JAERI-Research
2000-040

CRITICAL AND SUBCRITICAL MASSES OF CURIUM-245, -246 AND -247 CALCULATED WITH A COMBINATION OF MCNP4A CODE AND JENDL-3.2 LIBRARY

September 2000

Hiroshi OKUNO and Hiromitsu KAWASAKI*

日本原子力研究所
Japan Atomic Energy Research Institute

編集兼発行　日本原子力研究所

JAERI-Research 2000-040

Critical and Subcritical Masses of Curium-245, -246 and -247 Calculated with a Combination of MCNP4A Code and JENDL-3.2 Library

Hiroshi OKUNO and Hiromitsu KAWASAKI*

Department of Fuel Cycle Safety Research
Nuclear Safety Research Center
Tokai Research Establishment
Japan Atomic Energy Research Institute
Tokai-mura, Naka-gun, Ibaraki-ken

(Received August 4, 2000)

Critical masses of three curium isotopes, ^{245}Cm, ^{246}Cm and ^{247}Cm, were calculated with a combination of the current version of the Japanese Evaluated Nuclear Data Library, JENDL-3.2, and a continuous energy Monte Carlo neutron transport code, MCNP4A. The subcritical masses corresponding to the neutron multiplication factor $k_{eff} = 0.9$ and 0.8 were also computed in the same way. The subcritical masses that correspond to $k_{eff} = 0.9$ for ^{246}Cm metal and ^{246}CmO$_2$ with a 30-cm-thick stainless steel reflector were computed as 25.2 kg and 41.8 kg, respectively. The minimum critical mass for ^{245}Cm was obtained as 65.6 g in a sphere of a homogeneous mixture of granulated ^{245}Cm metal and water surrounded by a fully thick water reflector. The corresponding quantity for ^{247}Cm was found to be 2.19 kg. The critical masses of ^{245}Cm, ^{246}Cm and ^{247}Cm metals were computed also for reference by replacing the JENDL-3.2 with the ENDF/B-VI; they were reduced by 23 %, 45 % and 2 %, respectively, from each corresponding value, which revealed a large dependence of the results on the evaluated nuclear data libraries. The present report was prepared for revision of the ANSI/ANS-8.15, *the American National Standard for Nuclear Criticality Control of Special Actinide Elements.*

Keywords: Critical Mass, Subcritical Mass, Curium-245, Curium-246, Curium-247, JENDL-3.2, MCNP4A, ENDF/B-VI, Minor Actinides, ANSI/ANS-8.15

This work was carried out by the Japan Atomic Energy Research Institute (JAERI) under entrustment by the Science and Technology Agency (STA) of Japan.

* CRC Research Institute, Inc.

JAERI-Research 2000-040

MCNP4A コード及び JENDL-3.2 ライブラリを用いたキュリウム-245、-246 及び-247 の臨界及び未臨界質量の計算

日本原子力研究所東海研究所安全性試験研究センター燃料サイクル安全工学部
奥野　浩・川崎　弘光*

（2000 年 8 月 4 日受理）

キュリウム同位体 3 核種（^{245}Cm、^{246}Cm 及び ^{247}Cm）の臨界質量を日本の評価済核データライブラリの JENDL-3.2 と連続エネルギーモンテカルロ中性子輸送計算コード MCNP4A とを用いて計算した。中性子増倍率 k_{eff} = 0.9 及び 0.8 に対応する質量も同様な方法で算出した。30cm 厚さのステンレス鋼を反射体とする ^{246}Cm 金属及び ^{246}CmO$_2$ の未臨界質量（k_{eff} = 0.9 に対応）は、それぞれ 25.2 kg 及び 41.8 kg と計算された。^{245}Cm の最小臨界質量として、微粒状 ^{245}Cm 金属と水の均質混合球状体系で十分な厚さの水反射体に囲まれた場合に 65.6 g との結果を得た。^{247}Cm の対応量は 2.19 kg と求められた。参考までに ^{245}Cm、^{246}Cm 及び ^{247}Cm の裸の金属体系で、評価済核データライブラリを JENDL-3.2 から ENDF/B-VI に置き換えて臨界質量を計算したところ、対応量はそれぞれ 23％、45％及び 2％だけ小さくなり、核データライブラリの依存性が大きいことが分かった。本報告書は、米国原子力学会基準 ANSI/ANS-8.15（特別なアクチニド核種の臨界管理）改訂のため準備した。

この報告書は、電源開発促進対策特別会計法に基づく科学技術庁からの受託として行った研究成果である。
東海研究所：〒319-1195　茨城県那珂郡東海村白方白根 2-4

*　㈱CRC 総合研究所

Contents

目次

This is a blank page.

1. Introduction

The American National Standard for Nuclear Criticality Control of Special Actinide Elements, ANSI/ANS-8.15-1981,(1) has been in a revising process by a work group chaired by Norman L. Pruvost since 1996,(2) which consisted of six US members and four non-US members. One of the authors of this report (HO) is the Japanese member of the group, who has been assigned three curium isotopes: ^{246}Cm for new inclusion, and ^{245}Cm and ^{247}Cm for revision.

Practical importance of critical and/or subcritical masses of curium isotopes is at least related to the revision of the IAEA's transport regulations (hereafter referred to as ST-1). (3) France proposed; (1) to adopt minor actinides such as ^{242}Am, ^{245}Cm and ^{251}Cf, as fissile materials, which are now limited only to the four nuclides ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu, and their mixtures, as prescribed in the Item 222 of ST-1; (2) to include minor actinides in the excepted fissile material from the requirements to be transported in packages stated in the Item 672 of ST-1.(4), (5)

In this aspect, JAERI-Tokai, e.g., has transported minor actinides three times for the past 7 years, the amount of which is less than 1 g, therefore there has been no fear of criticality. In the future, however, a larger amount of minor actinides may be treated and transported.

Curium isotopes are generated from ^{238}U in light-water reactors by several neutron captures and four gamma decays. Their amounts are negligibly small in an initial stage of burnup, however, increase remarkably as burnup proceeds. Some of the isotopes, ^{243}Cm, ^{245}Cm and ^{247}Cm, have half lives more than 20 years and are proton-even and neutron-odd like as the big three fissile nuclides, ^{233}U, ^{235}U and ^{239}Pu. Therefore, the criticality aspects of curium isotopes are important, next to the big three mentioned above and ^{241}Pu, from the standpoint of both safety and safeguards.

There have been no critical experiments and a very limited number of reactivity-worth experiments involving minor actinides.(6), (7) Therefore, we can almost only rely on calculation for the critical masses of these nuclides. There are several documents so far that treated criticality data on or subcritical limits of curium isotopes.(8)-(14) However, none of the documents covers the whole.

The purpose of this report is to obtain critical and subcritical masses of the curium isotopes as precisely as possible using modern computation tools based on recently published nuclear data for standardizing these quantities in reference to the previous data. The critical mass is defined as the mass corresponding to the neutron multiplication factor, $k_{eff} = 1$. The subcritical masses to be obtained in this report are limited to the masses corresponding to $k_{eff} = 0.9$ and 0.8. This selection of the subcritical masses was decided in referring to Ref. (15).

Chapter 2 of this report describes calculation methods and results. Chapter 3 devotes comparisons with the corresponding data obtained based on another nuclear data library than the present calculations, and also with published data. The last Chapter concludes the report. Appendix A summarizes the formula and numbers adopted in calculating the atomic number densities for Cm-H_2O and CmO_2-H_2O. Appendix B presents the ^{244}Cm effect results on the minimum critical mass and the minimum subcritical masses of ^{245}Cm, and discusses the validity of the linear relation shown in the ANSI/ANS-8.15-1981. Appendix C briefly reviews the critical mass studies of curium isotopes. Appendix D collects graphs comparing evaluated nuclear data for various cross sections and for averaged number of neutrons produced by a neutron-induced fission of the curium isotopes.

2. Calculation of Critical and Subcritical Masses

2.1. Method of calculation

The neutron transport code we used for criticality calculation was a continuous energy Monte Carlo code MCNP4A(16) developed at the Los Alamos National Laboratory of the USA. The MCNP code impressed us about its popularity in the field of nuclear criticality safety at *the Topical Meeting on Physics and Methods in Criticality Safety* held at Nashville, TN, in 1993.(17) The ambiguity due to the statistical method has been lessened by a larger number of histories from typically 30 thousands in 1980's to an order of millions nowadays adopted in the calculations.

The number of tallied source histories of the MCNP4A calculation was set to a half million in our calculation. More precise information on our specification for the input is shown in Table 1.

The evaluated nuclear data library applied was the JENDL-3.2,(18) the current version of the Japanese Evaluated Nuclear Data Library. The number of available curium isotopes in the JENDL-3.2 were ten (from ^{241}Cm to ^{250}Cm), whereas six (from ^{243}Cm to ^{248}Cm) in the ENDF/B-VI.(19) Comparison of the critical masses obtained based on the JENDL-3.2 with those on the ENDF/B-VI will be discussed in Section 3.1.

As mentioned in Chapter 1, we call in this report the mass corresponding to $k_{eff} = 1$ calculated with the computation system the critical mass. By this definition, the critical mass depends on the nuclear data library and/or other data assumed in the calculation and computation methods employed.

The estimated critical mass would be determined by obtaining the estimated critical neutron multiplication factor that could be derived by benchmark calculations of similar critical experiments, which were, however, not available for curium isotopes. Therefore, masses corresponding to $k_{eff} = 0.9$ and 0.8, which we call subcritical masses, of curium isotopes (^{245}Cm, ^{246}Cm and ^{247}Cm) were calculated as well as the critical mass. These masses were assumed both in metallic and in dioxide forms. The atomic number densities of Cm, CmO_2 and their water mixtures were obtained according to the formulae and numbers described in Appendix A. The reflector conditions are summarized in Table 2. For both of water and stainless-steel (SUS) reflectors, the thickness was assumed to be 30 cm. The kind of SUS we assumed in our calculation was Type 304, the composition of which is shown in Table 3.(20)

For determining the spherical radius of fuel, we adopted the algebraic form proposed by Rombough et al.,(21) which was adopted in "Nuclear Criticality Safety Guide."(15) Namely, to determine the value of the sphere radius r for a particular value of k_{eff}, an appropriate set of calculational results, at least 4 points, were fitted to a continuous curve having the following algebraic form:

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

where k_∞ is the neutron multiplication factor of water mixture of fuel in infinite media, and α, β and γ are parameters.

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

$$M = \rho \cdot 4\pi r^3/3,$$

where ρ is the mass density of curium. It is worth pointing out that we took into account the difference in the density ρ by the mass numbers of isotopes.

Note in Table 2 that the critical/subcritical masses for ^{246}Cm were calculated only at the theoretical density, because the fission cross section dominates absorption cross section only above hundreds keV of the neutron energy (see Appendix D and Ref. (22)). For ^{245}Cm and ^{247}Cm, however, enhanced moderation with water decreases the critical mass of the curium isotopes due to higher fission cross sections at low energies. Therefore, for these nuclides curium concentration was changed in the calculations.

2.2. Results

The relation between the radius of a fuel sphere and k_{eff} is shown in Fig.1 for ^{245}Cm without reflectors. This figure explains how to obtain the fuel radii corresponding to $k_{eff} = 1$, 0.9 and 0.8. They are 6.00, 5.35 and 4.72 cm, respectively. The critical mass was derived as 12.3 kgCm, and subcritical masses corresponding to $k_{eff} = 0.9$ and 0.8 were calculated as 8.71 and 5.99 kgCm, respectively, by multiplying the fuel density 13.57 gCm/cm^3 to the corresponding volumes.

The critical and subcritical masses of ^{245}Cm metal with a water reflector, and ^{245}Cm dioxide with/without a water reflector were similarly obtained, and all the results are summarized in Table 4. The critical/subcritical masses of fuel without reflectors are only one-third of the corresponding masses of fuel with a water reflector. The effect of the reflector is remarkably larger than for typical fission nuclides, ^{235}U and ^{239}Pu: the critical masses ^{235}U and ^{239}Pu in a bare metallic sphere are 49.0 kg and 10.0 kg, respectively, which are reduced to 22.8 kg and 5.42 kg, respectively, with a fully-thick water reflector. (23) Therefore, the water reflector makes the critical mass almost a half of the corresponding mass of the bare fuel in these two popular nuclides.

Tables 5 and 6 show similar amounts for ^{246}Cm and ^{247}Cm. Note, however, that an SUS reflector was assumed for ^{246}Cm fuel instead of water reflector, because the former reflector was known to act more reactively than the latter reflector (see Table C7 of Appendix C).(17) For ^{247}Cm fuel, both reflectors have comparative effects; therefore both calculations were made attaching either an SUS or a water reflector (see Table 6).

For ^{245}Cm, critical and subcritical masses of fuel in a sphere were calculated with changing ^{245}Cm concentration in its water mixture. Figures 2 and 3 show the results for two reflector conditions, with and without a water reflector, respectively. Table 7 summarizes the minimum critical and subcritical masses with and without a water reflector. This table shows, e.g., that the minimum ^{245}Cm mass was calculated as 138 g at the Cm concentration of 10.0 gCm/L (= 0.01 gCm/cm^3) for a bare sphere of its water mixture.

Quite similarly to ^{245}Cm, critical and subcritical masses of fuel in a sphere were calculated with a change in ^{247}Cm concentration in its water mixture. The only difference is that these masses were obtained for ^{247}Cm in three reflector conditions, i.e., with water reflector, with SUS reflector and without reflector, instead of the two reflector conditions as for ^{245}Cm. Figures 4 through 6 show overall results, and Table 8 summarizes the relevant minimum critical and subcritical masses. It is found out from this table that SUS acts more reactively than water in the aspect of the minimum critical/subcritical masses of ^{247}Cm in its mixture with water. However, this is not true for the ^{245}Cm metal; as we have seen in Table 6, SUS acts comparatively equal to or less reactively than water.

3. Discussions

3.1. Comparison with the results based on ENDF/B-VI

The critical masses of Cm isotopes in metallic form were calculated using the ENDF/B-

VI library instead of the JENDL-3.2 library to see the effect of different nuclear data libraries. Table 9 shows the results. The differences due to 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.

The critical curium masses of a sphere of ^{245}Cm-H_2O without and with a water reflector were also calculated as a function of ^{245}Cm concentration to be seen in Figs. 7 and 8, respectively, comparing the results 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 fuel concentration ranges. Table 10 summarizes the calculated results of the minimum critical masses and the corresponding concentrations of curium.

Qualitative explanation of the differences in the critical masses and the minimum critical mass can be made through comparison of ν-value, fission and capture cross sections, as follows.

Figures 9 to 11 compare the mean number of neutrons produced by a neutron-induced fission (ν-value) for the three curium isotopes as a function of neutron energy. Figures 9 and 10 explain that the ENDF/B-VI gives larger k_{eff} from the aspect of ν-value compared to the JENDL-3.2 for ^{245}Cm and ^{246}Cm; Figure 11 implies the tendency is opposite for ^{247}Cm. Table 11 compares the ν-value at the neutron energy of 2 MeV. It explicates numerically the above statement: ν-value of ENDF/B-VI is 4.3% and 7.8 % larger for ^{245}Cm and ^{246}Cm, respectively, than the corresponding values at 2 MeV than JENDL-3.2, whereas 5.4 % smaller for 247Cm.

The neutron spectra for the critical spheres of bare curium metals of 245Cm, 246Cm and 247Cm have a peak from 1 to 3 MeV (See see Fig.ure 12). This fact implies that that the differences in keff mainly characterized by the _-value as mentioned above are additionally modified by the behaviors of cross section data around that energy region.

Figures 13 and 14 compare the neutron-induced fission and neutron capture cross-sections, respectively, for ^{245}Cm between JENDL-3.2 and ENDF/B-VI. As the fission cross section of ENDF/B-VI is larger than that of JENDL-3.2, and the neutron capture cross section has an opposite tendency, the differences in the cross sections raise the k_{eff} based on the ENDF/B-VI than based on JENDL-3.2. This enforces the tendency originated from the differences in the ν-value. Similar statements should be said for ^{246}Cm from Figs. 15 and 16. Comparison of cross sections shows a similar tendency for ^{247}Cm, which compensates the differences in the ν-value (see Fig. 11).

As shown in Figs. 7 and 8, and also in Table 10, the minimum critical masses for a homogeneous mixture of granulated ^{245}Cm metal and water without and with a water reflector are smaller when the calculations are made based on ENDF/B-VI than JENDL-3.2. The neutron energy spectrum for the bare sphere of curium-water mixture has two peaks: at 50 meV and at 3 MeV. The relations between ENDF/B-VI and JENDL-3.2 for the averaged number of neutrons emitted by a fission of ^{245}Cm, fission cross section and capture cross section at 50 meV are similar to those at 3 MeV (see Figs. D3 and D5 in Appendix D(24)). Therefore, it is reasonable that the calculated minimum critical mass of ^{245}Cm based on ENDF/B-VI is smaller than that of JENDL-3.2.

3.2. Comparison with other data

Appendix C summarizes a survey of studies made on critical masses of curium isotopes since the publication of the ANSI/ANS-8.15-1981. Hereafter we compare our results with those studies and the subcritical mass limits given in the ANSI standard. (The subcritical limit was defined as *the limiting value assigned to a controlled parameter that results in a* *system known to be subcritical, provided the limiting value of no other controlled parameter of the system is violated. The subcritical limit allows for uncertainties in the calculations used in its derivation but not for contingencies; e.g., double batching or failure of analytical techniques to yield accurate values of process variables.*(1) The subcritical margin Δk_{eff}, which should be subtracted from unity to find the k_{eff} corresponding the subcritical limit, depends on the supporting experimental data. We can find some examples from the literature for the value: 0.05 for mixed uranium and plutonium oxides with full water reflection specified(15) where a limited number of applicable experiments are available; 0.07 was thought to be an adequate margin for ^{235}U moderated with the unfamiliar moderators as SiO_2, C, Be or D_2O(25); and 0.1 or larger for those systems without applicable critical experiments.(14))

(1) ^{245}Cm

Table 12 gives a comparison of the critical masses of ^{245}Cm metal with and without a water reflector. The present calculation result based on the JENDL-3.2 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, and agrees well with the results based on JENDL-3.1, ENDL-82 and -85. However, the Anno's result based on JEF-2, 6.81 kg, was far smaller. The other result based on the ENDF/B-V has a similar value to our ENDF/B-VI result, 9.41 kg. A similar tendency was found for the results with a water reflector.

Various results for the minimum critical masses of a water mixture of ^{245}Cm metal granules are shown in Table 13. The present result based on the JENDL-3.2 is almost equal to that of Srinivasan's for the case without reflector, but a little larger than the corresponding for the water reflected case. Rossignol supplied a considerably smaller value, 42.7 g, which is consistent with the subcritical mass limit, 30 g, of ANSI/ANS-8.15-1981 (see Table 14). Both of the minimum subcritical masses, corresponding to $k_{eff} = 0.9$, obtained by calculations based on JENDL-3.2 and ENDF/B-VI are larger than 40 g, therefore, considerably larger than the value suggested by ANSI/ANS-8.15-1981.

(2) ^{246}Cm

Table 15 compares the critical masses of ^{246}Cm with a water reflector, with an SUS reflector, and without reflectors. The consistencies with Nojiri's results maintained: the JENDL-3.2 results are about 80 % larger than those based the ENDF/B-V or -VI results. The ENDL-85 gave a result more than twice as large as the ENDF/Bs' results. The critical mass of a bare sphere of ^{246}Cm metal based on the JEF-2 was found about 10% larger than the corresponding mass based on the ENDF/B-V and -VI.

There is no description on the subcritical mass limit for ^{246}Cm in the ANSI/ANS-8.15-1981. The present calculation based on JENDL-3.2 suggests 25 kg for the subcritical mass limit for SUS reflected ^{246}Cm. Considering that Nojiri's SCALE4.3 result based on the ENDF/B-V gives 20.5 kg as the critical mass, the subcritical mass limits should be smaller than the value just suggested. For subcritical mass limits of ^{246}Cm with a water reflector and with a SUS reflector, 23 kg and 14 kg, respectively, would be appropriate, corresponding to 70 % of the critical masses 32.9 kg and 20.5 kg, each of which is the minimum of the corresponding values obtained.

(3) ^{247}Cm

Table 16 compares the critical masses of ^{247}Cm with a water reflector, with an SUS reflector, and without reflectors. In comparison with the other two nuclides mentioned above, the differences among these results are small: they are in a band from 6.9 to 7.9. Looking more carefully at Table 16, however, we would notice the following: (1) ENDLs' results are larger than the results based on other libraries, i.e., JENDLs, ENDF/Bs and JEF-2. (2) Rather large discrepancies are observed between our JENDL-3.2 result and the corresponding of Nojiri's. This might be due to the difference in the densities of ^{247}Cm metal: Nojiri used 13.5 g/cm^3, whereas we assumed 13.68 g/cm^3, considering mass differences among different isotopes.

The minimum critical masses of a water mixture of ^{247}Cm metal granules are compared in Table 17. The present results based on the JENDL-3.2 are 16 % and 20 % smaller than those of Srinivasan for bare and water reflected cases, respectively. These differences are due to the difference in the nuclear data libraries.

The subcritical mass limit of the ANSI/ANS-8.15-1981 for water reflected ^{247}Cm, 900 g, are small enough compared to our subcritical mass of water reflected ^{247}Cm, 1.49 kg for k_{eff} = 0.9 (see Table 8). It should be noted here that Clark calculated the minimum mass at k_{eff} = 0.9 for ^{247}Cm in CmO_2 as 1360 g. Because of the uncertainty of ν-value, he concluded that a suitable subcritical mass limit was 900 g.(14) For discussing revision of the ANSI/ANS-8.15-1981, therefore, we should be cautious to calculate the corresponding value using other data libraries as well.

3.3. Amount of curium isotopes in typical burnup fuels

Ando and Takano(26) calculated compositions of UO_2 and mixed plutonium and uranium oxide (MOX) fuels irradiated in typical boiling water reactor (BWR) and pressurized water reactor (PWR) conditions up to the burnup of 33, 45 and 60 GWd/tHM (tHM: ton heavy metal). Contents of curium isotopes are displayed in Figs. 19 and 20. Anno and Sert also calculated spent fuel compositions, their results being shown in Table 18 for typical fuels irradiated for 35 GWd/t and cooling time of 90 days.(5), (13) The indicated values are the average amounts of 8 selected cases for respective type of fuels (UO_2 fuels in PWRs and BWRs, and MOX fuels in PWRs).

In comparing the two independent sources above, we see that the curium isotope contents for UO_2 fuels discharged in PWR and BWR, and MOX fuels in PWR are in good agreement. It is clear from the figures that (1) the amount of curium isotopes increases dramatically as the fuel burnup increases. From both the figures and the table, it is further obvious that (2) ^{244}Cm dominates other curium isotopes in quantity; (3) curium contents in MOX fuels are one order larger than UO_2 fuels at the same burnup; ^{247}Cm is far less important than ^{245}Cm in evaluating criticality safety of burnup fuels both in reactivity and produced amount; and (4) the amount of ^{242}Cm is not negligible, e.g. in comparison with the amount of ^{246}Cm, especially when no cooling time is assumed.

4. Concluding Remarks

Critical (k_{eff} = 1.0) and subcritical (k_{eff} = 0.9 and 0.8) masses of three curium isotopes, ^{245}Cm, ^{246}Cm and ^{247}Cm, were calculated with a combination of the current version of the Japanese Evaluated Nuclear Data Library, JENDL-3.2, and a continuous energy Monte Carlo neutron transport code, MCNP4A. The subcritical masses corresponding to k_{eff} = 0.9 for ^{246}Cm metal and $^{246}CmO_2$ with a 30-cm-thick stainless-steel reflector were computed as 25.2 kg and 41.8 kg, respectively. These quantities were not included in ANSI/ANS-8.15-1981, and requested by the leader of ANSI/ANS-8.15 Work Group, Mr. N.L. Pruvost. As we found Nojiri's independent results for the critical mass of ^{246}Cm metal with water using SCALE4.3 code system had smaller values, 20.5 kg, than our subcritical mass, we suggested 14 kg would be appropriate to be included into the new version of ANSI/ANS-8.15 complying with conservatism.

