 58 g, was based on our result based on ENDF/B-VI, 117 g.¹²⁾ As there were no steel-

reflected data in November 2001, when the Working Group meeting was held, the Group did not recommend any subcritical mass limit for steel-reflected $^{245}\text{Cm-H}_2\text{O}$ system.

4. Cm-246

Table 12 compares the critical masses of ^{246}Cm metal in bare, water-reflected and steel-reflected conditions. The critical masses obtained as a result of the present calculations

Table 12 Comparison of critical and subcritical masses of ^{246}Cm in metal systems, and the subcritical mass limits accepted by the ANSI/ANS-8.15 Working Group (kg)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
70.1	57.4	38.1	1	JENDL-3.2	MCNP4A	12)
45.5	37.0	25.2	0.9	JENDL-3.2	MCNP4A	12)
28.9	23.5	16.2	0.8	JENDL-3.2	MCNP4A	12)
39.0	—	—	1	ENDF/B-VI	MCNP4A	12)
40.96	—	—	1	JEF-2.2	APOLLO-II	25)
37.9	32.9	20.5	1	ENDF/B-V	SCALE4.3	8)
84.1	65.8	44.7	1	ENDL-85	MCNP4A	8)
70.0	58.6	38.7	1	JENDL-3.2	MCNP4A	8)
—	—	—	ANSI/ANS8.15-1981			1)
19	16	10	Accepted subcritical mass limits			

Table 13 Comparison of critical and subcritical masses of ^{247}Cm in metal systems, and the subcritical mass limits accepted by the ANSI/ANS-8.15 Working Group (kg)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
7.06	3.10	3.07	1	JENDL-3.2	MCNP4A	12)
5.02	2.18	2.28	0.9	JENDL-3.2	MCNP4A	12)
3.44	1.49	1.63	0.8	JENDL-3.2	MCNP4A	12)
6.94	—	—	1	ENDF/B-VI	MCNP4A	12)
7.00	2.91	—	1	JENDL-3.0	MULTI-KENO-3.0	7)
7.15	3.66	2.94	1	ENDF/B-V	SCALE4.3	8)
7.88	3.71	3.42	1	ENDL-85	MCNP4A	8)
7.25	3.01	3.15	1	JENDL-3.2	MCNP4A	8)
7.87	—	3.36	1	ENDL-82	DTF-IV	4)
7.21	—	—	1	JEF-2.2	APOLLO-II	9)
—	—	—	ANSI/ANS8.15-1981			1)
3.5	1.5	1.4	Accepted subcritical mass limits			

based on the JENDL-3.2 library are almost equal to Nojiri's results⁸⁾ based on the same library, considering the errors associated with the Monte Carlo calculations. The JENDL-3.2 results are about 80% larger than those based on the ENDF/B-V or -VI libraries.^{8,12)} Those calculation results give 15 to 20% smaller values than those based on the ENDL-85 library.⁸⁾ The critical mass of a bare sphere of ^{246}Cm metal based on the JEF-2.2 library, 40.96 kg,²⁵⁾ was 5% larger than the corresponding mass, 39.0 kg,¹²⁾ based on the ENDF/B-VI library. As the original nuclear data are the same, the small difference may have been originated from the difference in the densities and/or the criticality calculation codes. The subcritical mass limits in bare, water-reflected and steel reflected conditions were decided, therefore, based on the ENDF/B-V and -VI results to be 19, 16 and 10 kg, respectively.

There was no description on subcritical mass limits for ^{246}Cm in the ANSI/ANS-8.15-1981; therefore these are new recommendations.

5. Cm-247

Table 13 gives a list of the critical masses of ^{247}Cm metal in bare, with a water-reflector, and with a steel-reflector. In comparison with the other four nuclides mentioned above, the differences among the results on ^{247}Cm are small: *e.g.*, the

critical ^{247}Cm mass in bare condition are in a band from 6.9 to 7.9. Looking more carefully at **Table 13**, however, we would notice that ENDLs' results^{4,8)} are larger than the results based on other libraries, *i.e.*, JENDLs, ENDF/Bs and JEF-2.2.

The minimum critical masses of homogeneous ^{247}Cm -H₂O are compared in **Table 14**. The critical mass of IPSN, 2.16 kg,²⁵⁾ which gives the minimum for the water-reflected case, is very close to the present result, 2.18 kg. The corresponding value by Srinivasan⁴⁾ is about 26% larger than these values.

