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Technical Note

Calculation of the Minimum Critical Mass of Fissile Nuclides

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Abstract—*The OB-1 method for the calculation of the minimum critical mass of fissile actinides in metal/water systems was described in a previous paper. A fit to the calculated minimum critical mass data using the extended criticality parameter is the basis of the revised method. The solution density (grams/liter) for the minimum critical mass is also obtained by a fit to calculated values. Input to the calculation consists of the Maxwellian averaged fission and absorption cross sections and the thermal values of nubar. The revised method gives more accurate values than the original method does for both the minimum critical mass and the solution densities. The OB-1 method has been extended to calculate the uncertainties in the minimum critical mass for 12 different fissile nuclides. The uncertainties for the fission and capture cross sections and the estimated nubar uncertainties are used to determine the uncertainties in the minimum critical mass, either in percent or grams. Results have been obtained for* ^{233}*U,* ^{235}*U,* ^{236}*Pu,* ^{239}*Pu,* ^{241}*Pu,* ^{242m}*Am,* ^{243}*Cm,* ^{245}*Cm,* ^{249}*Cf,* ^{251}*Cf,* ^{253}*Cf, and* ^{254}*Es. Eight of these 12 nuclides are included in the ANS-8.15 standard.*

I. INTRODUCTION

The OB-1 method for the calculation of the minimum critical mass (mcm) of fissile actinides in metal/water systems was described in a previous paper.[1] In the previous paper a very good fit to the mcm data was obtained using the extended criticality parameter, xk_1, where

$$xk_1 = k_1 \times \eta/\eta_{235} \ ,$$

$$\eta = \nu\sigma_F/\sigma_A \ ,$$

$$k_1 = \nu\sigma_F - \sigma_A \ ,$$

and

$$\sigma_A = \sigma_F + \sigma_c \ .$$

Each of the above is specified for the nuclides of interest. The terms σ_F and σ_A are the one-group fission and absorption cross sections, respectively, and ν is the average number of neutrons per fission. The σ_F and σ_A are weighted with a Maxwellian at 296 K or some other suitable thermal spectrum, and ν is to be evaluated at 0.0253 eV. The capture cross section σ_c is not needed for the basic calculation but is needed for the uncertainty calculation. The η_{235} is the eta of ^{235}U.

In the original paper, the mcm was determined by first calculating the solution density and then multiplying by the critical volume. The method gave mcm estimates within about ±20%. A normalization factor of 1.09 was applied to the calculated values for all nuclides. The values obtained for the solution densities and volumes were rather inaccurate in some cases.

One important difference between this paper and the original method is that the solution densities are now calculated by a least-squares fit to calculated values. This results in considerably improved accuracy relative to the original method. In order for the mcm and the solution densities to be consistent, the mcm is also calculated by a least-squares fit to calculated values. The values used for the least-squares fits were obtained from XSDRN calculations. These values had been obtained as part of the validation procedure for the previous paper and were not repeated for this work. Once the least-squares fits have been done, it is not necessary to repeat this step in order to use this revised procedure. The user does not have to repeat the transport calculations or the least-squares fits in order to use the method; the only input

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required is the thermal values for the fission and absorption cross sections and nubar. These values can be obtained by either visual inspection of the evaluated nuclear data file or by using a retrieval program.

The user does not need to generate a multigroup or continuous energy cross-section file, which can be a significant effort in itself. In fact, a complete evaluated nuclear data file is not even required. In this sense, the revised method is much easier to use than the standard transport methods.

There are several different evaluated nuclear data files available for use in transport calculations, e.g., ENDF/B (United States), JENDL (Japan), JEF (Europe), CENDL (China), and BROND (Russia). One application of the current method is to quickly and easily determine the mcm and solution densities predicted by these various files for a number of different nuclides. The criticality data or nuclear data specialist can use this method to predict the impact of a new or proposed nuclear data evaluation without even going to the trouble of generating a multigroup or continuous energy data file.