The minimum critical mass of a homogeneous mixture of ^{245}Cm granules and water in a sphere with a fully thick water reflector was obtained as 65.6 g. The corresponding quantity for ^{247}Cm was found to be 2.19 kg. The critical masses of ^{245}Cm, ^{246}Cm and ^{247}Cm metals were computed also by replacing the JENDL-3.2 with the ENDF/B-VI, and they were reduced by 12 %, 45 % and 2 %, respectively, from each corresponding value, which revealed a large dependence of the results on the evaluated nuclear data libraries.

Importance of evaluating subcritical limits for ^{242}Cm is suggested in this report, which has larger abundance compared to ^{246}Cm if no cooling time is assumed. Curium-242 has a half-life of 162.8 days; therefore, its amount will reduce to 21% after one-year of cooling time.

Acknowledgments

We are grateful to all the members of the Work Group for revising ANSI/ANS-8.15, for having welcomed one of us (HO) into the group; especially to the chair of the Group, N.L. Pruvost, for having assigned him the curium isotopes, and to J. Anno for showing the group members the French results. We thank I. Nojiri and Y. Fukasaku for supplying us with their both published and unpublished results; to C. Nordborg for permitting us to reproduce a part of the report *NEA/WPEC*-8 published from OECD/NEA; to T. Nakagawa for preparing for us graphs comparing cross sections for the curium isotopes ^{246}Cm and ^{247}Cm as shown in Appendix D; to R. Ando and H. Takano for allowing us to refer their burnup calculation results; to Y. Komuro for providing us useful information; and A. Hasegawa for a careful proof-reading of the manuscript.

References

(1) "American National Standard for Nuclear Criticality Control of Special Actinide Elements," *ANSI/ANS*-8.15-1981 (1981).

(2) R.W. Brewer, N.L. Pruvost and C.T. Rombough, "ANSI/ANS-8.15-1981 (R87): Nuclear Criticality Control of Special Actinide Elements," *Trans. Am. Nucl. Soc.* **75**, 200 (1996).

(3) "Regulations for the Safe Transport of Radioactive Material, 1996 Edition" *ST*-1, Intl. Atomic Energy Agency, Vienna (1996).

(4) IAEA, TRANSAC-III, *TC*-983.3 Working Paper No. 22, "Towards an Extension of the Definition of Fissile Materials and their Mass Exemption Limits," (1998).

(5) J. Anno and G. Sert, "French Participation at ANS/ANSI 8/15 Working Group Updating Criticality Data on ^{237}Np - Criticality & Transportation Proposals," *Proc. of the Sixth Intl. Conf. on Nucl. Criticality Safety, ICNC'99*, Vol.I, 447, Sept. 20-24, 1999, Versailles (1999).

(6) D. M. Barton, "Central Reactivity Contributions of ^{244}Cm, ^{239}Pu, and ^{235}U in a Bare Critical Assembly of Plutonium Metal," *Nucl. Sci. and Eng.*, **33**, 51 (1968); R.W. Brewer, "Replacement Measurements Performed with Curium-244, Plutonium-239, and HEU Using JEZEBEL," *NEA/NSC/DOC*(95)03/VII, *SPEC-MET-FAST*-001.

(7) R. G. Sanchez, "Critical Mass of Np-237," *Proc. of the Fifth Intl. Conf. on Nucl. Criticality Safety, ICNC'95,* Vol.I, 6•182, September 17-21, 1995, Albuquerque, NM, (1995); R.W. Brewer, "Neptunium-237 and Highly Enriched Uranium Replacement Measurements Performed Using Flattop," *NEA/NSC/DOC*(95)03/VII, *SPEC-MET-FAST*-003.

(8) M. Srinivasan, K. Subba Rao, S.B. Garg and G.V. Acharya, "Systematics of Criticality Data of Special Actinides Deduced Through the Trombay Criticality Formula," *Nucl. Sci. and Eng.*, **102**, 295 (1989).

(9) M. Roussignol, "Détermination des Masses Critique Relatives à Certains Actinides Fissiles; Criticité de l'Américium et du Curium Issus de Combustibles REP Irradiees", *SEC/T*/208/94.332 (1994) [in French].

(10) R.R. Rahn, "Criticality Safety Evaluation of Mixtures Containing Americium and Curium," *Proc. of the Fifth Intl. Conf. on Nucl. Criticality Safety, ICNC'95*, Vol.II, 11•101, Sept. 17 - 21, 1995, Albuquerque, NM (1995).

(11) Y. Komuro, T. Takada and T. Arakawa, "Estimation of Critical Mass for Actinoids," *1995 Fall Mtg. of the Atomic Energy Soc. of Japan*, Oct. 17-20, 1995, Tokai-mura, B55 (1995) [Abstract form only, in Japanese].

(12) I. Nojiri and Y. Fukasaku, "Calculational Study for Criticality Safety Data of Fissionable Actinides," *Proc. of GLOBAL '97, Int. Conf. on Future Nuclear Systems*, Oct. 5-10, 1997, Yokohama, Japan, Vol.2, 1397-1401 (1997).

(13) J. Anno, "Données de criticité des actinides fissiles en métal et/ou en solution aqueuse sous forme de sphères nues et réfléchies," *SEC/T*/99.196 (1999) [in French].

(14) H.K. Clark, "Subcritical Limits for Special Fissile Actinides," *Nucl Technol.*, **48**, 164 (1980).

(15) N.L. Pruvost and H.C. Paxton, "Nuclear Criticality Safety Guide," *LA*-12808, *UC*-714 (1996).

(16) J.F. Briesmeister (ed.), "MCNP - A General Monte Carlo N-Particle Transport Code, Version 4A," *LA*-1265-*M* (1993).

(17) *Proceedings of the Topical Meeting on Physics and Methods in Criticality Safety*, September 19-23, 1993, Nashville, Tennessee (1993).

(18) T. Nakagawa et al., "Japanese Evaluated Nuclear Data Library, Version 3, Revision 2: JENDL 3.2," *J. Nucl. Sci. Technol.*, **32**, 1259 (1995).

(19) ENDF/B-VI Summary Documentation, *BNL-NCS*-17541 (ENDF-201), 4th edition (1991).

(20) Nuclear Materials Regulation Division/ Nuclear Safety Bureau/ Science and Technology Agency, "Nuclear Criticality Safety Handbook," *Nikkan-shobo*, Tokyo (1988) [in Japanese]; (tr.) Y. Naito and H. Okuno, "Nuclear Criticality Safety Handbook (English Translation)," *JAERI-Review* 95-013 (1995).

(21) C.T. Rombough, S.H. Martonak, and N.L. Pruvost, "Search Technique for Calculating Critical and Subcritical Configurations with MCNP," *ANS Trans.* **76**, 197, (1996).

(22) (Eds.) K. Shibata, T. Nakagawa , H. Sugano and H. Kawasaki, "Curves and Tables of Neutron Cross Sections in JENDL-3.2," *JAERI-Data/Code* 97-003 (1997).

(23) E.D. Clayton, "Anomalies of Nuclear Criticality," *PNL-SA*-4868 Rev. 5 (1979).

(24) T. Nakagawa, H. Takano and A. Hasegawa, "Present Status of Minor Actinide Data," *NEA/WPEC*-8 (1999).

(25) C.V. Parks, C.M. Hopper and J.L. Lichtenwalter, "Assessment and Recommendations for Fissile-Material Packaging Exemptions and General Licenses Within 10 CFRT part 71," NUREG/CR-5342, ORNL/TM-13607 (1998).

(26) Y. Ando and H. Takano, "Estimation of LWR Spent Fuel Composition," *JAERI-Research* 99-004 (1999) [in Japanese].

(27) J. Anno, *Private communication.*

Table 1. Specification for MCNP-4A calculation

Items	Value
Nominal number of source histories per source cycle	2000
Total number of cycles	270
Number of skipped cycles before tally	20
Initial spatial distribution for the source	uniform

Table 2. The calculation objects[*1]

Nuclide	Chemical form	Reflector[*2]	Note
^{245}Cm	Cm-H_2O, CmO_2-H_2O	None, water	
^{246}Cm	Cm, CmO_2	None, stainless steel	At the theoretical density only
^{247}Cm	Cm-H_2O, CmO_2-H_2O	None, water, stainless steel	

[*1] The geometrical shape was assumed to be a sphere.

[*2] The thickness of water and stainless steel was assumed to be 30 cm.

Table 3. Density and atomic composition of stainless steel (Type 304)$^{(20)}$

Density	7.93 g/cm^3
Atomic element	Ratio [wt.%]
C	0.08
Si	1
Mn	2
P	0.045
S	0.03
Ni	9.25
Cr	19
Fe	68.595
Total	100

Table 4. Critical and subcritical masses of ^{245}Cm in metallic and oxide forms

Chemical form	Fuel density [gCm/cm^3]	Reflector	Cm mass [kg]		
			k_{eff}=1.0	k_{eff}=0.9	k_{eff}=0.8
Cm	13.57	None	12.3	8.71	5.99
		Water	3.91	2.71	1.82
CmO_2	10.58	None	12.8	9.36	6.64
		Water	4.33	3.00	2.01

Table 5. Critical and subcritical masses of ^{246}Cm in metallic and oxide forms

Chemical form	Fuel density [gCm/cm^3]	Reflector	Cm mass [kg]		
			k_{eff}=1.0	k_{eff}=0.9	k_{eff}=0.8
Cm	13.62	None	70.1	45.5	28.9
		Stainless steel	38.1	25.2	16.2
CmO_2	10.62	None	111	69.0	42.6
		Stainless steel	67.7	41.8	25.9

Table 6. Critical and subcritical masses of ^{247}Cm in metallic and oxide forms

Chemical form	Fuel density [gCm/cm^3]	Reflector	Cm mass [kg]		
			k_{eff}=1.0	k_{eff}=0.9	k_{eff}=0.8
Cm	13.68	None	7.06	5.02	3.44
		Water	3.10	2.18	1.49
		Stainless steel	3.07	2.28	1.63
CmO_2	10.67	None	7.98	5.83	4.11
		Water	3.62	2.57	1.77
		Stainless steel	3.58	2.69	1.96

Table 7. Minimum critical/subcritical mass of ^{245}Cm in ^{245}Cm-H_2O

k_{eff}	Bare		Water-reflected	
	Concentration [gCm/L]	Mass [gCm]	Concentration [gCm/L]	Mass [gCm]
1.0	10.0	138	12.1	65.6
0.9	8.6	105	12.1	47.7
0.8	8.1	79	10.9	34.8

Table 8. Minimum critical/subcritical mass of ^{247}Cm in ^{247}Cm-H_2O

k_{eff}	Bare		Water-reflected		SUS-reflected	
	Concentration [gCm/L]	Mass [kgCm]	Concentration [gCm/L]	Mass [kgCm]	Concentration [gCm/L]	Mass [kgCm]
1.0	208	4.24	301	2.19	310	1.56
0.9	200	2.98	258	1.49	265	1.11
0.8	200	2.12	205	1.00	214	0.77

Table 9. Critical masses of Cm isotopes calculated with different nuclear data libraries

Cm isotopes	Reflector	Critical mass (k_{eff}=1) [kgCm]		M_E/M_J
		MJ(JENDL-3.2)	ME(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 10. Calculated minimum critical masses of ^{245}Cm-H_2O 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]
None	JENDL-3.2	10.0	138
	ENDF/B-6	8.1	117
Water	JENDL-3.2	12.1	65.6
	ENDF/B-6	11.6	54.9

Table 11. Comparison of averaged numbers of neutrons (ν-value) produced by a neutron-induced fission between JENDL-3.2 and ENDF/B-VI at the neutron energy of 2 MeV

	JENDL-3.2	ENDF/B- VI	B-VI /J-3.2
Cm-245	3.82	3.98	1.043
Cm-246	3.59	3.87	1.078
Cm-247	4.20	3.98	0.946

Table 12. Comparison of critical/subcritical masses of ^{245}Cm metal [kg]

Author	Nuclear Data Library	Computer Code	Bare		Water reflected	
			k_{eff} =1	k_{eff} =0.9	k_{eff} =1	k_{eff} =0.9
Present	JENDL-3.2	MCNP4A	12.3	8.71	3.91	2.71
Present	ENDF/B-VI	MCNP4A	9.41		3.03	
Anno	JEF-2	DTF-IV	6.81		2.61	
Komuro	JENDL-3.1	MULTIKENO	12.4		3.78	
Nojiri	ENDF/B-V	SCALE4.3	9.33		3.14	
Nojiri	ENDL-85	MCNP4A	12.5		3.86	
Nojiri	JENDL-3.2	MCNP4A	12.4		3.59	
Srinivasan	ENDL-82	DTF-IV	12.28			

Table 13. Comparison of the minimum critical masses of ^{245}Cm metal granules in a water mixture

Author	Nuclear Data Library	Computer Code	Bare		Water reflected	
			Cm mass [g]	Cm conc. [g/L]	Cm mass [g]	Cm conc. [g/L]
Present	JENDL-3.2	MCNP4A	138	10.0	65.6	12.1
Present	ENDF/B-VI	MCNP4A	117	8.1	54.9	11.6
Roussignol	JEF-1	DTF-IV			42.7	
Srinivasan	ENDL-82	DTF-IV	136	12	62	12

Table 14. Comparison of the minimum subcritical masses corresponding to k_{eff}= 0.9 of ^{245}Cm metal granules in a water mixture

Author	Nuclear Data Library	Computer Code	Bare		Water reflected	
			Cm mass [g]	Cm conc. [g/L]	Cm mass [g]	Cm conc. [g/L]
Present	JENDL-3.2	MCNP4A	105	8.6	47.7	12.1
Present	ENDF/B-VI	MCNP4A	89.2	8.1	40.5	11.4
Clark[*1]	ENDF/B-IV	ANSIN			30[*2]	12
Roussignol	JEF-1	DTF-IV			29.8[*3]	

*1 Adopted in ANSI/ANS-8.15-1981

*2 Obtained for a water-reflected CmO$_2$-H$_2$O mixture

*3 70% of the critical mass

Table 15. Comparison of critical/subcritical masses of ^{246}Cm metal [kg]

Author	Nuclear Data Library	Computer Code	Bare		Water reflected		SUS reflected	
			k_{eff} =1	k_{eff} =0.9	k_{eff} =1	k_{eff} =0.9	k_{eff} =1	k_{eff} =0.9
Present	JENDL-3.2	MCNP4A	70.1	45.5			38.1	25.2
Present	ENDF/B-VI	MCNP4A	39.0					
Anno	JEF-2	DTF-IV	42.5					
Nojiri	ENDF/B-V	SCALE4.3	37.9		32.9		20.5	
Nojiri	ENDL-85	MCNP4A	84.1		65.8		44.7	
Nojiri	JENDL-3.2	MCNP4A	70.0		58.6		38.7	

Table 16. Comparison of critical/subcritical masses of ^{247}Cm metal [kg]

Author	Nuclear Data Library	Computer Code	Bare		Water reflected		SUS reflected	
			k_{eff} =1	k_{eff} =0.9	k_{eff} =1	k_{eff} =0.9	k_{eff} =1	k_{eff} =0.9
Present	JENDL-3.2	MCNP4A	7.06	5.02	3.10	2.18	3.07	2.28
Present	ENDF/B-VI	MCNP4A	6.94					
Anno	JEF-2	DTF-IV	7.21					
Komuro	JENDL-3.1	MULTIKENO	7.00		2.91			
Nojiri	ENDF/B-V	SCALE4.3	7.15		3.66		2.94	
Nojiri	ENDL-85	MCNP4A	7.88		3.71		3.42	
Nojiri	JENDL-3.2	MCNP4A	7.25		3.01		3.15	
Srinivasan	ENDL-82	DTF-IV	7.87				3.36	

Table 17. Comparison of minimum critical masses of a mixture of ^{247}Cm metal and water

Author	Nuclear Data Library	Computer Code	Bare		Water reflected		SUS reflected	
			Cm mass [kg]	Cm conc. [g/L]	Cm mass [kg]	Cm conc. [g/L]	Cm mass [kg]	Cm conc. [g/L]
Present	JENDL-3.2	MCNP4A	4.24	208	2.19	301	1.56	310
Srinivasan	ENDL-82	DTF-IV	5.048	350	2.728	350		

Table 18. Amount of curium isotopes [g/tIHM] in spent fuels^{*1} according to Refs. (5) and (13)

Nuclide	UO$_2$ in PWRs	UO$_2$ in BWRs	MOX in PWR^{*2}
Cm-242	1.07E+01	6.44	9.54E+01
Cm-243	4.23E-01	2.40E-01	6.10E+00
Cm-244	3.48E+01	3.28E+01	4.97E+02
Cm-245	1.67	1.54	4.25E+01
Cm-246	2.59E-01	3.17E-01	4.97E+00
Cm-247	3.23E-03	4.53E-03	8.82E-02

*1 Burnup condition: 35 GWd/t, cooling time of 90 days

*2 The authors of Ref. (5) remarked the numbers appeared in the MOX column in Table 2 of Ref. (5) were false, whereas those in the original paper$^{(13)}$ are correct.$^{(27)}$

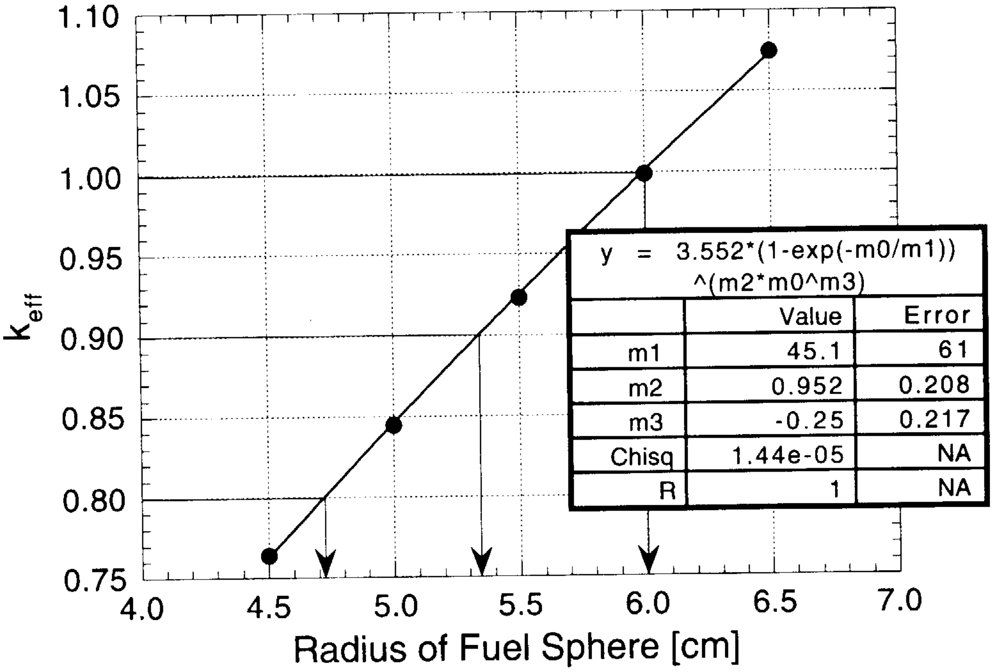

Fig. 1 Relation between spherical radius of ^{245}Cm metal without reflector and calculated values of k_{eff}

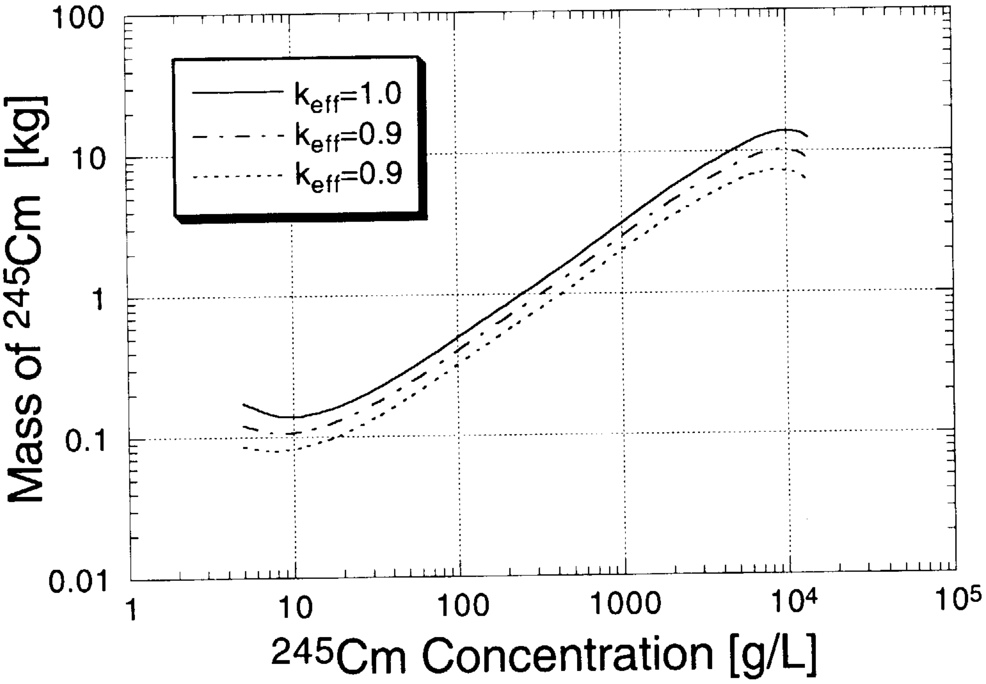

Fig. 2 Relation between ^{245}Cm concentration and critical/subcritical masses of ^{245}Cm for a sphere of ^{245}Cm-H_2O without reflector

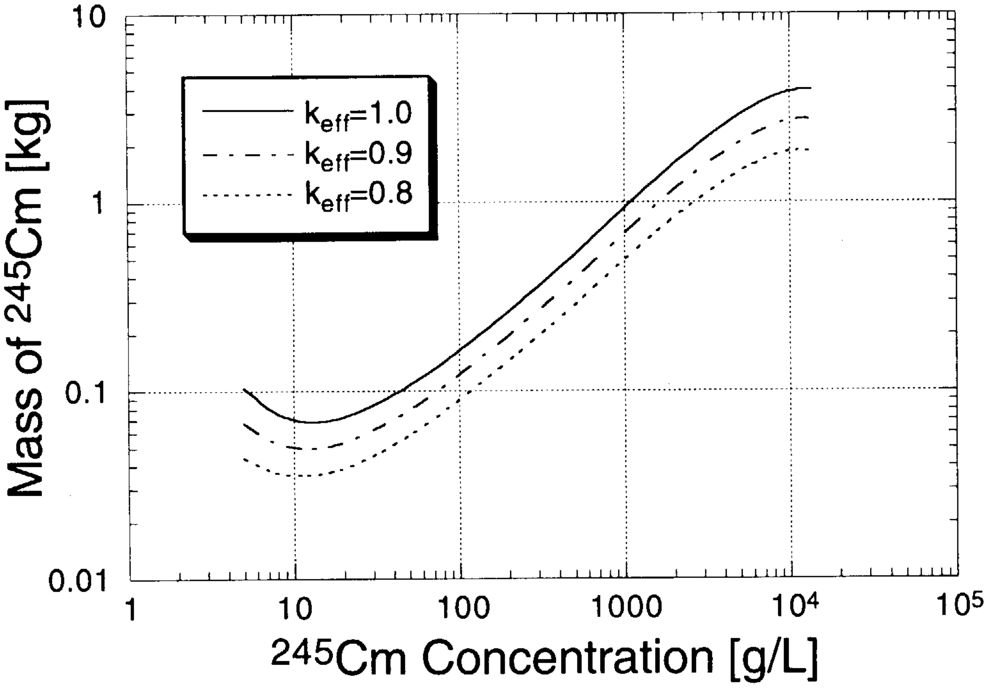

Fig. 3 Relation between ^{245}Cm concentration and critical/subcritical masses of ^{245}Cm for a sphere of ^{245}Cm-H_2O with water reflector