In deciding the subcritical mass limits, *e.g.* 1.0 kg for the water-reflected case, 50% mass rule was applied, however in a rather conservative way, considering there were no results based on ENDF/Bs.

6. Summary of Subcritical Mass Limits

Table 15 summarizes the subcritical mass limits for curium metal, on which the ANSI/ANS-8.15 Working Group reached a consensus at the meeting held in Los Alamos in November 2001. Subcritical mass limits agreed at the same time for homogeneous Cm-H₂O mixtures are listed in **Table 16**. A draft of revision for the ANSI/ANS-8.15 is going to proceed with voting by the ANSI/ANS-8 members in late 2002.

Table 14 Comparison of the minimum critical and subcritical masses of ^{247}Cm in water mixture system, and the subcritical mass limits accepted by the ANSI/ANS-8.15 Working Group (kg)

Bare	Water-reflected	Steel-reflected	k_{eff}	Nuclear Data Library	Computer code	Ref.
4.15	2.18	1.55	1	JENDL-3.2	MCNP4A	12)
2.98	1.47	1.09	0.9	JENDL-3.2	MCNP4A	12)
2.11	1.00	0.77	0.8	JENDL-3.2	MCNP4A	12)
5.048	2.728	—	1	ENDL-82	DTF-IV	4)
—	2.160	—	1	JEF-2.2	APOLLO-II	25)
—	0.9	—	ANSI/ANS8.15-1981			1)
2.1	1.0	0.7	Accepted subcritical mass limits			

Table 15 The subcritical mass limits for curium metal accepted by the ANSI/ANS-8.15 Working Group in November 2001 (kg)

Nuclide	Bare	Water-reflected	Steel-reflected
^{243}Cm	3.7	1.4	1.4
^{244}Cm	14	11	7
^{245}Cm	3.4	1.3	1.3
^{246}Cm	19	16	10
^{247}Cm	3.5	1.5	1.4

Table 16 The subcritical mass limits for homogeneous Cm-H₂O mixtures accepted by the ANSI/ANS-8.15 Working Group in November 2001 (kg)

Nuclide	Bare	Water-reflected	Steel-reflected
^{243}Cm	0.19	0.09	0.08
^{245}Cm	0.058	0.023	Not applicable
^{247}Cm	2.1	1.0	0.7

V. Conclusions

The following conclusions were reached through this study.

- (1) Critical ($k_{\text{eff}}=1$) and subcritical ($k_{\text{eff}}=0.9$ and 0.8) masses were calculated for a sphere of five curium isotopes, ^{243}Cm to ^{247}Cm , in metal and three curium isotopes, ^{243}Cm , ^{245}Cm and ^{247}Cm , in metal-water mixtures, considering three reflector conditions: bare, with a water or stainless steel reflector. Systematic relations were mentioned (a) between the critical mass of curium metal and k_{∞} , and (b) between the minimum critical mass of curium in homogeneous Cm-H₂O and the corresponding curium concentration.
- (2) Remarkable relative differences, $10\% \Delta k/k$ or more, were found in the neutron multiplication factor calculations based on different nuclear data files, JENDL-3.2, ENDF/B-VI and JEF-2.2, especially for (a) ^{243}Cm in a metal-water mixture, (b) ^{245}Cm in metal, and (c) ^{246}Cm metal in a finite sphere. The comparisons of ν -values, and fission and capture cross sections explained tendencies of the differences.
- (3) The present study supplied basic information to the ANSI/ANS-8.15 Working Group for revision of the standard for nuclear criticality control of special actinide el-

ements. Based on the present study, information from members of the Group and past literature, the new or revised values for the subcritical mass limits of curium isotopes were proposed, and the consensus values of the ANSI/ANS-8.15 Working Group were obtained.

Acknowledgments

The authors are grateful to all the members of the Working Group of ANSI/ANS-8.15 for discussing and deciding the subcritical mass limits for curium isotopes. Special thanks are due to the chair of the Group, N. L. Pruvost for having assigned one of the authors (HO) to take charge of the curium isotopes, and to J. Anno and R. M. Westfall for showing the group members their results. Contributions from domestic collaborators are also praised: I. Nojiri and Y. Fukasaku supplied the authors with their both published and unpublished results; and Y. Komuro provided the authors with his results and also useful information.

A part of this work was carried out by the Japan Atomic Energy Research Institute (JAERI) under entrustment by the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan.

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