The OB-1 program is a small C program, currently about 280 statements. The number of input variables is basically eight times the number of nuclides plus a few others. Currently there are 18 nuclides and about 150 input variables. A large part of the effort in developing the OB-1 program was to evaluate the input, which is based on ENDF/B-VI and JENDL-3.2 evaluated cross-section files. The required input is the average fission and capture cross sections at thermal energies and the value of nubar (the average number of neutrons/fission). One goal of the project was to determine which of the evaluated files had the "best" evaluated data for a particular nuclide, but there is no requirement to use data from an evaluated file. The user is at liberty to use any valid data that he or she may be able to obtain. This may be useful to assess the accuracy of the data in the evaluated nuclear data files (e.g., ENDF/B-VI or JENDL-3.2). The input used in this paper for ^{232}Pa is taken from data in the literature and does not correspond to an evaluated data file.

The reader may wonder why it is worthwhile to bother with an approximate method such as the OB-1 method when it is always possible to do a transport calculation and obtain a result that is generally thought to be more accurate. We believe that there are three very good reasons. First, the OB-1 method is much faster and easier to run. Results for 18 or more cases can be obtained in less time than it would take to prepare the input for a single transport case. Another important reason is that the mcm and the solution density are directly related to the input values of the basic thermal cross sections. This makes it very easy to assess the impact of cross-section changes on the calculated parameters. The third reason is that the uncertainties in the calculated parameters, resulting from uncertainties in the input cross sections, are also obtained.

When the basic accuracy of the input cross sections is considered, the accuracy of the more rigorous method may actually be no better because the uncertainty in the result can be larger than the difference between the OB-1 method and the "more accurate" result.

The most important goal of the study was to obtain uncertainties in the calculated mcm resulting from uncertainties in the basic cross sections. These uncertainties are perhaps the most useful results obtained.

Concerning the selection of the nuclides included in this work, we included several nuclides that are of "academic interest" as well as nuclides listed in U.S. Department of Energy (DOE) Order 420.1. This DOE order is quoted in LA-UR-04-6514, a Los Alamos National Laboratory report[2]; the specific requirements are given for 24 nuclides of criticality concern: ^{231}Pa, ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm, ^{247}Cm, ^{249}Cf, ^{250}Cf, ^{251}Cf, ^{252}Cf, and ^{254}Es.

Seven of the above nuclides exist in isolated quantities less than the potential critical mass [see ANSI/ANS-8.15-1981, reaffirmed 1987 (Ref. 3)]; 13 of the 24 nuclides are fissile, and 11 are not. Both ^{232}U and ^{252}Cf are exceptions to the general rule concerning even-even nuclides because the values of η at thermal are greater than 1; thus, ^{252}Cf is in fact a fissile nuclide.[2] Ten of the 13 fissile nuclides are included in this paper; ^{232}U, ^{247}Cm, and ^{252}Cf are not included.

Nuclides not included in DOE Order 420.1 but included in this study are ^{232}Pa, ^{236}Pu, ^{237}Pu, ^{238}Np, ^{244}Am, ^{241}Cm, ^{250}Bk, and ^{253}Cf. They will generally be of academic interest only because these nuclides currently exist in less than the potential critical mass and/or because each of these nuclides would be too "hot" either thermally or radiologically.

The evaluations for both ^{238}Np and ^{253}Cf in the ENDF/B-VI file are incomplete, so a transport calculation would give an incorrect result. There is no ^{253}Cf evaluation in the JENDL-3.2 file.

The fission, absorption, and ν input values used in this work are given in Table I. Considerable time and effort were devoted to the determination of these values. Cross sections for ^{237}Pu, ^{244}Am, ^{241}Cm, ^{250}Bk, ^{251}Cf, and ^{254}Es are from JENDL-3.2; cross sections for the other 12 nuclides are taken from ENDF/B-VI. The values in Table I are arranged in order of decreasing values of xk_1. The calculated parameters α, η, and k_1 are given in Table II. The value of σ_A is also included again for convenience. The parameters α, η, and k_1 can be defined as follows:

$$\alpha = (\sigma_A - \sigma_F)/\sigma_F \ ,$$

$$\eta = \nu/(1 + \alpha) \ ,$$

and

$$k_1 = (\eta - 1) \times \sigma_A \ .$$

TABLE I

OB-1 Input Cross Sections: Fission, Absorption, and Nubar

Nuclide	xk_1 (b)	σ_F (b)	σ_A (b)	Nubar
^{242m}Am	17 360	6470	7790	3.2640
^{251}Cf[a]	13 192	4196	6642	4.1059
^{254}Es[a]	10 340	1742	1767	4.0832
^{253}Cf	7 105	1152.1	1168.1	4.1500
^{245}Cm	6 849	1859	2144	3.6059
^{249}Cf	5 908	1495	1864	3.8869
^{244}Am[a]	4 728	2096	2640	3.1418
^{237}Pu[a]	4 125	2176	2619	2.8630
^{238}Np	3 643	1769	1946	2.7900
^{250}Bk[a]	2 220	803.2	1099.1	3.5720
^{243}Cm	2 187	613.01	664.40	3.4300
^{241}Cm[a]	1 846	620.36	744.43	3.3915
^{241}Pu	1 574	937.47	1263	2.9453
^{239}Pu	1 027	697.57	973.15	2.8800
^{233}U	731.4	470.32	510.66	2.4947
^{235}U	637.5	503.51	589.43	2.4367
^{232}Pa	350.5	865.8	1446	2.2858
^{236}Pu	269.9	146.2	173.9	2.8140

[a]From the JENDL-3.2 cross sections.

TABLE II

OB-1 Calculated Parameters for α, η, and k_1

Nuclide	α	η	σ_A (b)	k_1 (b)
^{242m}Am	0.2040	2.7110	7790	13 329
^{251}Cf	0.5829	2.5939	6642	10 586
^{254}Es	0.0144	4.0252	1767	5 347
^{253}Cf	0.0139	4.0932	1168.1	3 613
^{245}Cm	0.1532	3.1270	2144	4 559
^{249}Cf	0.2471	3.1168	1864	3 946
^{244}Am	0.2595	2.4944	2640	3 945
^{237}Pu	0.2037	2.3786	2619	3 610
^{238}Np	0.1001	2.5362	1946	2 989
^{250}Bk	0.3684	2.6103	1099.1	1 770
^{243}Cm	0.0838	3.1647	664.40	1 438
^{241}Cm	0.2000	2.8263	744.43	1 360
^{241}Pu	0.3472	2.1862	1263	1 498
^{239}Pu	0.3951	2.0644	973.15	1 036
^{233}U	0.0858	2.2976	510.66	662.65
^{235}U	0.1706	2.0815	589.43	637.47
^{232}Pa	0.6701	1.3686	1446	533.05
^{236}Pu	0.1895	2.3658	173.9	237.51

The parameter xk_1 can also be written in terms of η:

$$xk_1 = [\eta(\eta - 1)/\eta_{235}] \times \sigma_A \ .$$

The original method is revised and has been extended to calculate the uncertainties in the mcm. The fit using the parameter xk_1 uses the calculated masses from calculations using the XSDRN one-dimensional discrete ordinates code.[4] The values of the mcm (grams), and solution density ρ_F (grams/liter), are obtained by fitting the XSDRN calculated values. The revised method is discussed in Sec. II. We will also describe the extension to obtain the uncertainty in the mcm.

II. THE REVISED OB-1 METHOD

The calculated values of the mcm and the solution density ρ_F from the XSDRN (S32, P3) calculations for seven different nuclides are used to obtain fits for mcm and ρ_F. Equations (1) and (2) are used for the mcm and the solution density, respectively:

$$\text{mcm} = \exp[A + B \times \ln(xk_1)] \tag{1}$$

and

$$\rho_F = \exp[C + D \times \ln(xk_1)] \ , \tag{2}$$

where

$$xk_1 = k_1 \times \eta/\eta_{235} \ ,$$

$$\eta = \nu\sigma_F/\sigma_A \ ,$$

$$k_1 = \nu\sigma_F - \sigma_A \ ,$$

$$A = 13.99320279 \ ,$$

$$B = -1.13085615 \ ,$$

$$C = 9.068890124 \ ,$$

and

$$D = -0.79587973 \ . \tag{3}$$

The OB-1 results from Eq. (1) are compared with the XSDRN values in Table III. Very good agreement is obtained. The main advantage of the new method, compared with the original OB-1 method, is that the new method is more accurate. The normalization factor (1.09 in the original method) is not needed in the revised method. The accuracy of the fit is now within ±7.0%, and the standard deviation is 3.6% (compared to the XSDRN values); see Table III.