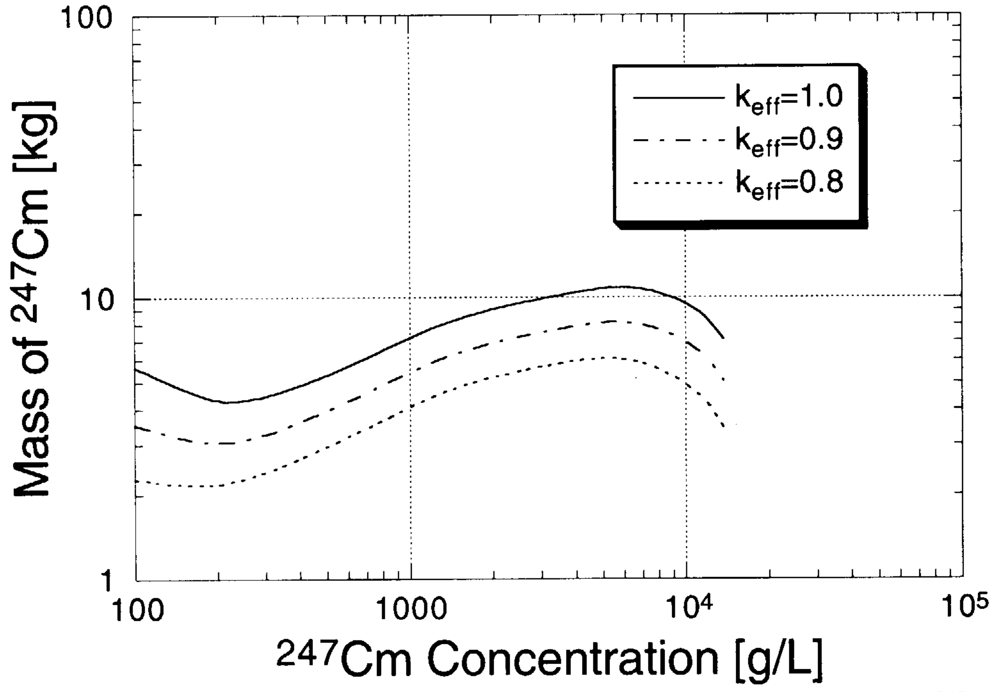

Fig. 4 Relation between ^{247}Cm concentration and critical/subcritical masses of ^{247}Cm for a sphere of ^{247}Cm-H_2O without reflector

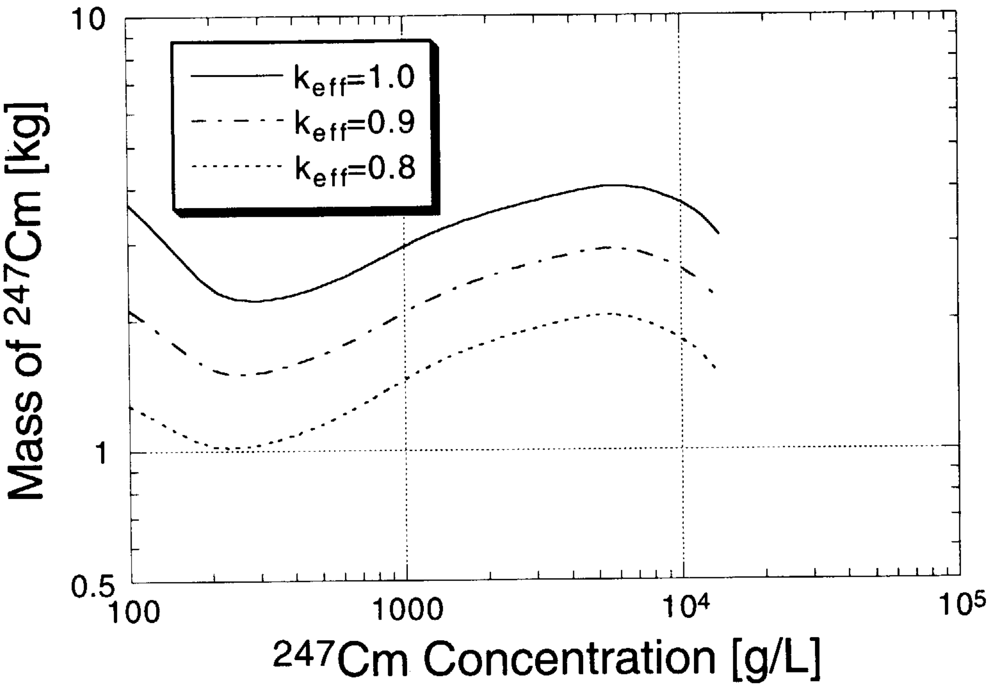

Fig. 5 Relation between ^{247}Cm concentration and critical/subcritical masses of ^{247}Cm for a sphere of ^{247}Cm-H_2O with water reflector

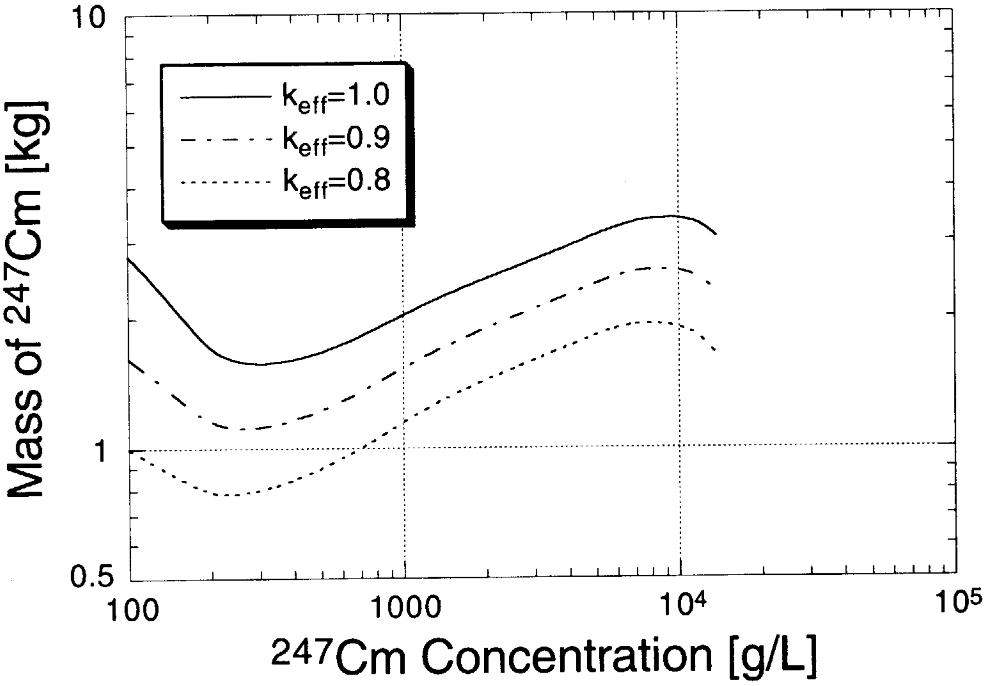

Fig. 6 Relation between ^{247}Cm concentration and critical/subcritical masses of ^{247}Cm for a sphere of ^{247}Cm-H_2O with SUS (Type 304) reflector

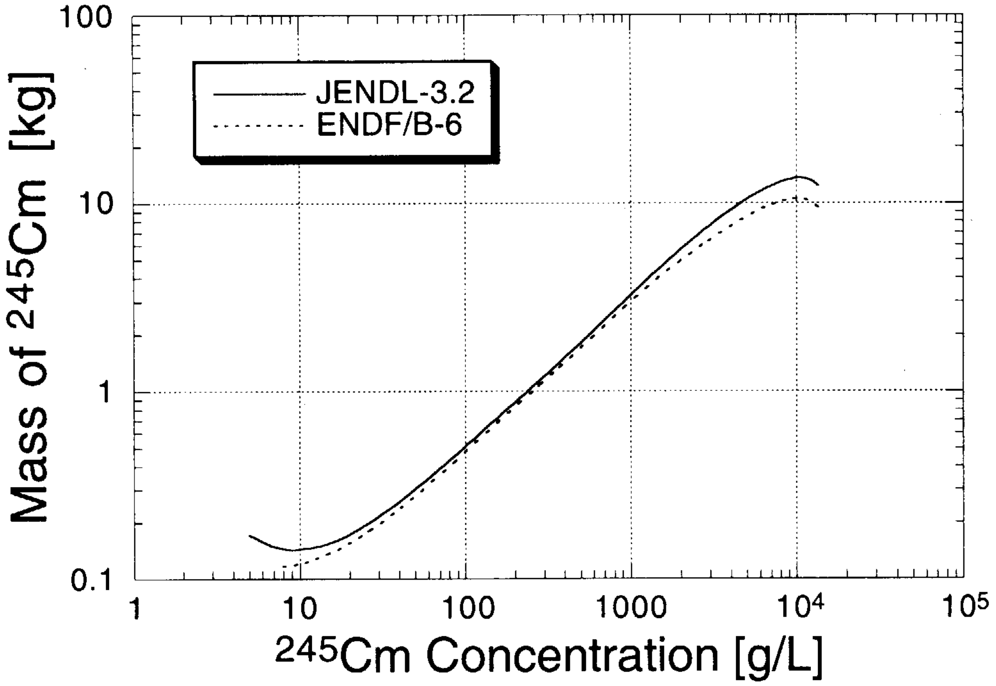

Fig. 7 Calculated critical masses based on the ENDF/B-VI library compared with those based on the JENDL-3.2 for a sphere of ^{245}Cm-H_2O without reflector

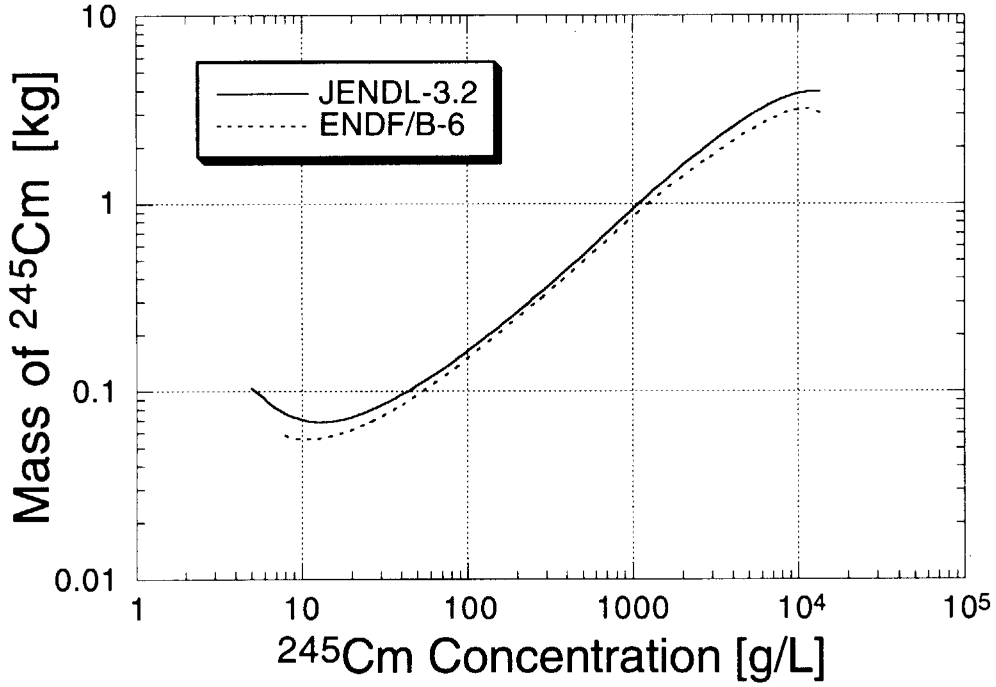

Fig. 8 Calculated critical masses based on the ENDF/B-VI library compared with those based on the JENDL-3.2 for a sphere of ^{245}Cm-H_2O with water reflector

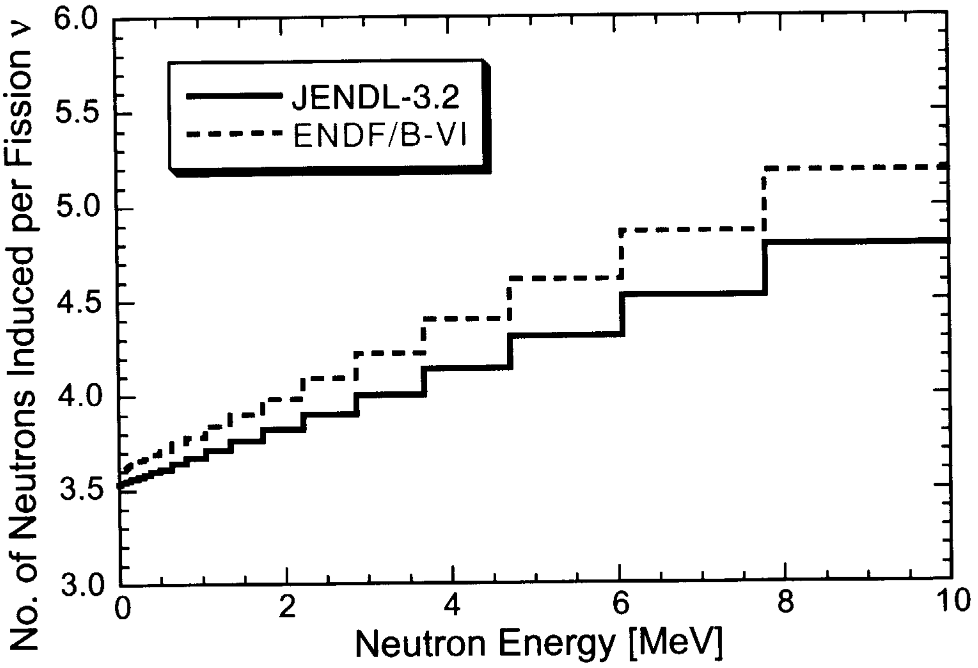

Fig. 9 The averaged number of neutrons produced by a neutron-induced fission of ^{245}Cm

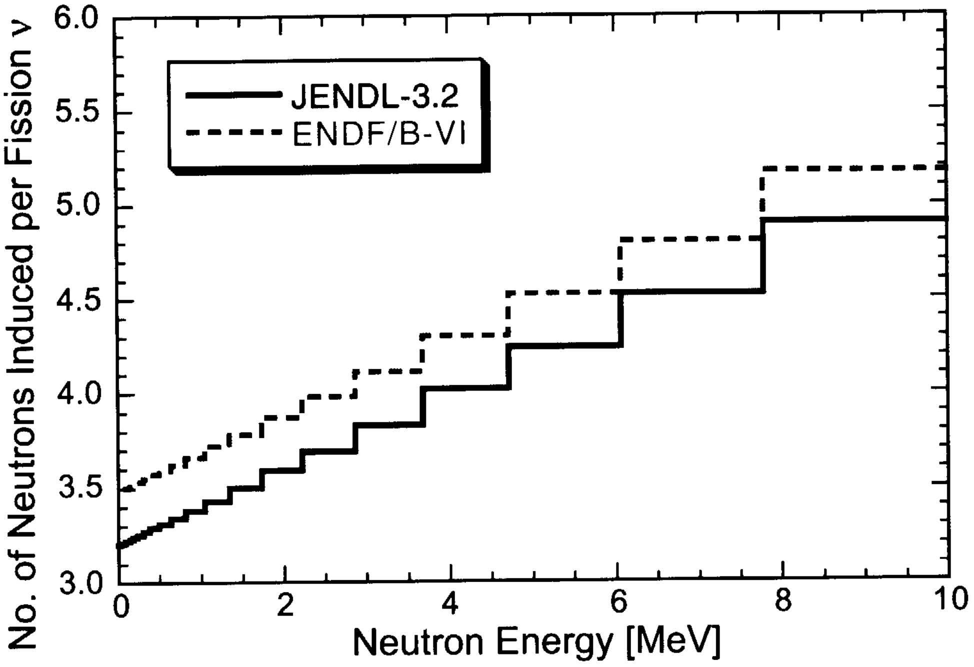

Fig. 10 The averaged number of neutrons produced by a neutron-induced fission of ^{246}Cm

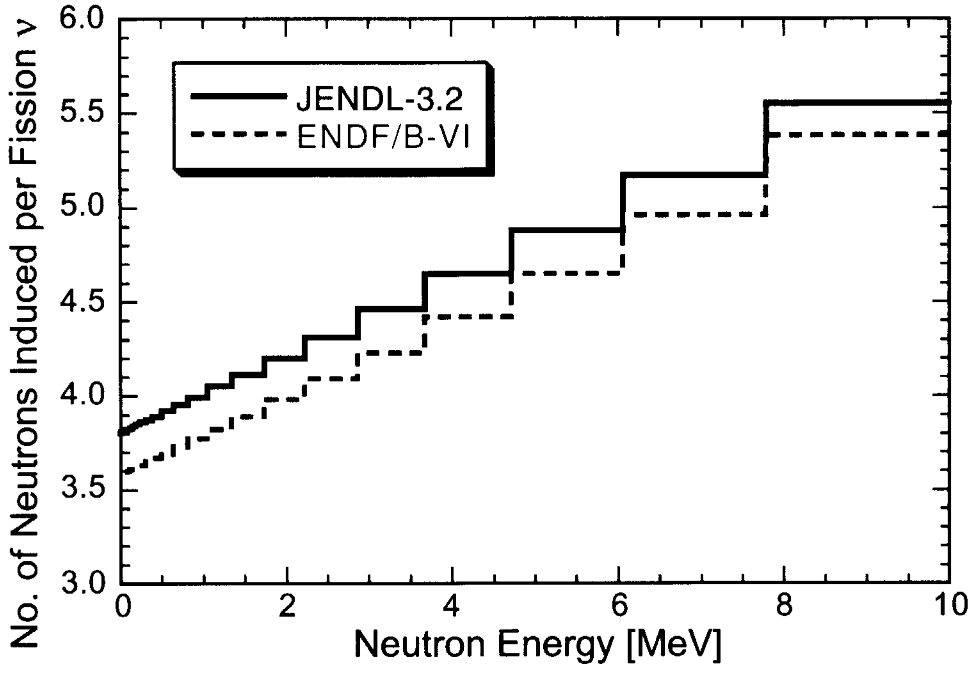

Fig. 11 The averaged number of neutrons produced by a neutron-induced fission of ^{247}Cm

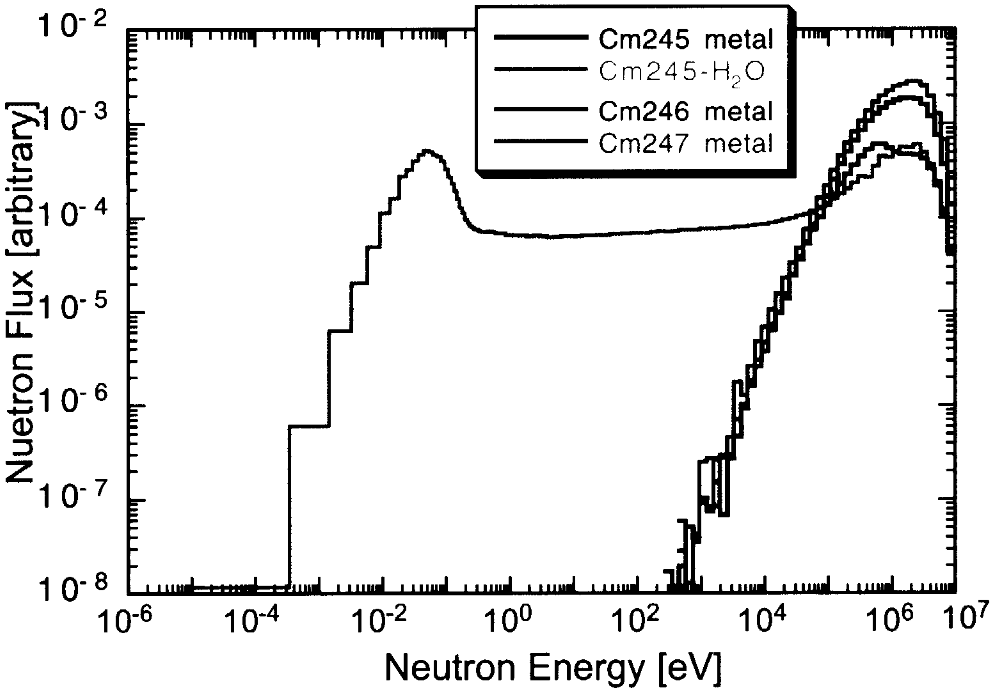

Fig. 12 The neutron spectra of critical bare spheres of ^{245}Cm-, ^{246}Cm- and ^{247}Cm-metals and a bare sphere of ^{245}Cm-H_2O (0.01 gCm/cm^3, radius: 14.887 cm)

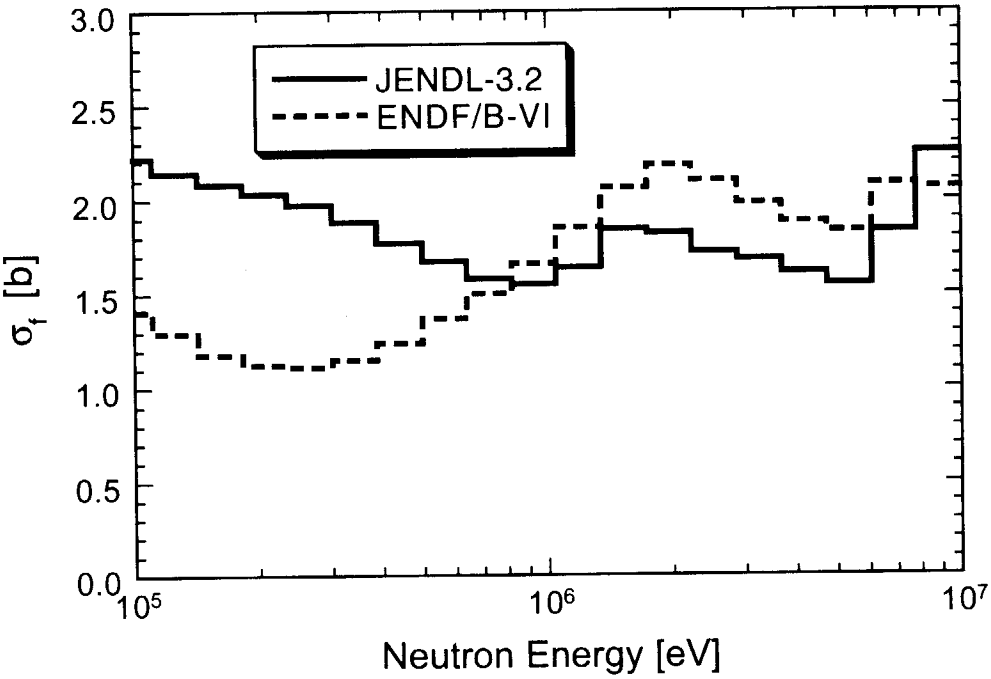

Fig. 13 Comparison of neutron induced fission cross sections of ^{245}Cm in JENDL-3.2 and in ENDF/B-VI

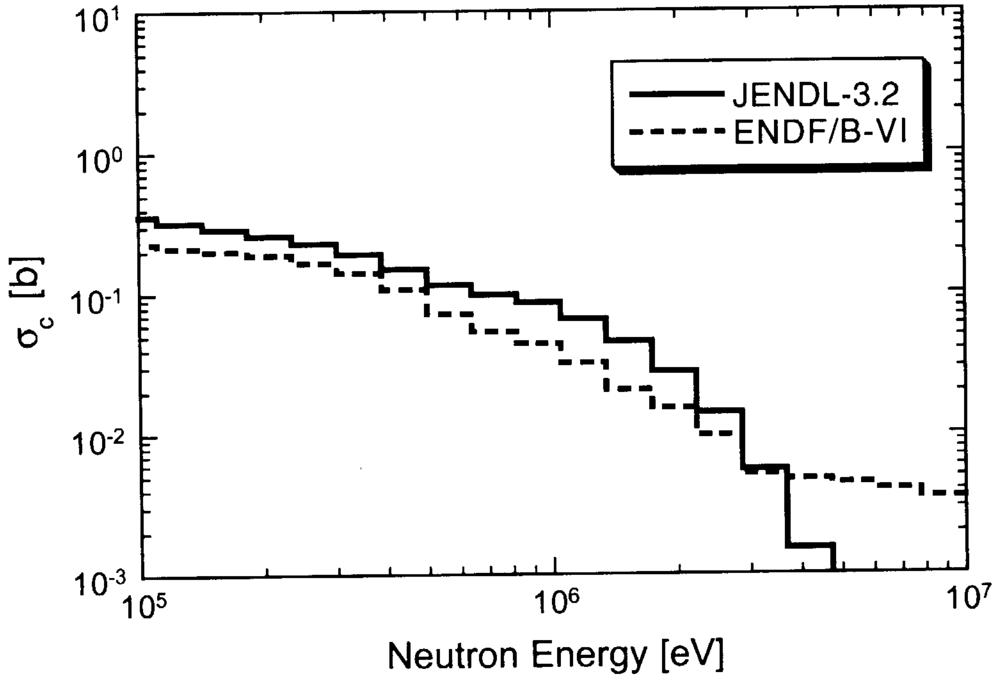

Fig. 14 Comparison of neutron capture cross sections of ^{245}Cm in JENDL-3.2 and in ENDF/B-VI

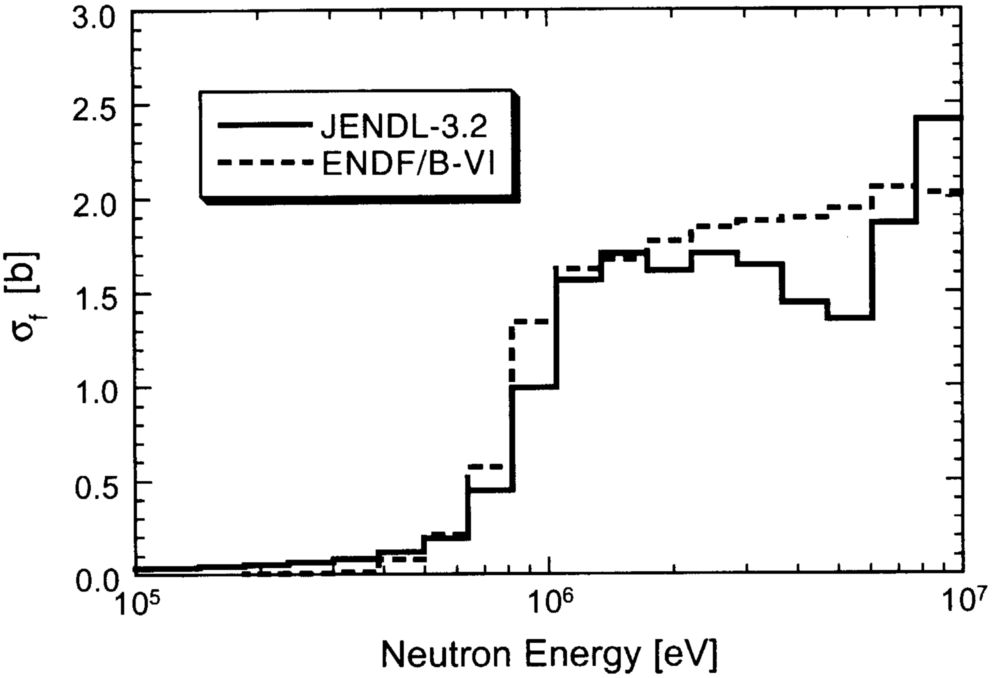

Fig. 15 Comparison of neutron induced fission cross sections of ^{246}Cm in JENDL-3.2 and in ENDF/B-VI

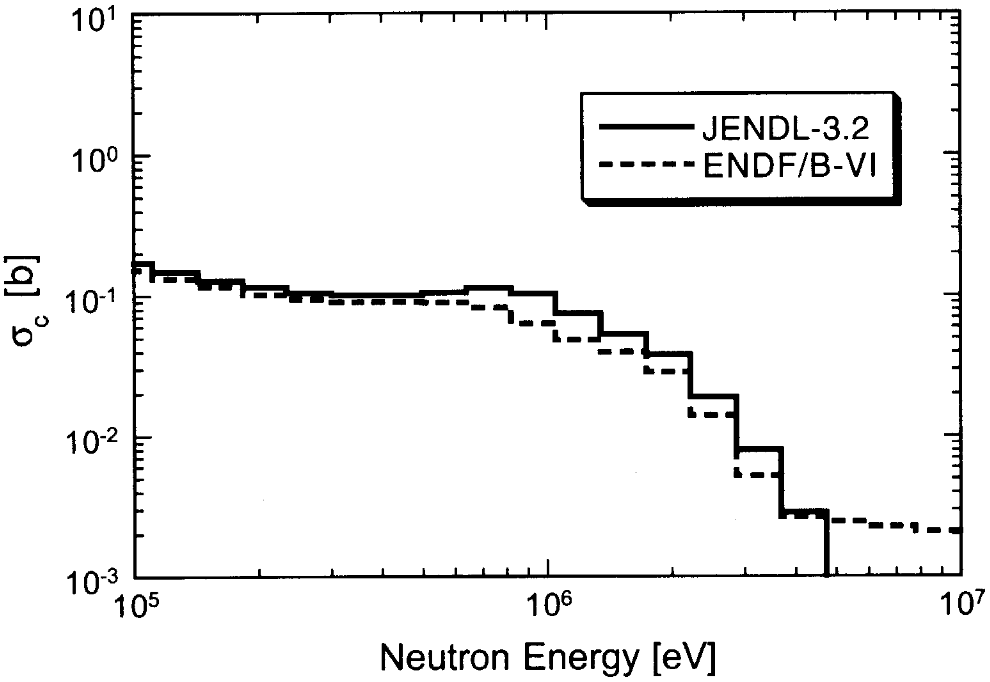

Fig. 16 Comparison of neutron capture cross sections of ^{246}Cm in JENDL-3.2 and in ENDF/B-VI

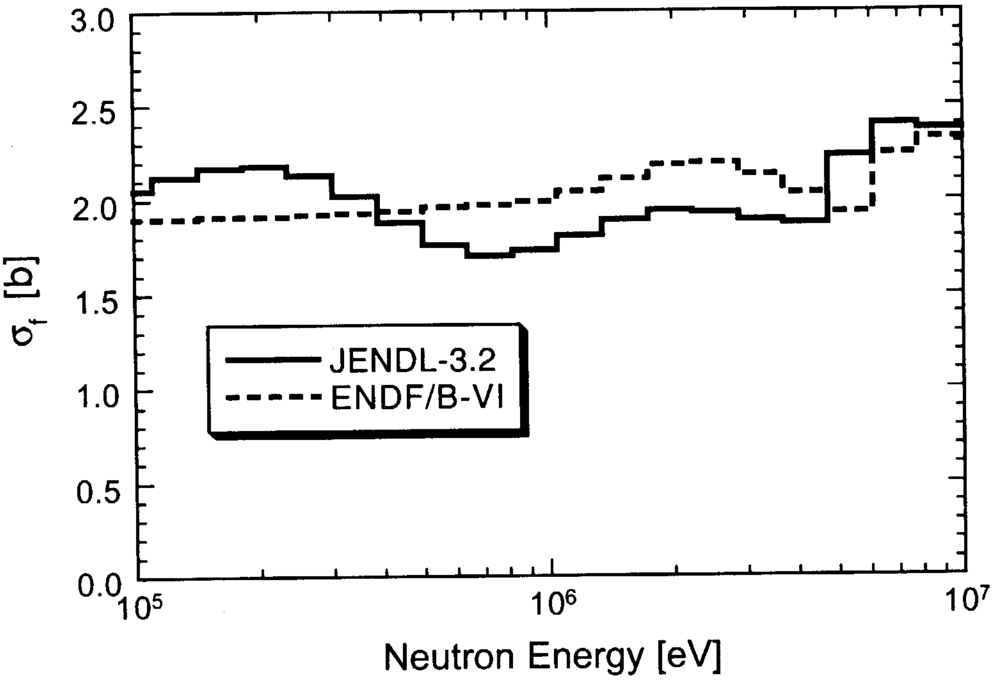

Fig. 17 Comparison of neutron induced fission cross sections of ^{247}Cm in JENDL-3.2 and in ENDF/B-VI

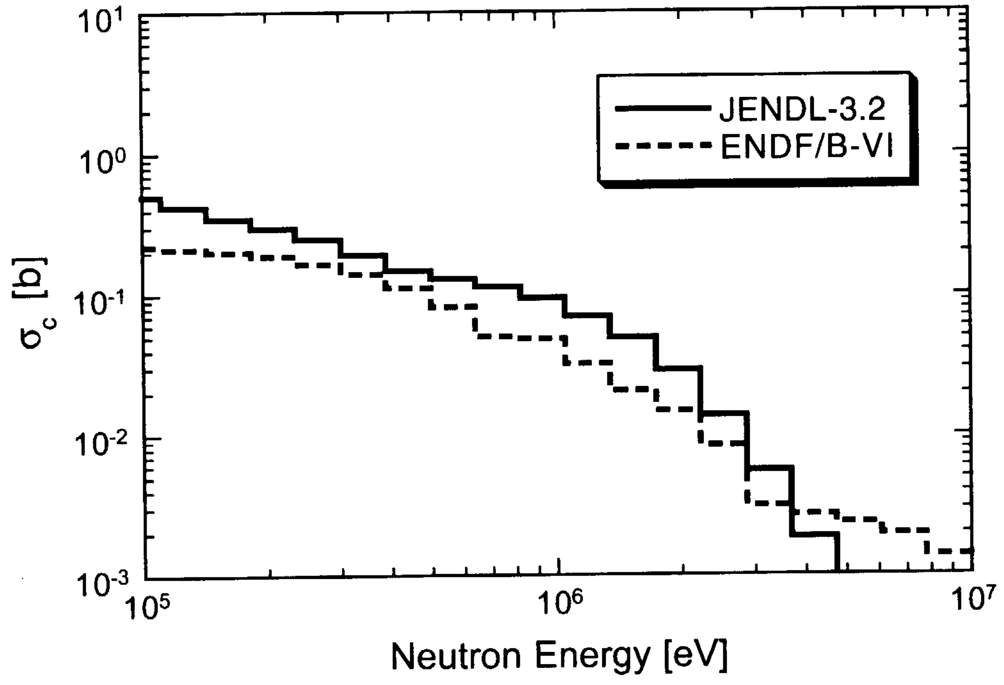

Fig. 18 Comparison of neutron capture cross sections of ^{247}Cm in JENDL-3.2 and in ENDF/B-VI

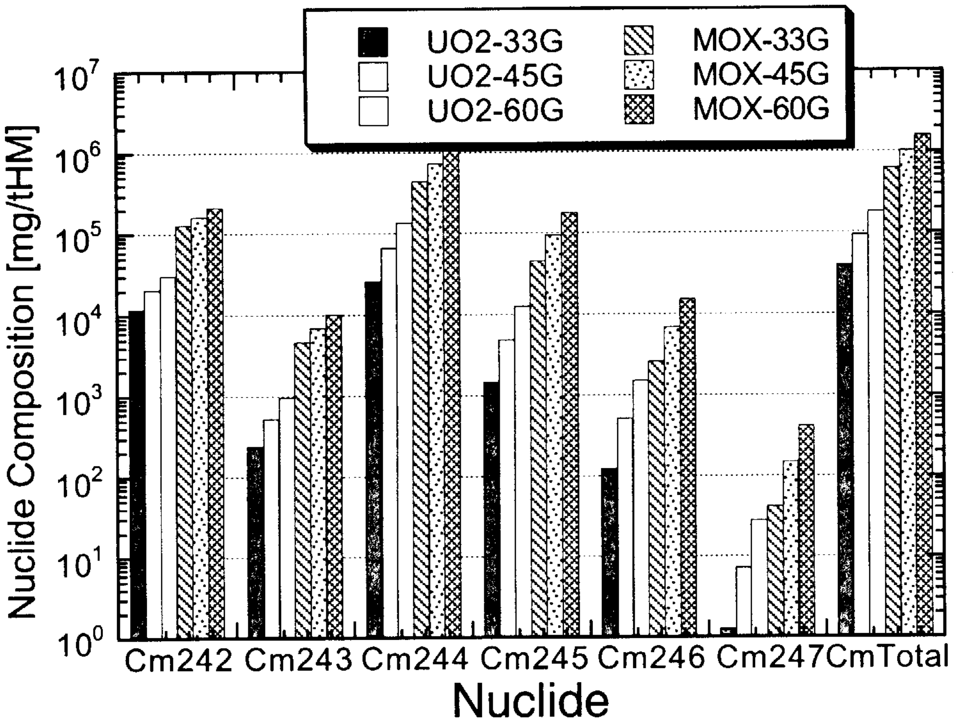

Fig. 19 Weights of curium isotopes in the UO_2 and MOX fuels irradiated in and just pulled out of PWR(26)

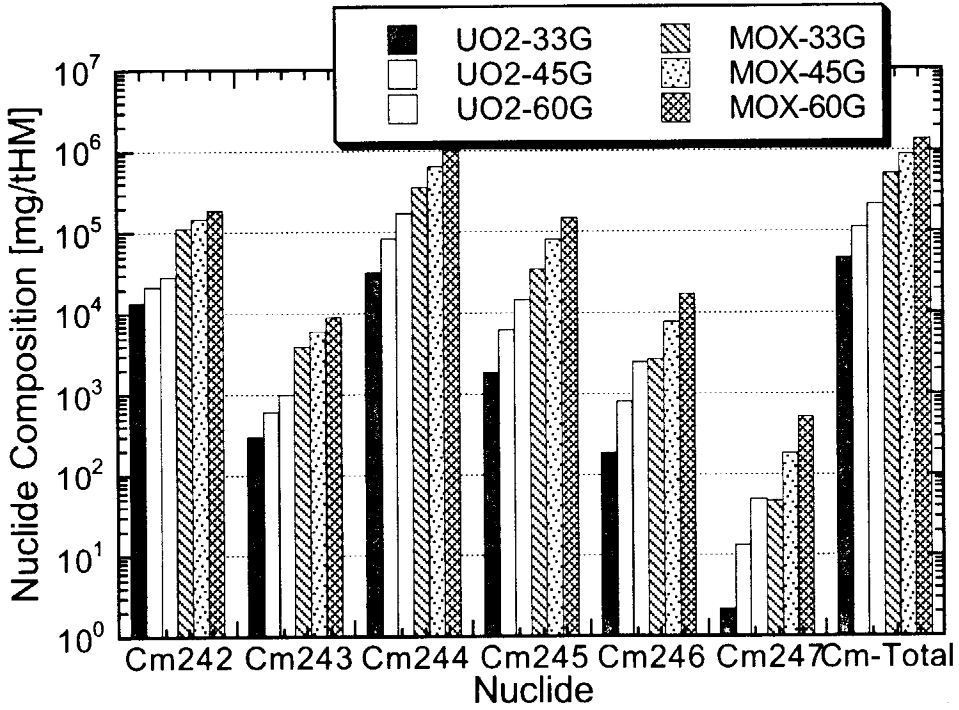

Fig. 20 Weights of curium isotopes in the UO_2 and MOX fuels irradiated in and just pulled out of BWR(26)

Appendix A. Formulae and Numbers Adopted in Calculating Atomic Number Densities for Cm-H_2O and CmO_2-H_2O

In this Appendix, the atomic number density formulae applied to in this report and the numbers adopted in the formulae are summarized. They are essentially in accordance with the *Nuclear Criticality Safety Handbook of Japan* (A1), taking into account the differences in mass of isotopes.

A.1. Constants

The Avogadro constant:
$N_A = 6.0221367\times10^{23}$ [#/mol].

Atomic mass of curium isotopes:
$A(^{244}\mathrm{Cm}) = 244.0627$ [g/mol],
$A(^{245}\mathrm{Cm}) = 245.0655$ [g/mol],
$A(^{246}\mathrm{Cm}) = 246.0672$ [g/mol], and
$A(^{247}\mathrm{Cm}) = 247.0703$ [g/mol].

Atomic mass of other elements:
$A(\mathrm{H}) = 1.0080$ [g/mol],
$A(\mathrm{O}) = 15.9949$ [g/mol].

A.2. Mass densities

Mass densities of curium isotopes:

$\rho(^{244}\mathrm{Cm}) = 13.51$ [g/cm^3](A2),

$$\rho(^{245}\mathrm{Cm}) = \rho(^{244}\mathrm{Cm})\cdot\frac{A(^{245}\mathrm{Cm})}{A(^{244}\mathrm{Cm})} = 13.57 \text{ [g/cm}^3\text{]},$$

$$\rho(^{246}\mathrm{Cm}) = \rho(^{244}\mathrm{Cm})\cdot\frac{A(^{246}\mathrm{Cm})}{A(^{244}\mathrm{Cm})} = 13.62 \text{ [g/cm}^3\text{]},$$

$$\rho(^{247}\mathrm{Cm}) = \rho(^{244}\mathrm{Cm})\cdot\frac{A(^{247}\mathrm{Cm})}{A(^{244}\mathrm{Cm})} = 13.68 \text{ [g/cm}^3\text{]}.$$

Mass densities of curium dioxide:

$$\rho(\mathrm{CmO_2}) = \frac{8A(\mathrm{O}) + 4A(Cm)}{a(\mathrm{CmO_2})^3\cdot N_A} \text{ [g/cm}^3\text{]},$$

where $a(\mathrm{CmO_2})$ is the cubic lattice parameter, which is equal to 5.3584 Å(A3). The relation above leads to the following numbers for mass densities of curium dioxides:

$\rho(^{245}\mathrm{CmO_2}) = 11.96$ [g/cm^3],
$\rho(^{246}\mathrm{CmO_2}) = 12.00$ [g/cm^3], and
$\rho(^{247}\mathrm{CmO_2}) = 12.05$ [g/cm^3].

Mass densities of water at 20 °C:

$\rho(\mathrm{H_2O}) = 0.99820$ [g/cm^3].

A.3. Atomic number densities

Atomic number densities of curium in curium metal, $N(\mathrm{Cm})$ [#/cm^3]:

$$N(\mathrm{Cm}) = \rho(\mathrm{Cm}) \cdot \frac{N_\mathrm{A}}{A(\mathrm{Cm})]}$$

Atomic number densities of curium, hydrogen and oxygen in a homogeneous mixture of curium metal and water, Cm-H_2O, at concentration of $C(\mathrm{Cm})$ [g/cm^3]:

$$N(\mathrm{Cm}) = C(\mathrm{Cm}) \cdot \frac{N_\mathrm{A}}{A(\mathrm{Cm})},$$

$$N(\mathrm{H_2O}) = N(\mathrm{Cm}) \cdot \frac{A(\mathrm{Cm})}{A(\mathrm{H_2O})} \cdot \rho(\mathrm{H_2O}) \cdot \left[\frac{1}{C(\mathrm{Cm})} - \frac{1}{\rho(\mathrm{Cm})} \right],$$

$N(\mathrm{H}) = 2 \cdot N(\mathrm{H_2O})$, and

$N(\mathrm{O}) = N(\mathrm{H_2O})$.

Atomic number densities of curium, hydrogen and oxygen in a homogeneous mixture of curium dioxide and water, CmO_2-H_2O, at concentration of $C(\mathrm{Cm})$ [g/cm^3]:

$$N(\mathrm{Cm}) = C(\mathrm{Cm}) \cdot \frac{N_\mathrm{A}}{A(\mathrm{Cm})},$$

$$N(\mathrm{H_2O}) = N(\mathrm{Cm}) \cdot \frac{1}{A(\mathrm{H_2O})} \cdot \rho(\mathrm{H_2O}) \cdot \left[\frac{A(\mathrm{Cm})}{C(\mathrm{Cm})} - \frac{A(\mathrm{CmO_2})}{\rho(\mathrm{CmO_2})} \right],$$

$N(\mathrm{H}) = 2 \cdot N(\mathrm{H_2O})$, and

$N(\mathrm{O}) = N(\mathrm{H_2O}) + 2 \cdot N(\mathrm{Cm})$.

References to Appendix A

(A1) (Tr.) Y. Naito and H. Okuno, "Nuclear Criticality Safety Handbook (English Translation)," *JAERI-Review* 95-013 (1995).

(A2) B.B. Cunningham and J.C. Wallmann, "Crystal Structure and Melting Point of Curium Metal," *J. Inorg. Nucl. Chem.*(1964) **26**, 271-5.

(A3) J. J. Katz, G. T. Seaborg and L. R. Morss, *The Chemistry of Actinide Elements, Second edition, Volume 2*, Chapman and Hall, London (1986).

Appendix B. Effect of ^{244}Cm on Minimum Critical and Subcritical Masses of ^{245}Cm

The American National Standard ANSI/ANS-8.15-1981 shows a table on subcritical mass limits for ^{245}Cm in (^{244}Cm + ^{245}Cm) for uniform water-reflected CmO_2-H_2O mixtures, as reproduced in Table B1. The critical ($k_{eff} = 1$) and subcritical ($k_{eff} = 0.9$ and 0.8) masses were calculated with less data points, i.e., for ^{245}Cm contents of 100, 5, 1.25 wt%, whereas in Table B1, 10 and 2.5 wt% enriched ^{245}Cm cases were also included. A graph is drawn with these values in Fig. B1 in comparison with the values indicated in Table B1. It reveals that ANSI's values are less than our results corresponding to $k_{eff} = 0.8$. The differences become larger as the ^{245}Cm contents become smaller. This fact suggests that the subcritical masses of ^{244}Cm may be larger than those described in ANSI/ANS-8.15-1981.

Table B1. Subcritical Mass Limits for ^{245}Cm in (^{244}Cm + ^{245}Cm) for Uniform Water-Reflected CmO_2-H_2O Mixtures, according to in ANSI/ANS-8.15-1981

^{245}Cm (wt%)	Mass Limit	
	^{245}Cm (g)	Total Quantity of Cm (g)
100	30.0	30
10	32.7	327
5	35.7	714
2.5	41.7	1670
1.25	53.7	4300

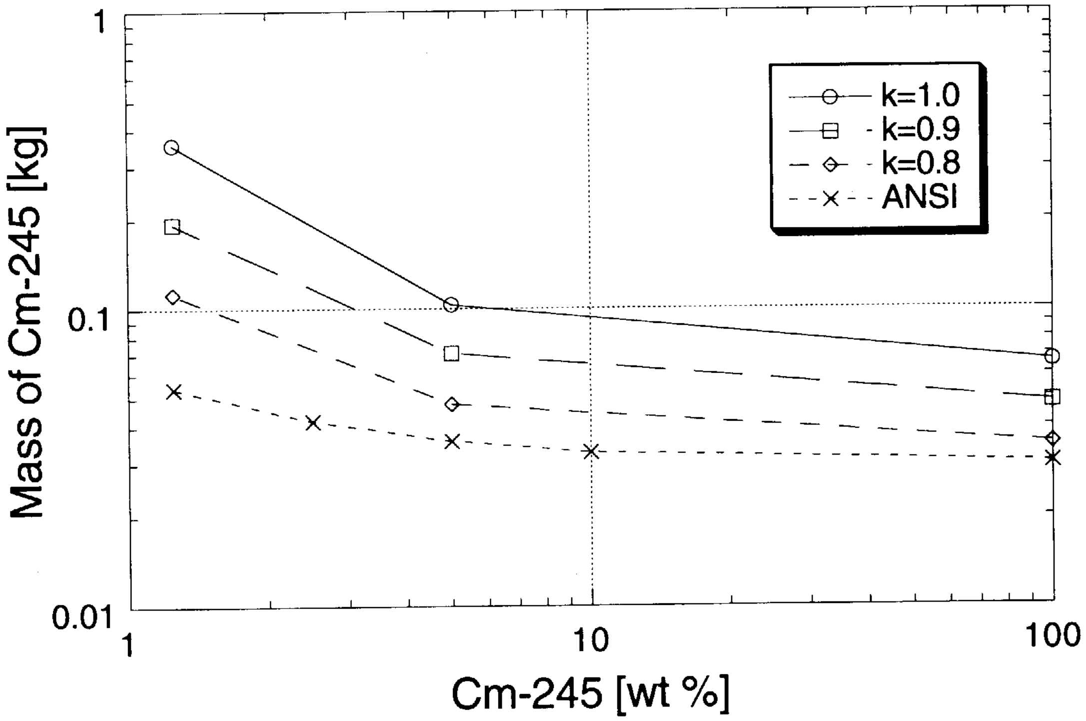

Fig. B1. Critical and subcritical masses of ^{245}Cm in (^{244}Cm + ^{245}Cm) for Uniform Water-Reflected CmO_2-H_2O Mixtures compared with the subcritical mass limits found in ANSI/ANS-8.15-1981

Appendix C. A Brief Review of Studies on Critical Masses of Curium Isotopes

There have been several studies made concerning criticality mass data on curium (Cm) isotopes since the publication of ANSI/ANS-8.15 in 1981. This appendix summarizes their findings.