The calculated values of the mass, density, and volume for 18 fissile actinides are given in Table IV. Table IV also gives the values of the extended criticality parameter xk_1.

A plot of the calculated masses on a log-log scale is shown in Fig. 1. The same results are shown in Fig. 2 for xk_1 = 500 to 12 000 on a linear-linear scale. Eighteen nuclides have been included in this paper. In the previous paper[1] we had calculated masses for 20 nuclides.

TABLE III

Minimum Critical Mass Fit to the XSDRN Values

Nuclide	XSDRN (g)	OB-1 (g)	Difference (%)
^{235}U	800	804.88	0.61
^{239}Pu	504	469.19	−6.91
^{241}Pu	273.2	289.73	6.05
^{237}Pu[a]	133.7	133.05	−0.49
^{245}Cm	54.37	54.91	1.00
^{254}Es	32.40	34.46	6.36
^{242m}Am	20.47	19.18	−6.30
Standard deviation			3.55

[a]The ^{237}Pu cross sections are ENDF/B-VI in Table III; JENDL-3.2 is used elsewhere in this paper. We decided that JENDL-3.2 values were better than ENDF/B-VI values.

TABLE IV

Revised OB-1 Calculations for 18 Nuclides

Nuclide	xk_1 (b)	Mass (g)	Density (g/ℓ)	Volume (ℓ)
^{242m}Am	17 360	19.18	[a]	—
^{251}Cf	13 192	26.16	[a]	—
^{254}Es	10 340	34.46	5.540	6.220
^{253}Cf	7 105	52.68	7.468	7.054
^{245}Cm	6 849	54.91	7.689	7.141
^{249}Cf	5 908	64.89	8.649	7.503
^{244}Am	4 728	83.50	10.33	8.083
^{237}Pu	4 125	97.41	11.51	8.463
^{238}Np	3 643	112.1	12.71	8.820
^{250}Bk	2 220	196.3	18.85	10.41
^{243}Cm	2 187	199.7	19.08	10.47
^{241}Cm	1 846	241.9	21.83	11.08
^{241}Pu	1 574	289.7	24.79	11.69
^{239}Pu	1 027	469.2	34.80	13.48
^{233}U[b]	872.6	564.3	55.68	10.13
^{235}U	637.5	804.9	50.88	15.82
^{232}Pa	350.5	1583	81.91	19.33
^{236}Pu[b]	429.2	1259	170.3	7.39

[a]Densities from the OB-1 fit are inaccurate for ^{242m}Am and ^{251}Cf.

[b]The ^{233}U and ^{236}Pu nuclides used the LERC correction to the value of xk_1.

We find that the method does not work well for ^{233}U or ^{236}Pu. We are reasonably sure that this is because of significant contributions to both the fission and capture rates from resonances in the 1- to 10-eV range (failure of the one-group approximation). A low-energy-resonance correction (LERC) can be used for ^{233}U and ^{236}Pu. This is done by applying a correction factor to the value of xk_1.

The method used for the LERC is analogous to the fast-fission factor in the four-factor formula. A correction factor is applied to the calculated value of the xk_1 parameter. There is not a rigorous method to evaluate the factor; instead, it is determined empirically. The values for the LERC factors are 1.193 for ^{233}U and 1.59 for ^{236}Pu. The LERC factor is applied to the calculated values of xk_1, and the calculation then proceeds as for the other nuclides.

The values in Table IV are calculated using LERC factors for ^{233}U and ^{236}Pu. Both the mcm and ρ_F for these two cases are greatly improved using the LERC correction factor. Since both the mcm and ρ_F are improved, it is judged to be worthwhile to use the LERC. The reason for the failure of the basic OB-1 method will be discussed next.