Srinivasan et al. (C1)

Systematic relations were deduced by analyzing the criticality data of special actinides. Criticality data were calculated with a combination of nuclear data libraries based on ENDL-82 and DTF-IV transport theory code. Critical masses of Cm isotopes with and without iron reflector are shown in Table C1. Table C2 shows the minimum critical masses of odd-N Cm isotopes in solution with and without water reflector.

Roussignol(C2)

Critical masses of Pu-241, Am-242m, Cm-243 and -245 isotopes were calculated with APPOLO-I and DTF-IV (Nuclear data libraries are not specified in the literature). The minimum critical mass, admissible mass (70% of the minimum critical mass) and the corresponding ANSI value for Cm-243 and -245 are compared in Table C3. The 20-cm-thick water reflector was assumed in the calculation.

Rahn(C3)

A nuclear criticality safety evaluation was performed to establish conditions for which the contents of an F Canyon tank would remain subcritical. As a check of the calculation made with GLASS-ANISN, the minimum critical mass of Cm-245 was calculated to find 42.5 g (see Fig. C1). He used a data library based on ENDF/B-III.

Komuro et al. (C4)

Criticality data for actinides were calculated with a combination of a 137-grouped neutron cross section library MGCL-J3 and a Monte Carlo neutron transportation code MULTI-KENO-3.0. Search calculations were made for $k_{eff} = 1.0 \pm 0.5\sigma$. Typical values for σ were from 0.003 to 0.005. The critical masses obtained for Cm isotopes were summarized in Table C4.

Nojiri and Fukasaku (C5)

Critical masses of curium isotopes in metal and in dioxide without reflector, with water and with Stainless Steel (SUS) in Type 304 were calculated in three ways, SCALE 4.3 (44Gr.ENDF/V + KENO V.a), MCNP-4A combined with JENDL-3.2 and with ENDL-85 libraries. The results are summarized in three tables, from Tables C5 to C7, according to the calculation methods applied.

Anno (C6)

Critical masses of curium isotopes in metal were calculated with Apollo2 - Sn (172-energy-grouped CEA 93 Library, condensed in 20 energy groups for Sn). The critical masses from ^{242}Cm to ^{247}Cm were obtained with various reflector conditions, and the results are shown in Table C8. In addition, useful formulae were derived on actinides between ^{232}U to ^{247}Cm that approximately show relationships between reflected (y) and bare (x) critical mass (in kg):

water: $y = 0.7743x + 0.0006x^2$,
concrete: $y = 0.6495x + 0.0005x^2$,
steel: $y = 0.4997x + 0.0003x^2$,
lead + water: $y = 0.5159x - 0.0001x^2$.

References to Appendix C

(C1) M. Srinivasan, K. Subba Rao, S.B. Garg and G.V. Acharya, "Systematics of Criticality Data of Special Actinides Deduced Through the Trombay Criticality Formula," *Nucl. Sci. and Eng.*, **102**, 295 (1989).

(C2) M. Roussignol, "Détermination des Masses Critique Relatives à Certains Actinides Fissiles; Criticité de l'Américium et du Curium Issus de Combustibles REP Irradiees", SEC/T/208/94.332 (1994) [in French].

(C3) R.R. Rahn, "Criticality Safety Evaluation of Mixtures Containing Americium and Curium," *Proc. of the Fifth Intl. Conf. on Nucl. Criticality Safety, ICNC'95*, Vol.2, 11•101, Sept. 17 - 21, 1995, Albuquerque (1995); K. Yates, private communication.

(C4) Y. Komuro, T. Takada and T. Arakawa, "Estimation of Critical Mass for Actinoids," *1995 Fall Mtg. of the Atomic Energy Soc. of Japan*, Oct. 17-20, 1995, Tokai-mura, B55 (1995) [Abstract form only, in Japanese].

(C5) I. Nojiri and Y. Fukasaku, "Calculational Study for Criticality Safety Data of Fissionable Actinides," *Proc. of the Intl. Conf. Future Nuclear Systems, Global '97*, Vol.2, 1397, Oct. 5 - 10, 1997, Yokohama (1997).

(C6) J. Anno, "Données de criticité des actinides fissiles en métal et/ou en solution aqueuse sous forme de sphères nues et réfléchies," SEC/T/99.196 (1999) [in French].

Table C1. ***Critical masses of Cm isotopes obtained by Srinivasan et al.***

Nuclide	Critical Mass [kg]	
	Bare	Iron reflected*1
Cm-243	8.10	3.28
Cm-244	21.2	10.59
Cm-245	12.28	5.80
Cm-247	7.87	3.36

*1 30-cm-thick

Table C2. ***Minimum critical masses of Cm isotopes in solution obtained by Srinivasan et al.***

Nuclide	Concentration [gCm/L]	H/Cm	Critical Mass [g]	
			Bare	Water reflected*1
Cm-243	40	672	739	382
Cm-245	12	2267	136	62
Cm-247	350	76	5048	2728

*1 20-cm-thick

Table C3. ***Minimum critical mass and admissible maximum mass of Cm isotopes calculated by Roussignol***

Nuclide	Minimum Critical Mass [g]	Maximum Admissible Mass [g]	Maximum Admissible Mass of ANSI [g]
Cm-243	116	81.2	90
Cm-245	42.7	29.8	30

Table C4. ***Critical masses of Cm isotopes obtained by Komuro et al.***

Nuclide	Critical Mass [kg]	
	Bare	Water reflected*1
Cm-242	15.4	10.4
Cm-243	9.72	3.35
Cm-244	27.6	22.8
Cm-245	12.4	3.78
Cm-247	7.00	2.91

*1 30-cm-thick

Table C5. Criticality masses calculated with SCALE4.3 (LIB:44-Gr. ENDF/B-V) by Nojiri and Fukasaku

Reflector	Reflector thickness [cm]	Cm mass [kg]			CmO_2 mass [kg]		
		Cm-245	Cm-246	Cm-247	Cm-245	Cm-246	Cm-247
None	---	9.33	37.9	7.15	11.8	67.0	9.21
Water	2.5	5.53	34.7	5.21	7.03	62.5	67.0
	30	3.14	32.9	3.66	3.85	58.8	4.70
SUS (Type304)	10	4.32	21.6	3.43	5.64	43.1	4.60
	20	3.79	20.5	3.11	4.82	41.7	4.05
	30	3.59	20.5	2.94	4.57	40.2	3.89

Table C6. Criticality masses calculated with MCNP-4A combined with JENDL-3.2 by Nojiri and Fukasaku

Reflector	Reflector thickness [cm]	Cm mass [kg]			CmO_2 mass [kg]		
		Cm-245	Cm-246	Cm-247	Cm-245	Cm-246	Cm-247
None	---	12.4	70.0	7.25	14.6	126	9.15
Water	2.5	7.30	64.1	4.76	8.90	116	6.22
	30	3.59	58.6	3.01	4.42	104	3.89
SUS (Type304)	10	5.77	40.8	3.67	7.29	83.1	4.88
	20	5.10	39.5	3.32	6.51	79.2	4.43
	30	4.80	38.7	3.15	6.13	77.6	4.21

Table C7. Criticality masses calculated with MCNP-4A combined with ENDL-85 by Nojiri and Fukasaku

Reflector	Reflector thickness [cm]	Cm mass [kg]			CmO_2 mass [kg]		
		Cm-245	Cm-246	Cm-247	Cm-245	Cm-246	Cm-247
None	---	12.5	84.1	7.88	14.6	140	9.79
Water	2.5	7.40	73.3	5.36	8.98	126	6.94
	30	3.86	65.8	3.71	4.78	114	4.97
SUS (Type304)	10	5.77	48.5	3.88	7.16	89.0	5.18
	20	4.98	45.0	3.54	6.26	86.2	4.67
	30	4.77	44.7	3.42	5.95	84.9	4.50

Table C8. Critical masses of Cm isotopes obtained by Anno

Nuclide	Critical Mass [kg]				
	Bare	Steel reflected*1	Water reflected*2	Concrete reflected*3	Lead + Water reflected*4
Cm-242	25.152				
Cm-243	7.336	2.758	2.829	2.939	3.390
Cm-244	32.965	16.007	26.871	21.605	15.479
Cm-245	6.809	2.657	2.607	2.620	3.206
Cm-246	42.529				
Cm-247	7.206				

*1 30 cm thick

*2 20 cm thick

*3 60 cm thick

*4 25-cm-thick lead + 20-cm-thick water

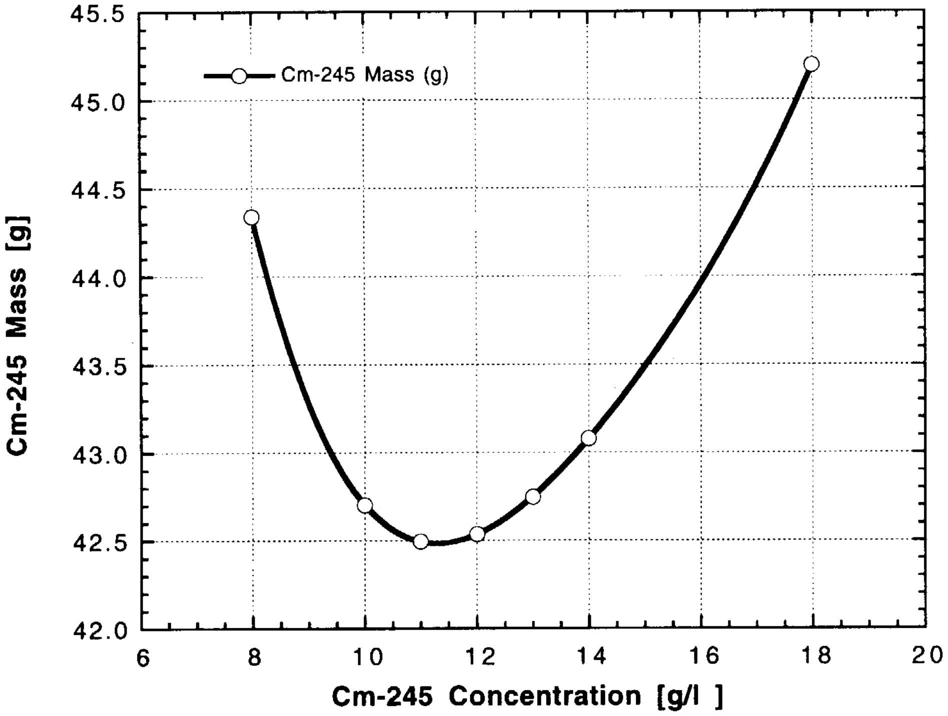

Fig. C1. Rahn's result for the minimum critical mass of Cm-245

Appendix D. Comparison of Evaluated Nuclear Data for ^{245}Cm, ^{246}Cm and ^{247}Cm

T. Nakagawa et al. published a report summarizing the present status of evaluated nuclear data of minor actinides: ^{237}Np, ^{241}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm and ^{245}Cm(D1). By courtesy of T. Nakagawa, figures comparing evaluated nuclear data for ^{245}Cm are cited in this Appendix. He is kind enough to produce also similar figures for ^{246}Cm and ^{247}Cm.

References to Appendix D

(D1) T. Nakagawa, H. Takano and A. Hasegawa, "Present Status of Minor Actinide Data," *NEA/WPEC*-8 (1999).

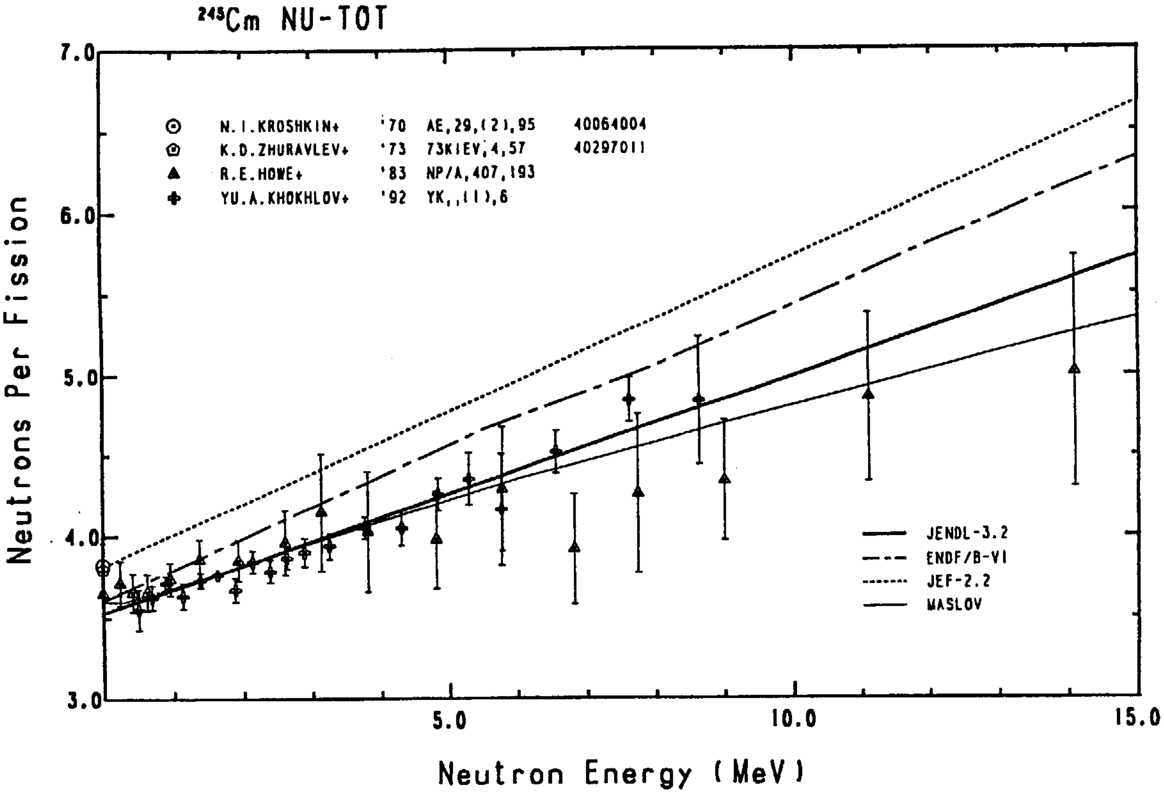

Fig. D1. ^{245}Cm number of neutrons per fission

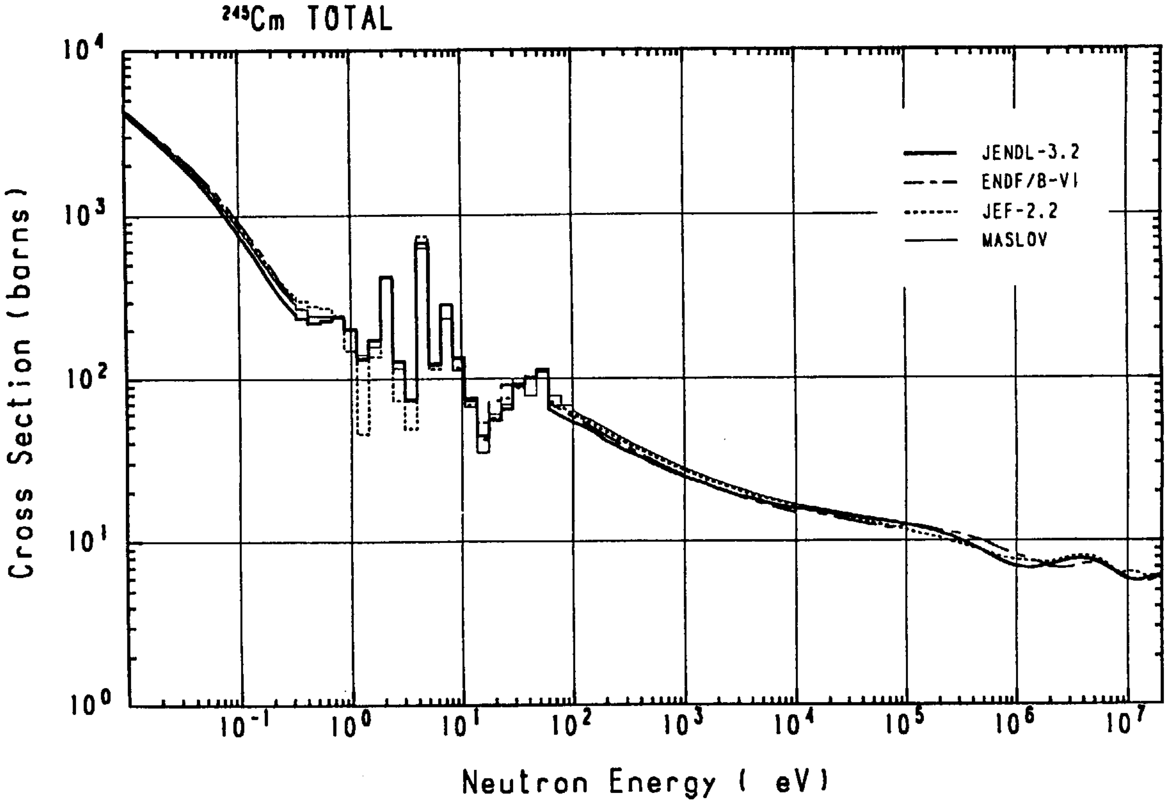

Fig. D2. ^{245}Cm total cross section

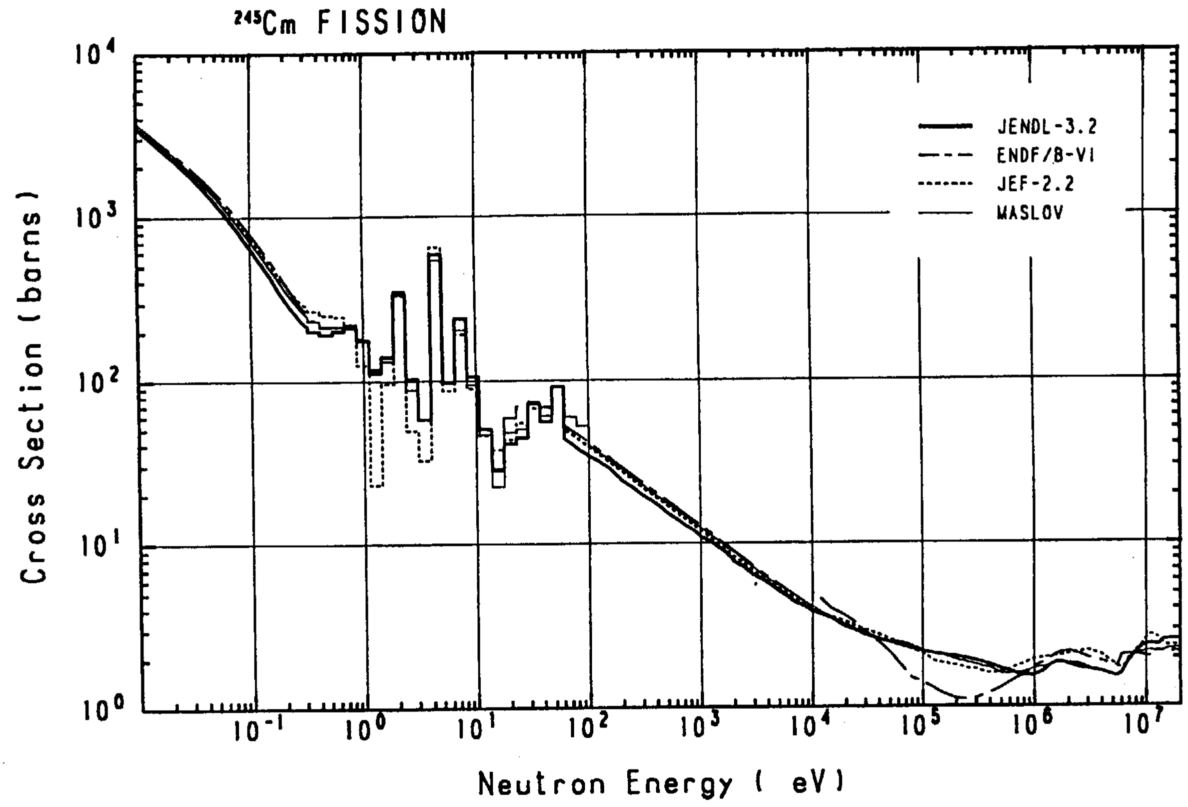

Fig. D3. ^{245}Cm fission cross section

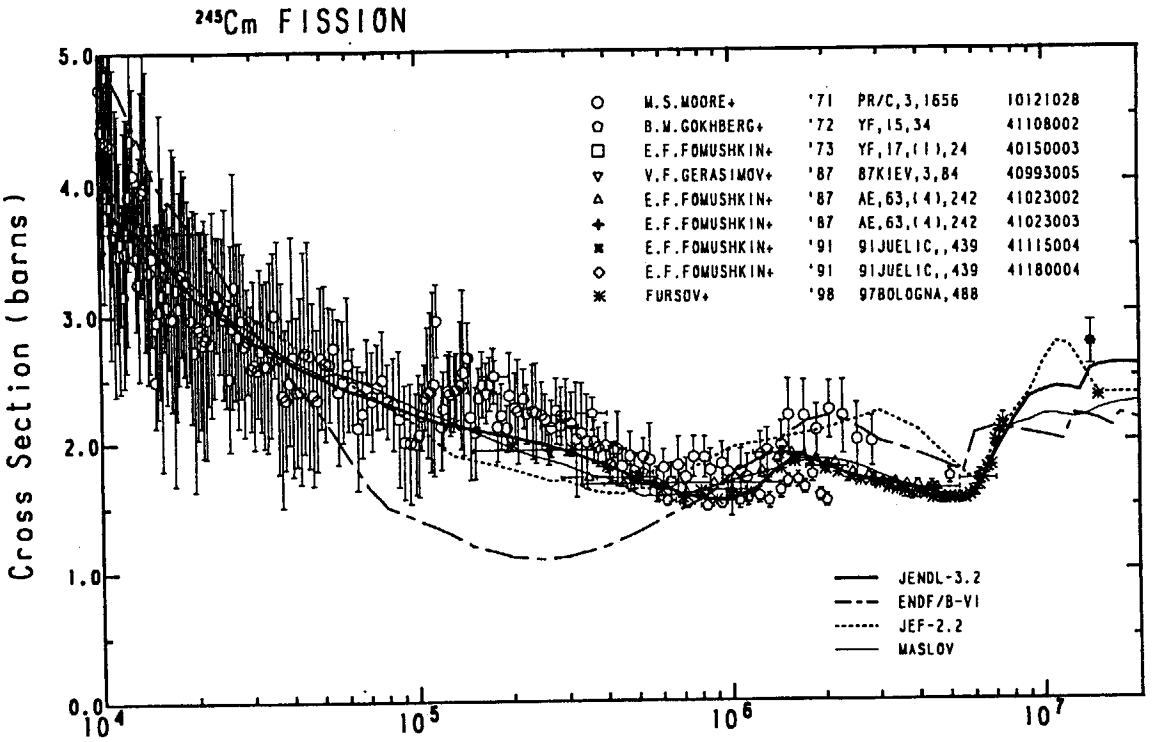

Fig. D4. ^{245}Cm fission cross section compared with experimental data in the neutron energy range above 10 keV

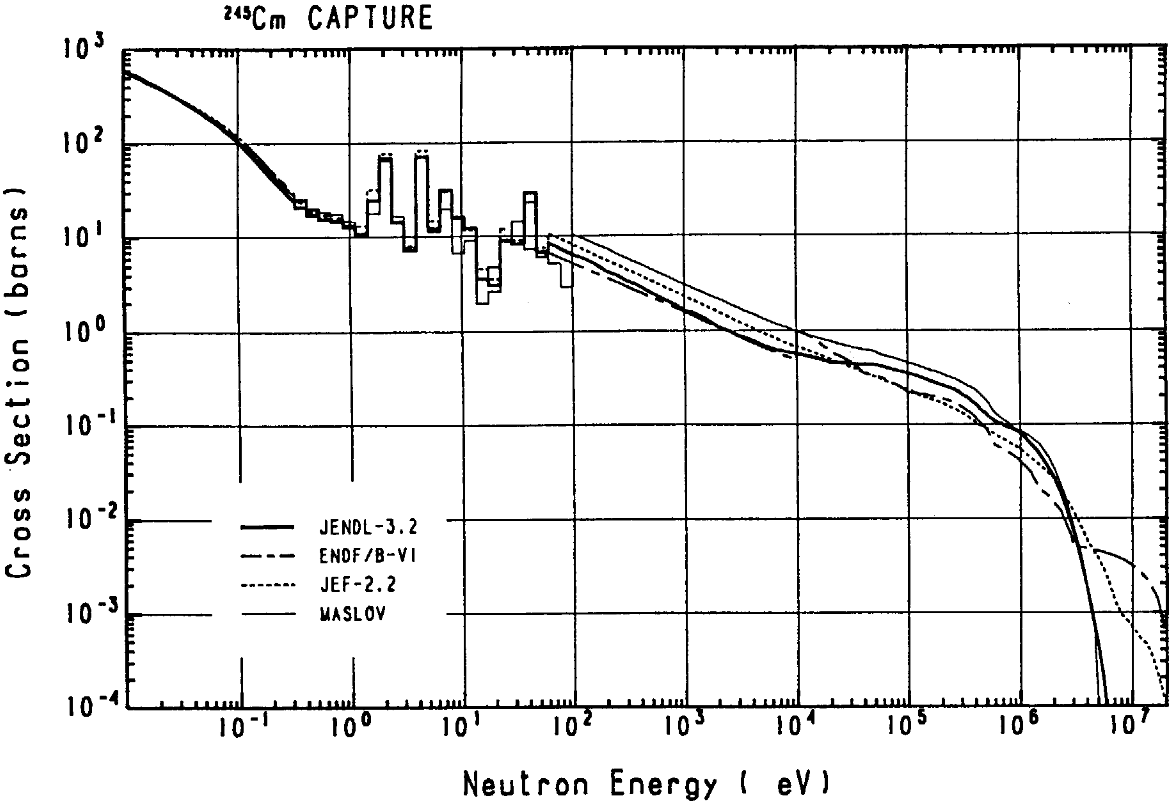

Fig. D5. ^{245}Cm capture cross section

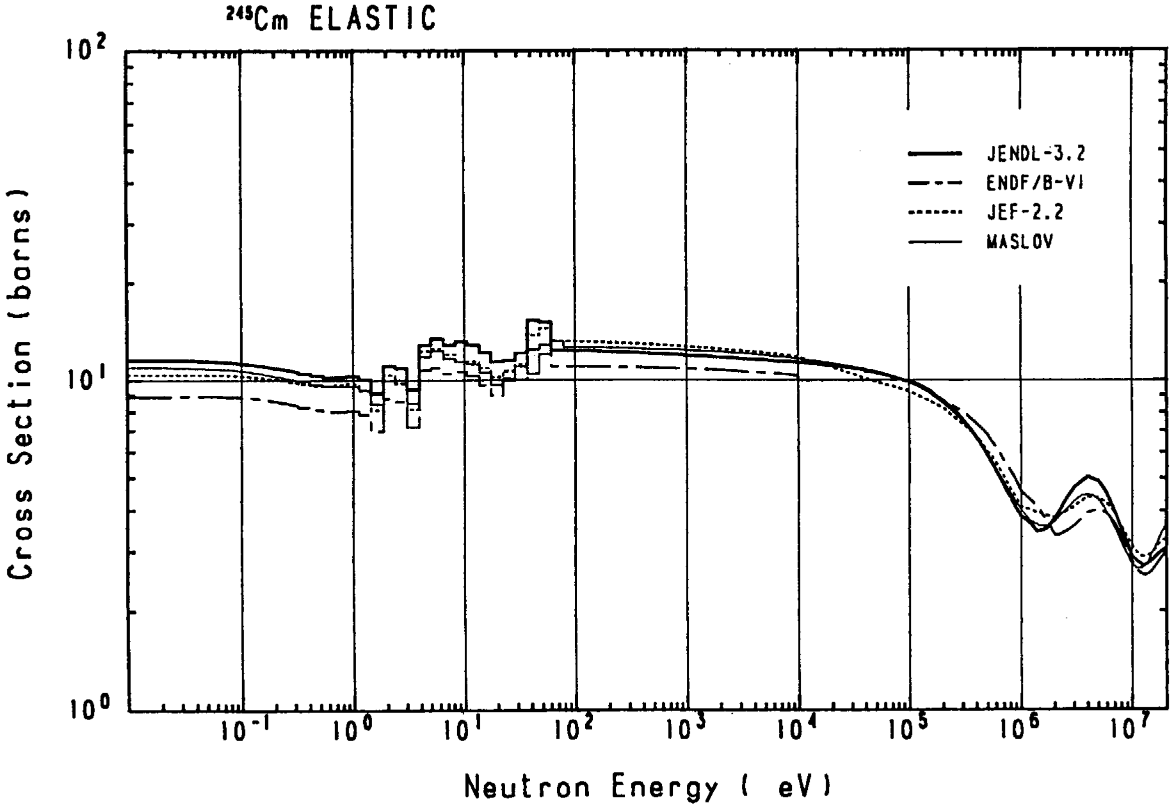

Fig. D6. ^{245}Cm elastic scattering cross section

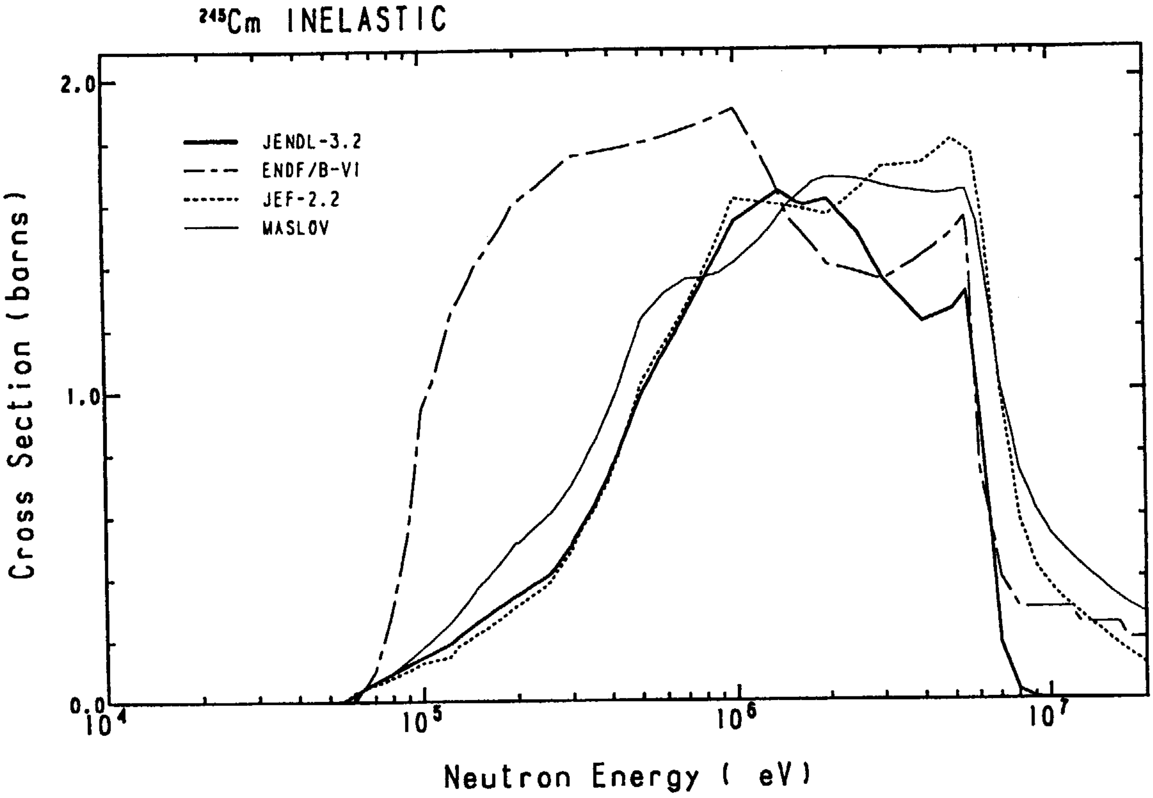

Fig. D7. ^{245}Cm total inelastic scattering cross section

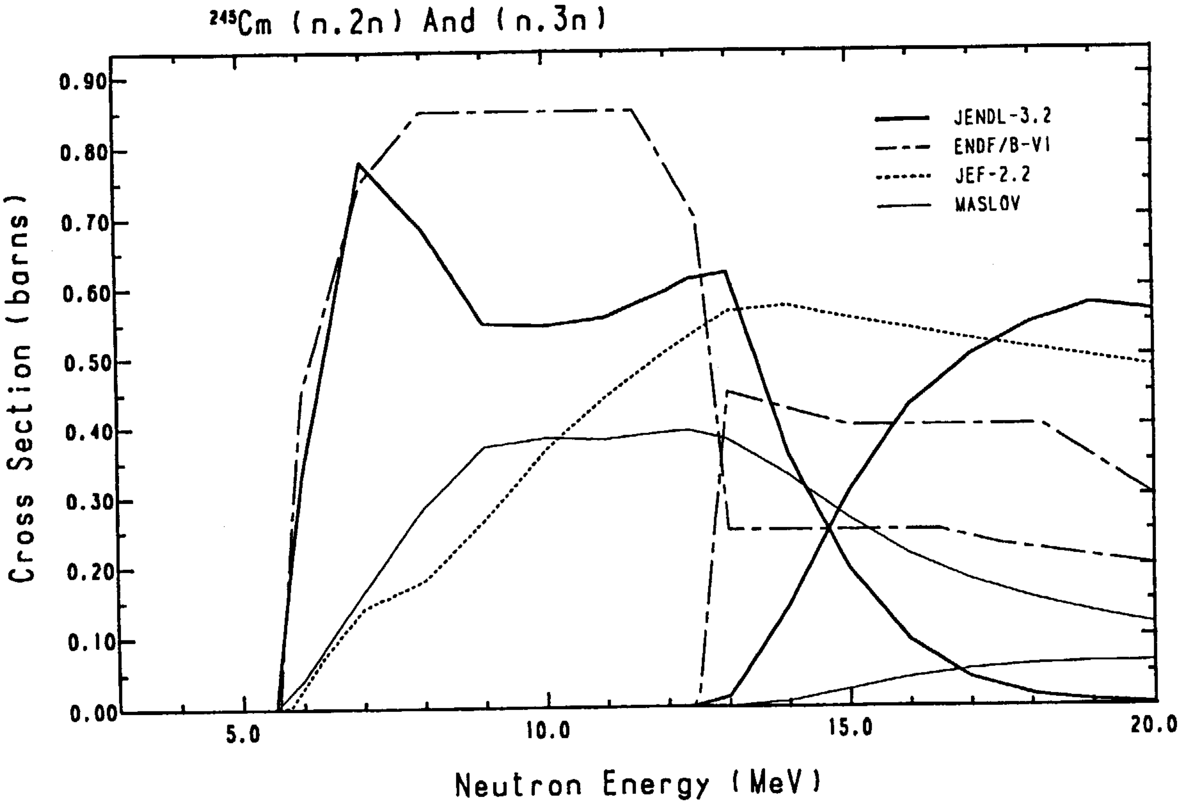

Fig. D8. ^{245}Cm(n,2n) and (n,3n) cross sections

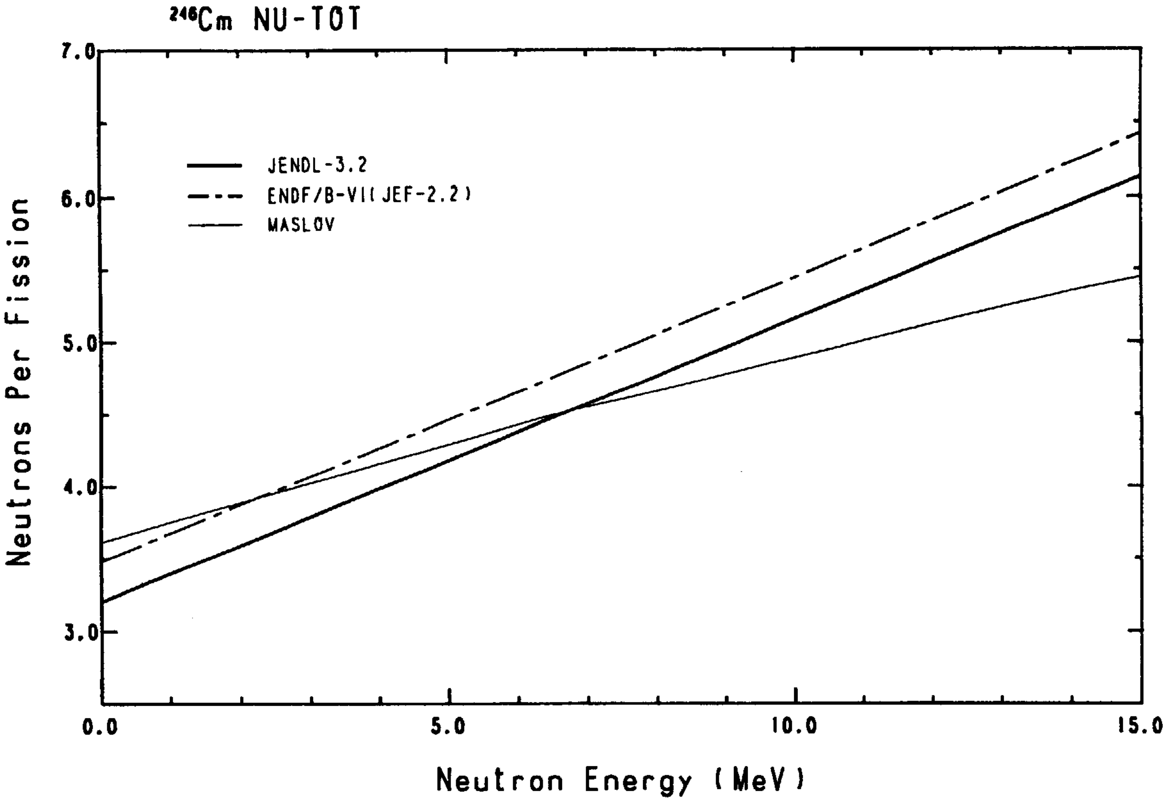

Fig. D9. ^{246}Cm number of neutrons per fission

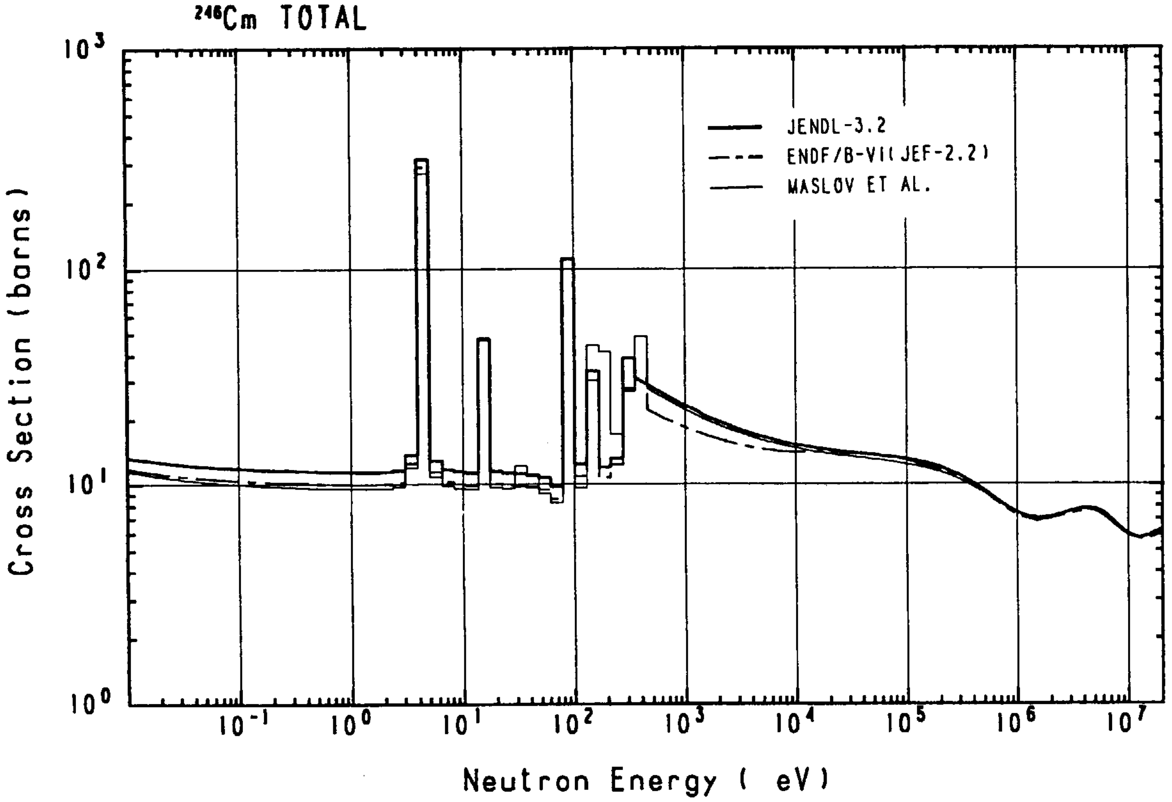

Fig. D10. ^{246}Cm total cross section

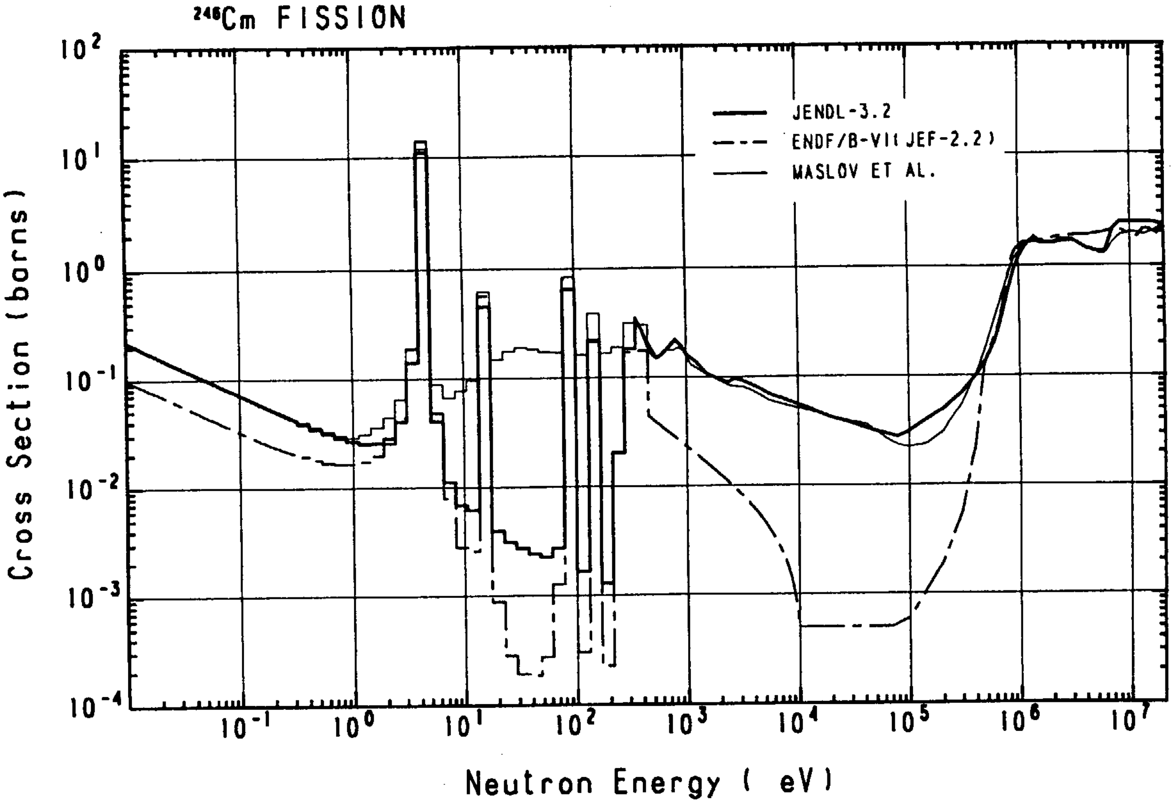

Fig. D11. ^{246}Cm fission cross section

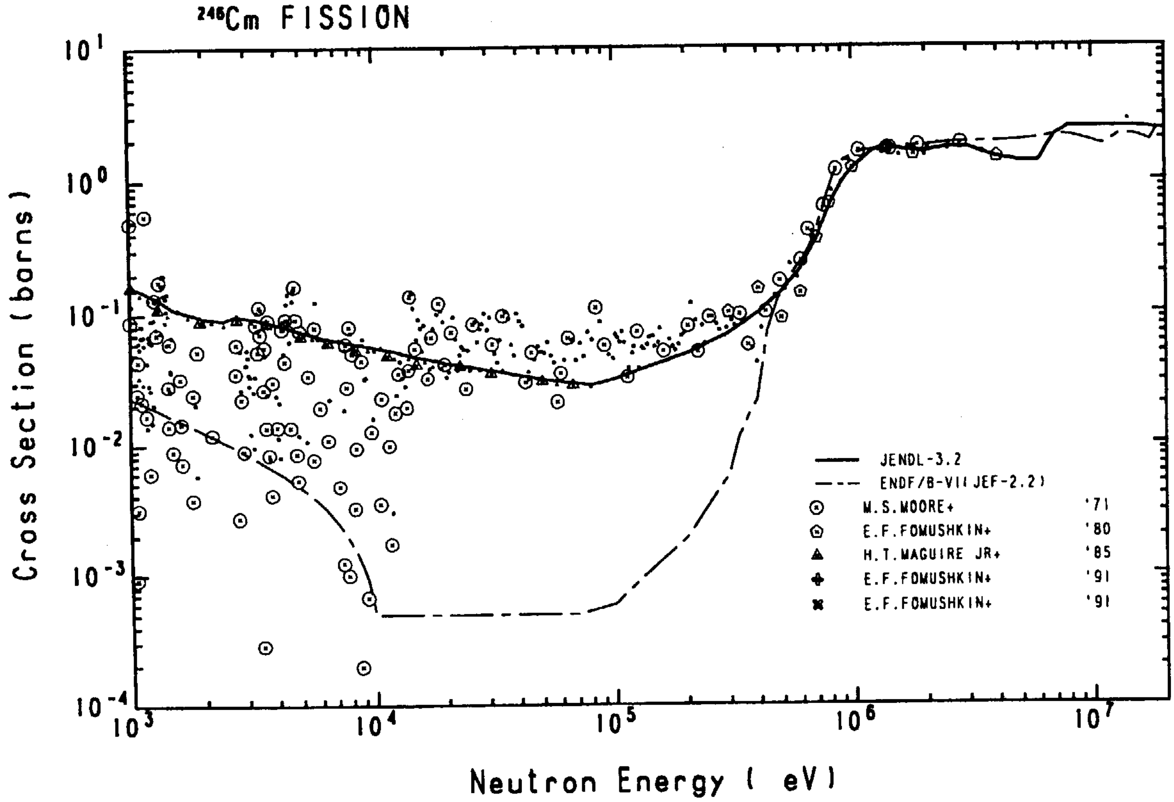

Fig. D12. ^{246}Cm fission cross section compared with experimental data in the neutron energy range above 1 keV

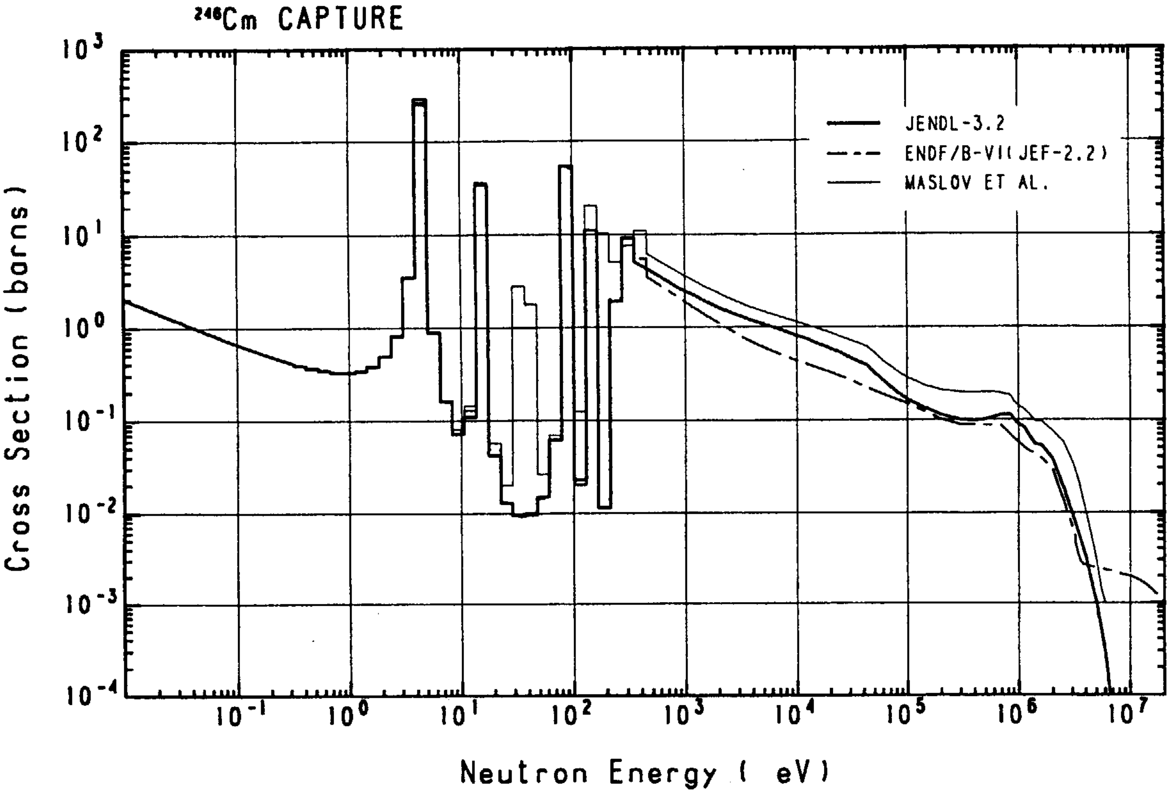

Fig. D13. ^{246}Cm capture cross section

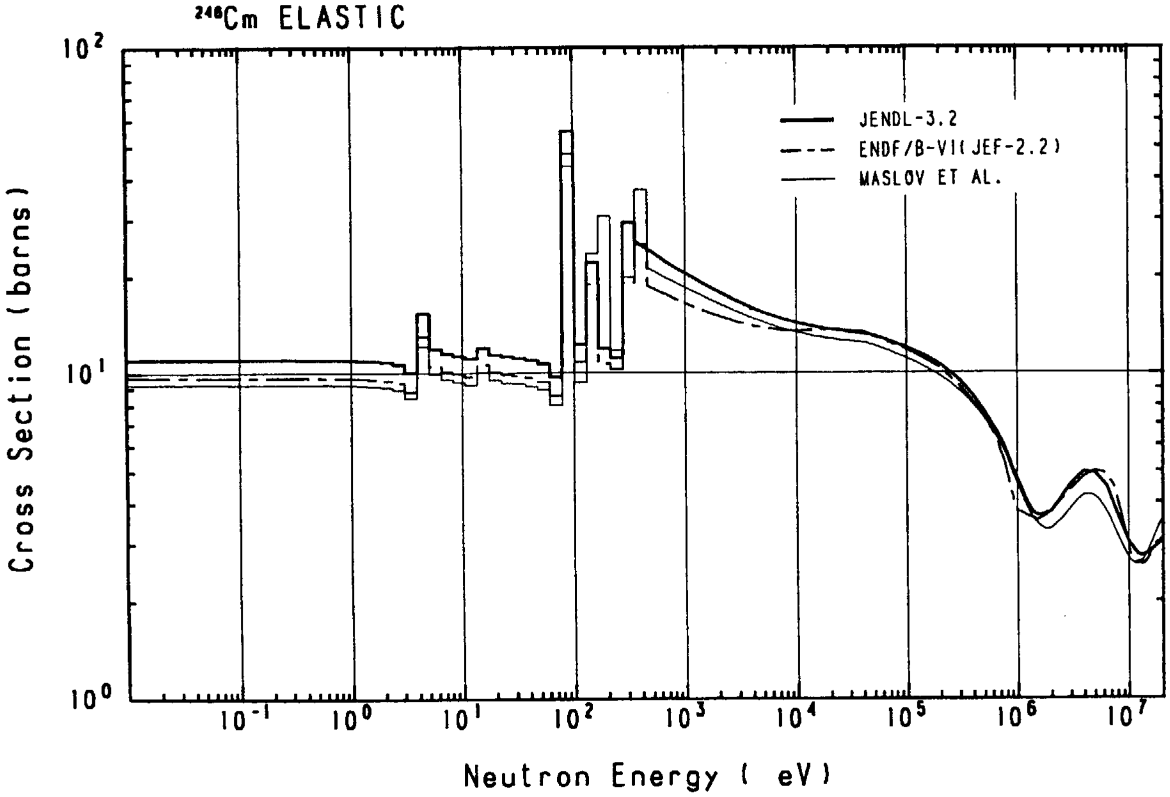

Fig. D14. ^{246}Cm elastic scattering cross section

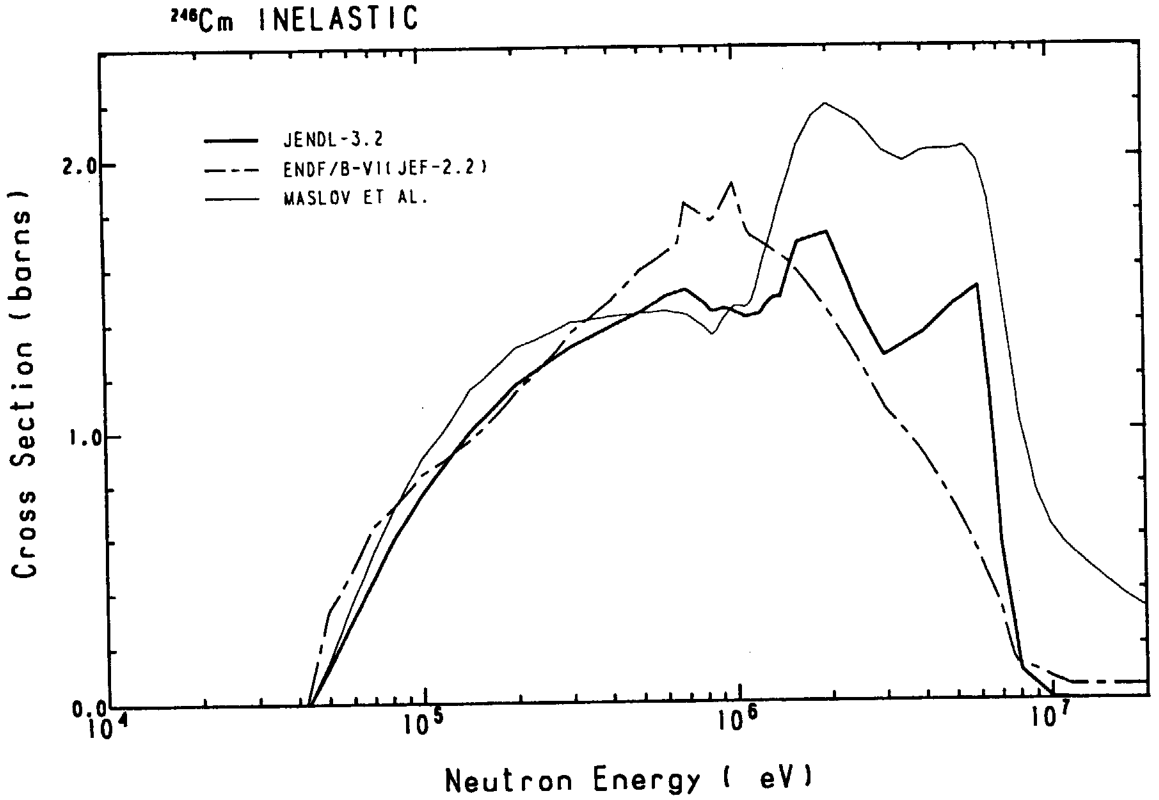

Fig. D15. ^{246}Cm total inelastic scattering cross section

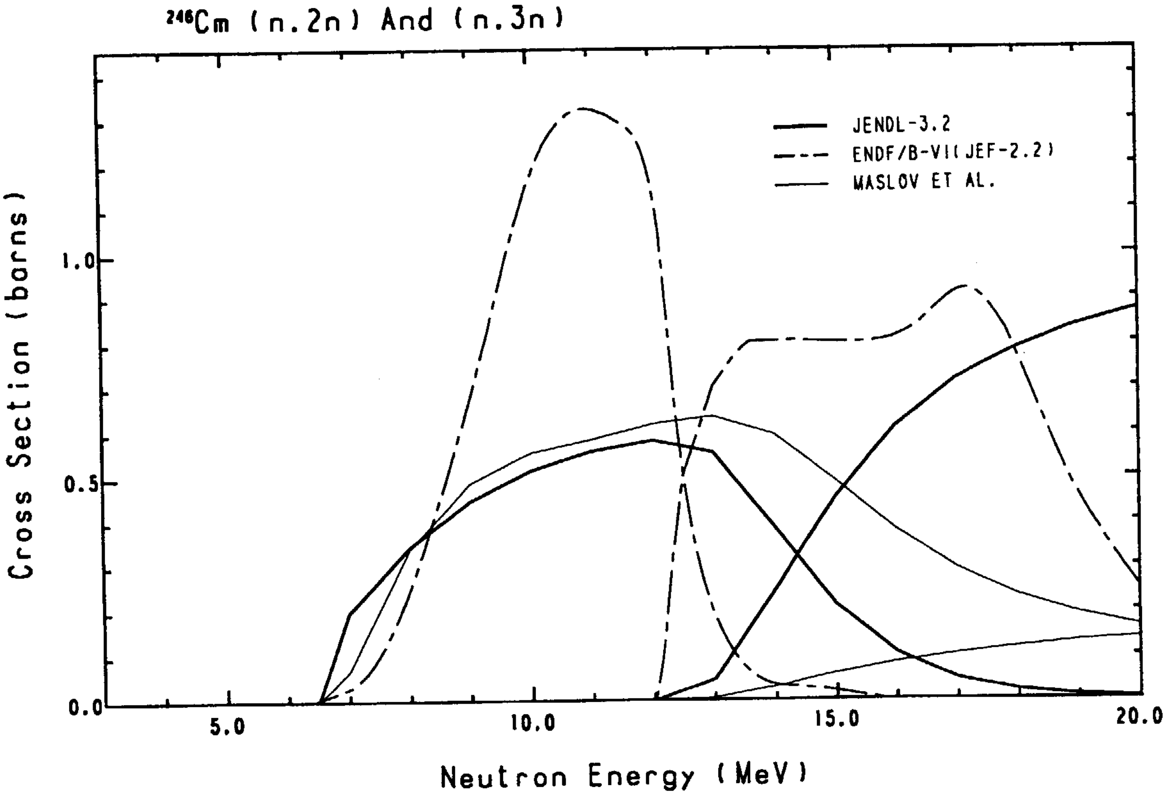

Fig. D16. ^{246}Cm(n,2n) and (n,3n) cross sections

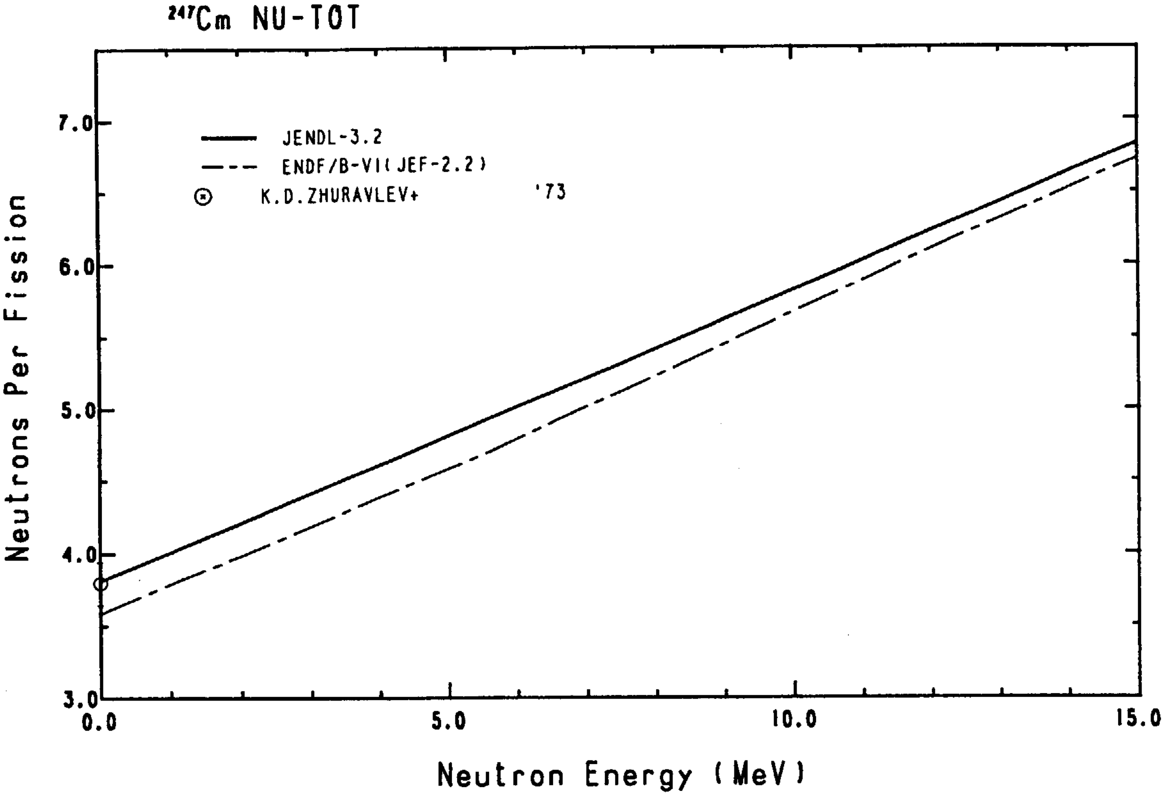

Fig. D17. ^{247}Cm number of neutrons per fission

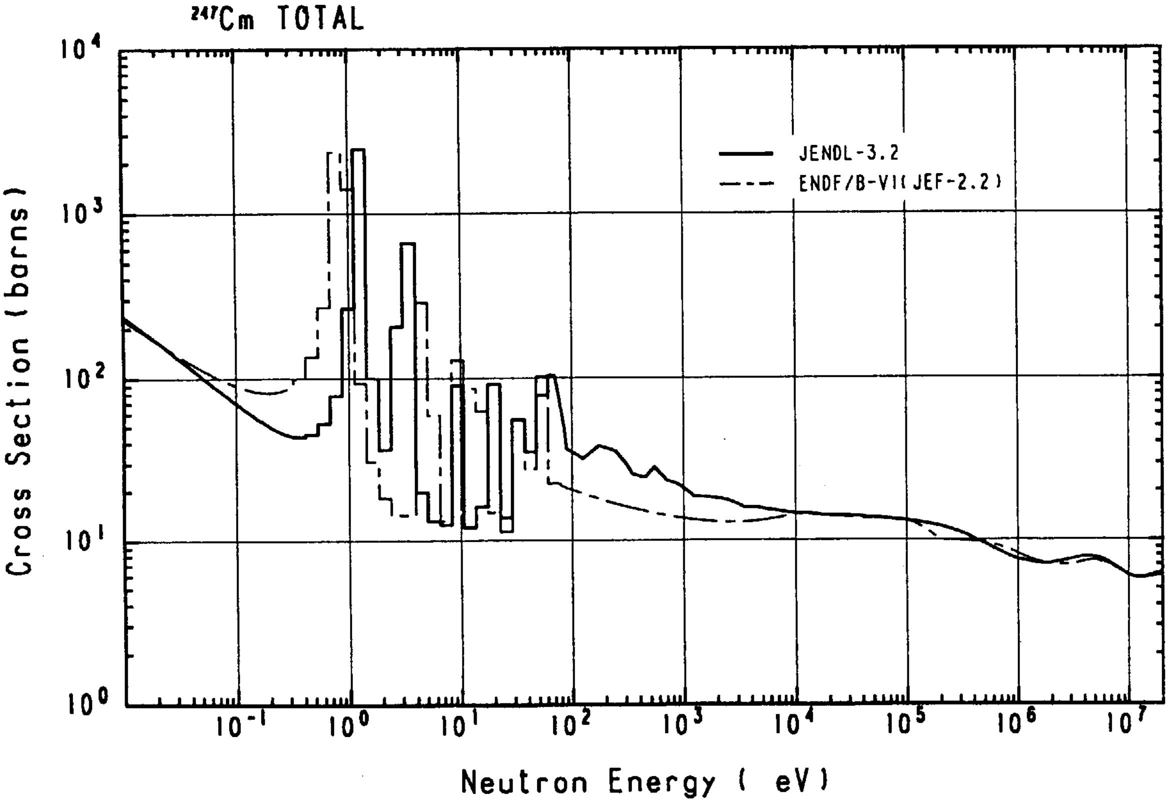

Fig. D18. ^{247}Cm total cross section

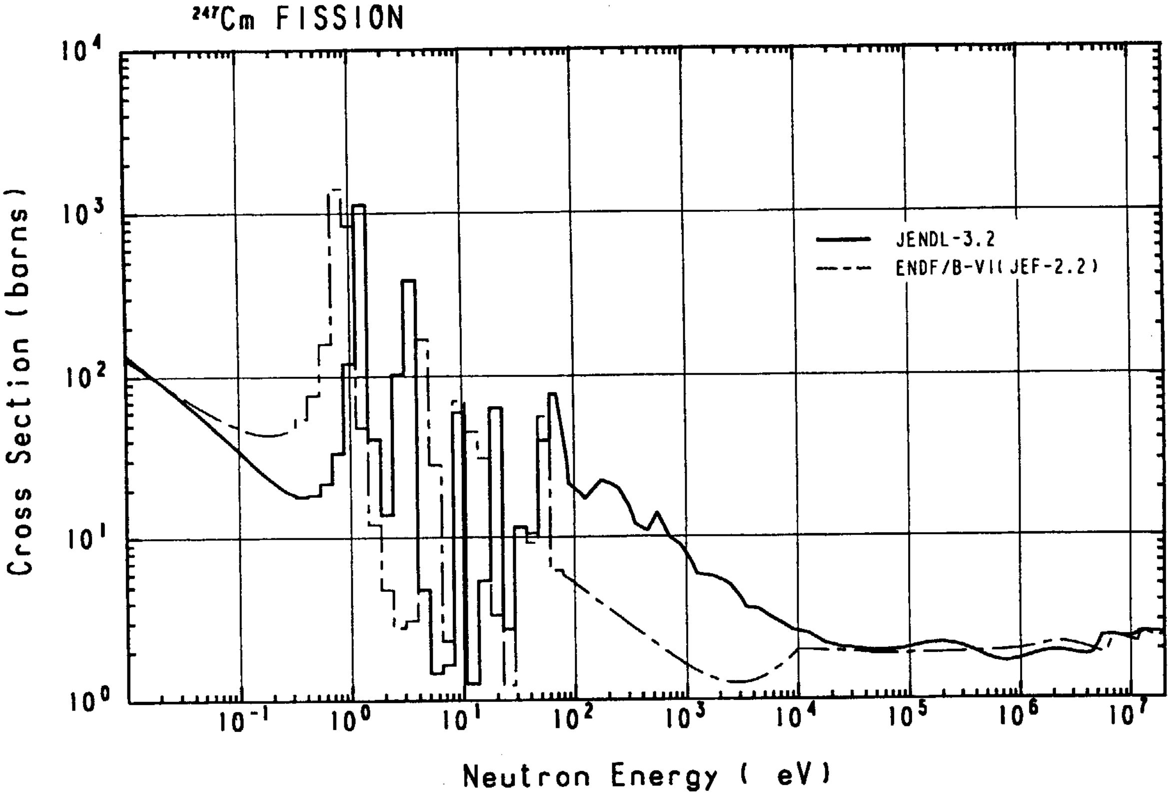

Fig. D19. ^{247}Cm fission cross section

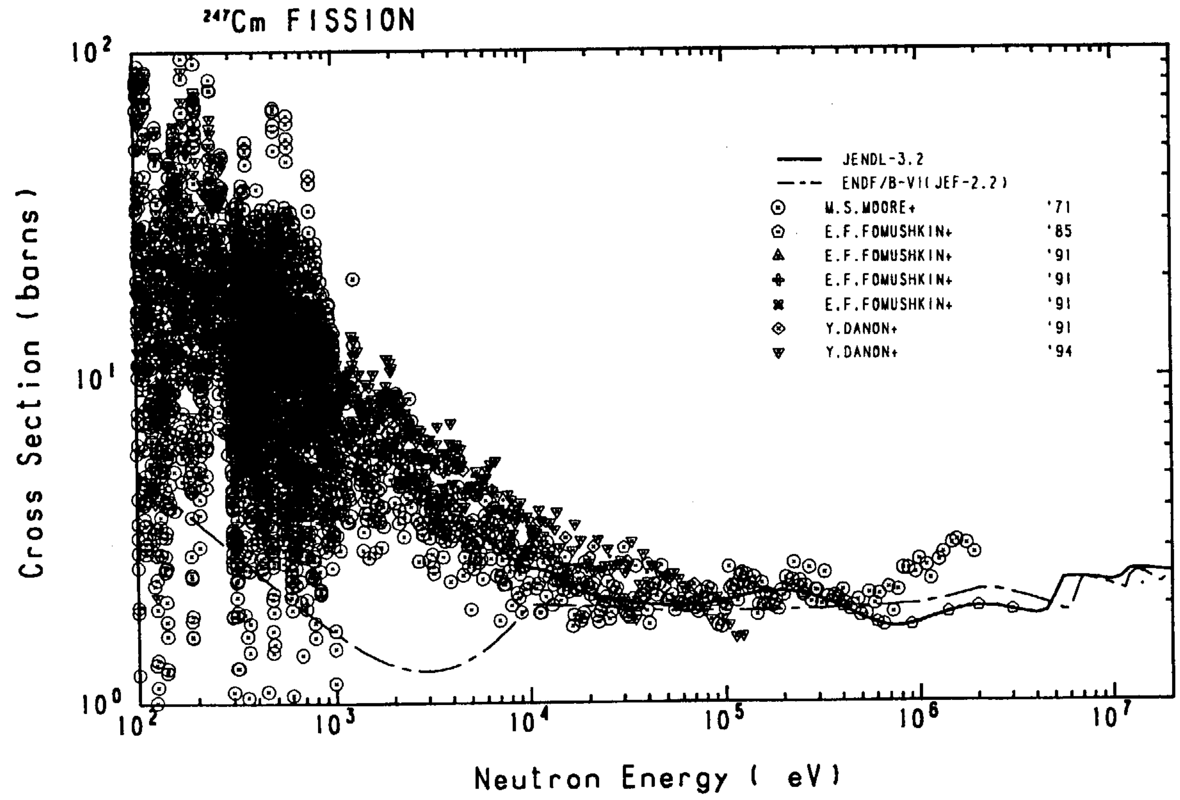

Fig. D20a. ^{247}Cm fission cross section compared with experimental data in the neutron energy range above 100 eV

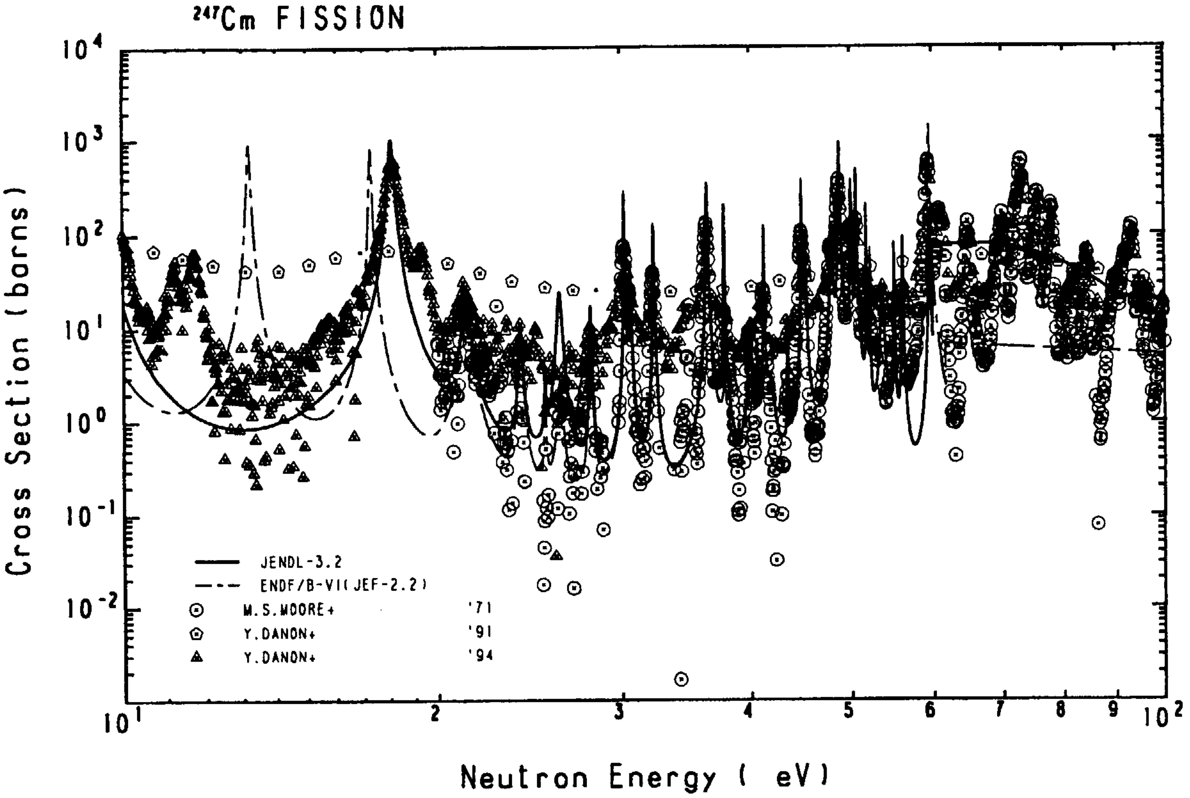

Fig. D20b. ^{247}Cm fission cross section compared with experimental data in the neutron energy range from 10 eV to 100 eV

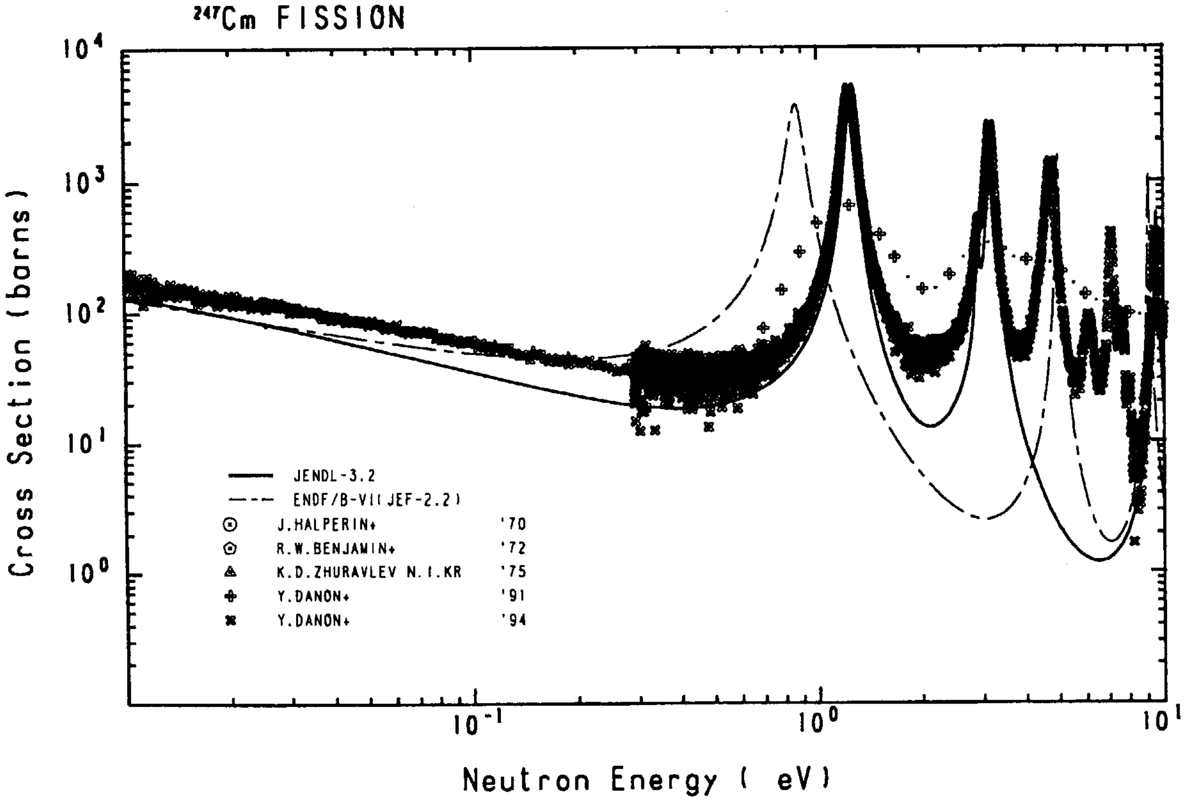

Fig. D20c. ^{247}Cm fission cross section compared with experimental data in the neutron energy range below 10 eV

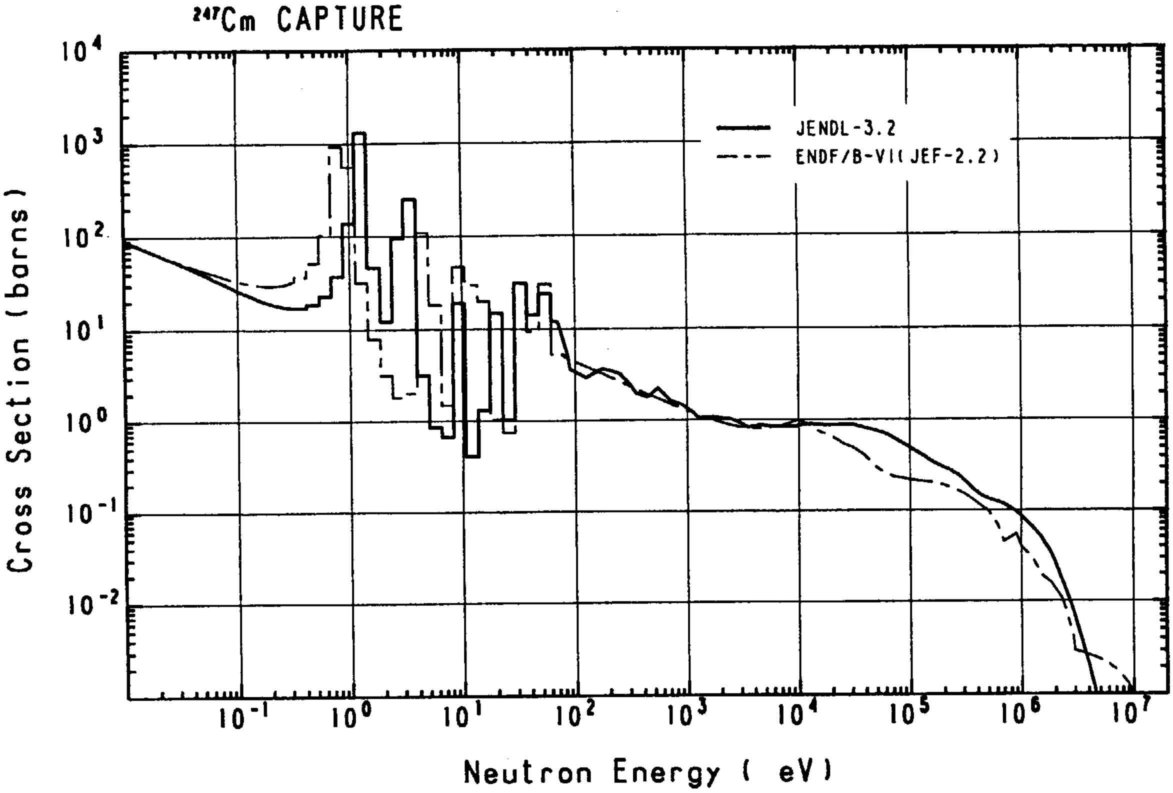

Fig. D21. ^{247}Cm capture cross section

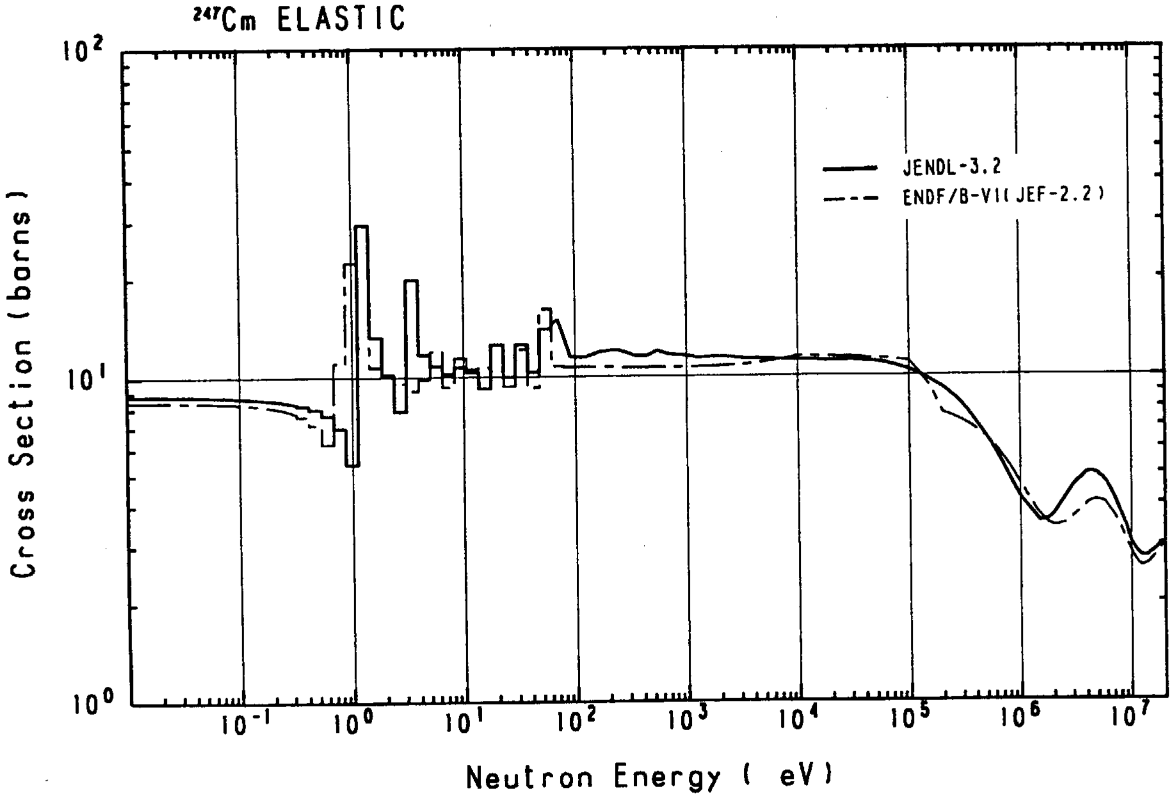

Fig. D22. ^{247}Cm elastic scattering cross section

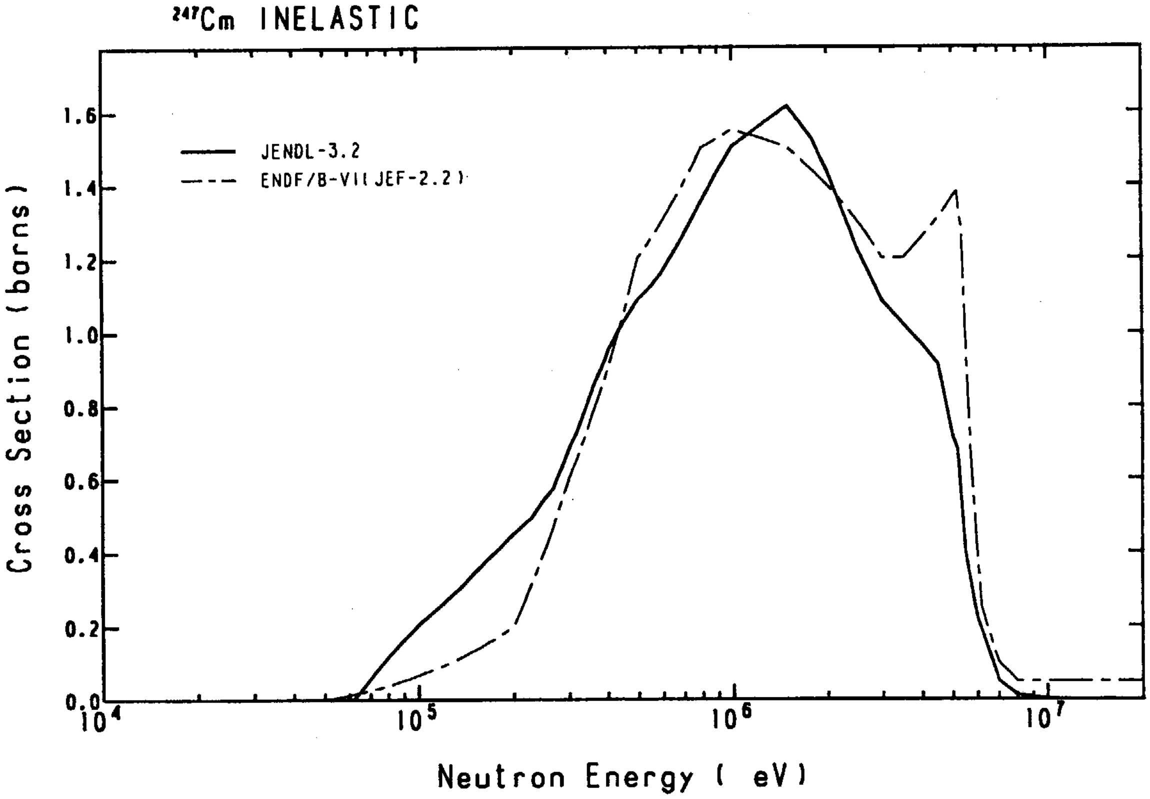

Fig. D23. ^{247}Cm total inelastic scattering cross section

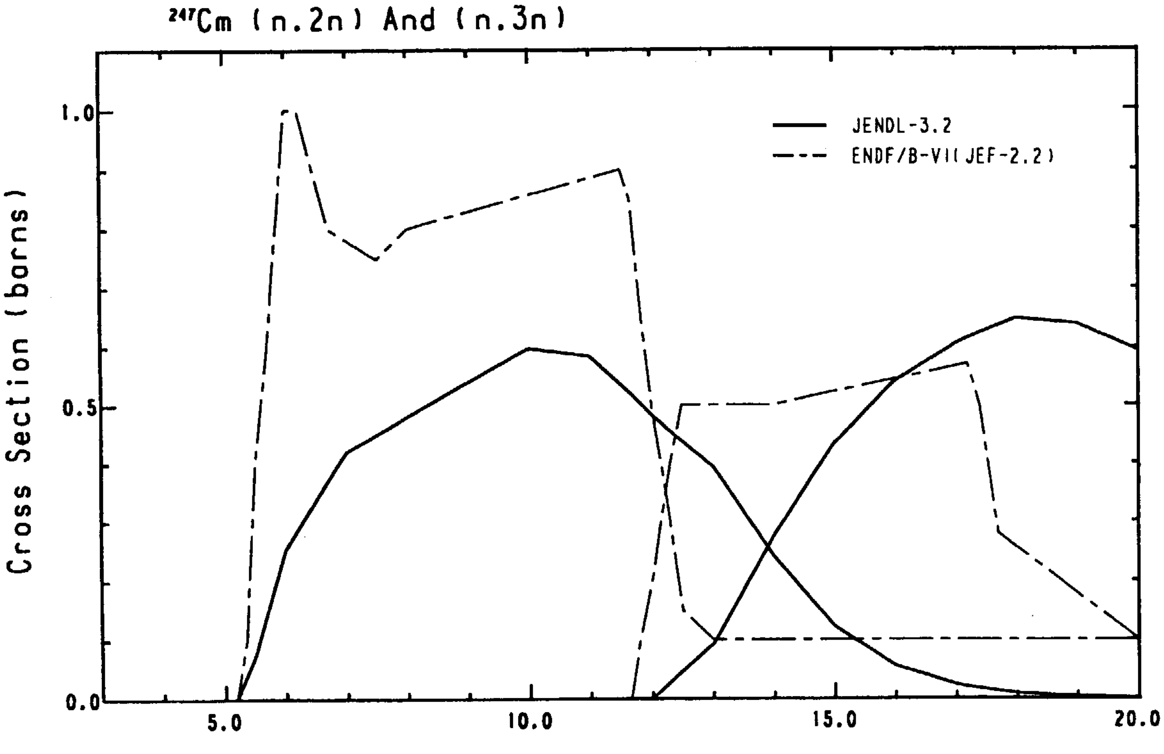

Fig. D24. ^{247}Cm(n,2n) and (n,3n) cross sections

国際単位系（SI）と換算表

表 1　SI基本単位および補助単位

量	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd
平面角	ラジアン	rad
立体角	ステラジアン	sr

表 3　固有の名称をもつSI組立単位

量	名称	記号	他のSI単位による表現
周波数	ヘルツ	Hz	s^{-1}
力	ニュートン	N	m·kg/s^{2}
圧力，応力	パスカル	Pa	N/m^{2}
エネルギー,仕事,熱量	ジュール	J	N·m
工率，放射束	ワット	W	J/s
電気量，電荷	クーロン	C	A·s
電位，電圧，起電力	ボルト	V	W/A
静電容量	ファラド	F	C/V
電気抵抗	オーム	Ω	V/A
コンダクタンス	ジーメンス	S	A/V
磁束	ウェーバ	Wb	V·s
磁束密度	テスラ	T	Wb/m^{2}
インダクタンス	ヘンリー	H	Wb/A
セルシウス温度	セルシウス度	℃	
光束	ルーメン	lm	cd·sr
照度	ルクス	lx	lm/m^{2}
放射能	ベクレル	Bq	s^{-1}
吸収線量	グレイ	Gy	J/kg
線量等量	シーベルト	Sv	J/kg

表 2　SIと併用される単位

名称	記号
分，時，日	min，h，d
度，分，秒	°，′，″
リットル	l，L
トン	t
電子ボルト	eV
原子質量単位	u

1 eV=1.60218×10^{-19}J
1 u=1.66054×10^{-27}kg

表 4　SIと共に暫定的に維持される単位

名称	記号
オングストローム	Å
バーン	b
バール	bar
ガル	Gal
キュリー	Ci
レントゲン	R
ラド	rad
レム	rem

1 Å=0.1nm=10^{-10}m
1 b=100fm^{2}=10^{-28}m^{2}
1 bar=0.1MPa=10^{5}Pa
1 Gal=1cm/s^{2}=10^{-2}m/s^{2}
1 Ci=3.7×10^{10}Bq
1 R=2.58×10^{-4}C/kg
1 rad=1cGy=10^{-2}Gy
1 rem=1cSv=10^{-2}Sv

表 5　SI接頭語

倍数	接頭語	記号
10^{18}	エクサ	E
10^{15}	ペタ	P
10^{12}	テラ	T
10^{9}	ギガ	G
10^{6}	メガ	M
10^{3}	キロ	k
10^{2}	ヘクト	h
10^{1}	デカ	da
10^{-1}	デシ	d
10^{-2}	センチ	c
10^{-3}	ミリ	m
10^{-6}	マイクロ	μ
10^{-9}	ナノ	n
10^{-12}	ピコ	p
10^{-15}	フェムト	f
10^{-18}	アト	a

（注）

1．表1－5は「国際単位系」第5版，国際度量衡局1985年刊行による。ただし，1 eVおよび1 uの値はCODATAの1986年推奨値によった。
2．表4には海里，ノット，アール，ヘクタールも含まれているが日常の単位なのでここでは省略した。
3．bar は，JISでは流体の圧力を表わす場合に限り表2のカテゴリーに分類されている。
4．EC閣僚理事会指令では bar，barnおよび「血圧の単位」mmHgを表2のカテゴリーに入れている。

換　算　表

力	N(=10^{5}dyn)	kgf	lbf
	1	0.101972	0.224809
	9.80665	1	2.20462
	4.44822	0.453592	1

粘　度　1 Pa·s(N·s/m^{2})=10 P(ポアズ)(g/(cm·s))

動粘度　1m^{2}/s=10^{4}St(ストークス)(cm^{2}/s)

圧力	MPa(=10bar)	kgf/cm^{2}	atm	mmHg(Torr)	lbf/in^{2}(psi)
	1	10.1972	9.86923	7.50062×10^{3}	145.038
	0.0980665	1	0.967841	735.559	14.2233
	0.101325	1.03323	1	760	14.6959
	1.33322×10^{-4}	1.35951×10^{-3}	1.31579×10^{-3}	1	1.93368×10^{-2}
	6.89476×10^{-3}	7.03070×10^{-2}	6.80460×10^{-2}	51.7149	1

エネルギー・仕事・熱量	J(=10^{7} erg)	kgf·m	kW·h	cal(計量法)	Btu	ft·lbf	eV
	1	0.101972	2.77778×10^{-7}	0.238889	9.47813×10^{-4}	0.737562	6.24150×10^{18}
	9.80665	1	2.72407×10^{-6}	2.34270	9.29487×10^{-3}	7.23301	6.12082×10^{19}
	3.6×10^{6}	3.67098×10^{5}	1	8.59999×10^{5}	3412.13	2.65522×10^{6}	2.24694×10^{25}
	4.18605	0.426858	1.16279×10^{-6}	1	3.96759×10^{-3}	3.08747	2.61272×10^{19}
	1055.06	107.586	2.93072×10^{-4}	252.042	1	778.172	6.58515×10^{21}
	1.35582	0.138255	3.76616×10^{-7}	0.323890	1.28506×10^{-3}	1	8.46233×10^{18}
	1.60218×10^{-19}	1.63377×10^{-20}	4.45050×10^{-26}	3.82743×10^{-20}	1.51857×10^{-22}	1.18171×10^{-19}	1

1 cal= 4.18605J（計量法）
= 4.184J（熱化学）
= 4.1855J（15℃）
= 4.1868J（国際蒸気表）

仕事率　1 PS(仏馬力)
= 75 kgf·m/s
= 735.499W

放射能	Bq	Ci
	1	2.70270×10^{-11}
	3.7×10^{10}	1

吸収線量	Gy	rad
	1	100
	0.01	1

照射線量	C/kg	R
	1	3876
	2.58×10^{-4}	1

線量当量	Sv	rem
	1	100
	0.01	1

（86年12月26日現在）

Critical and Subcritical Masses of Curium-245, -246 and -247 Calculated with a Combination of MCNP4A Code and JENDL-3.2 Library