The ^{233}U nuclide has two large resonances, just below and just above 2 eV. The E_0 values are 1.763 and 2.290 eV. The peak cross sections do not occur at exactly the E_0 values. The thermal (Maxwellian average) fission and absorption cross sections are 470 and 511 b, respectively. The fission cross sections are 1.85 and 1.39 times larger at 1.763 and 2.290 eV, respectively. The absorption cross sections are 2.08 and 2.16 times larger, respectively, at the same energies. It appears that the one-group approximation does not work well for ^{233}U because there are very significant contributions to the fission and absorption rates from the 1.763- and 2.290-eV resonances.

A similar problem is also observed for ^{236}Pu, for which the thermal (0.0253-eV) fission cross section is 165 b. The ^{236}Pu nuclide has large resonances at 3.16 and 6.30 eV. The peak fission cross sections are 6000 and 1750 b, respectively. The ^{236}Pu fission cross section is shown in Fig. 3. The ENDF/B-VI evaluation of the fission cross section is based on the measured data of Gromova et al.[5] The fission cross section at the peak of the 3.16-eV resonance is 36 times larger than the value at thermal. The fission rate per unit lethargy for the ^{236}Pu/water system was calculated by XSDRN; the mcm was 1.1 kg, and the solution density was 174 g/ℓ. The fission rate at the peak of the 3.16-eV resonance is 50% higher than the thermal value at 0.04 to 0.05 eV. The one-group approximation basically does not work at all for ^{236}Pu. The OB-1 result for the ^{236}Pu/water system is high by about a factor of two.

The original OB-1 method gave better results for both ^{233}U and ^{236}Pu. As far as we can tell, this was fortuitous. A recent XSDRN calculation for ^{233}U gave an mcm of 562 g at 56 g/ℓ. The original OB-1 method gave a value of 581 g at 57 g/ℓ. The revised OB-1 result for ^{233}U, when the LERC correction is not used, is 686 g at 0.046 g/ℓ. For nuclides other than ^{233}U and ^{236}Pu, the revised OB-1 method produces more accurate values than the original method did[1] for both the mcm and the solution density.

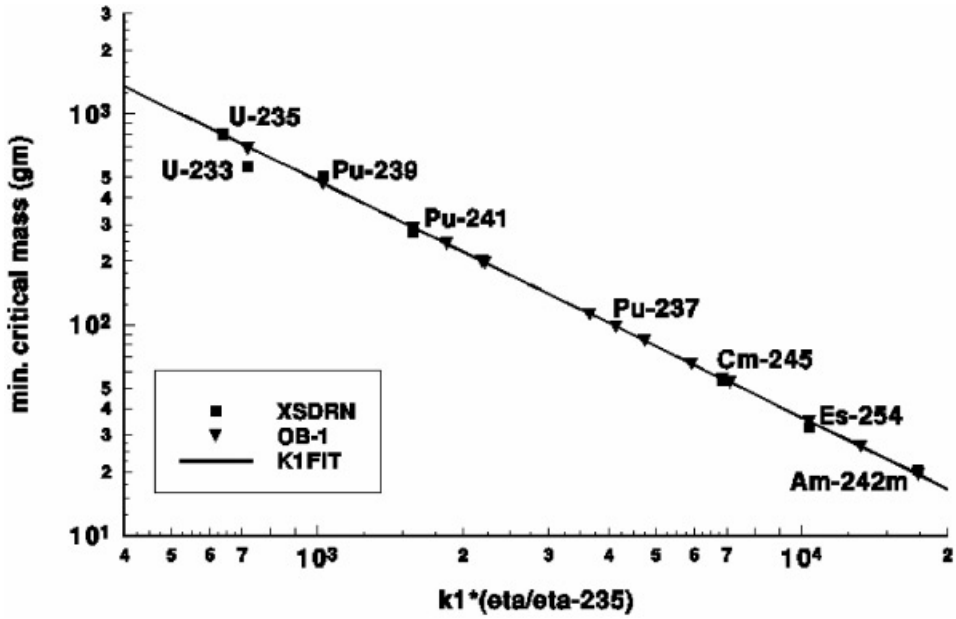

Fig. 1. Minimum critical masses, log-log plot. Values are listed in Table IV.

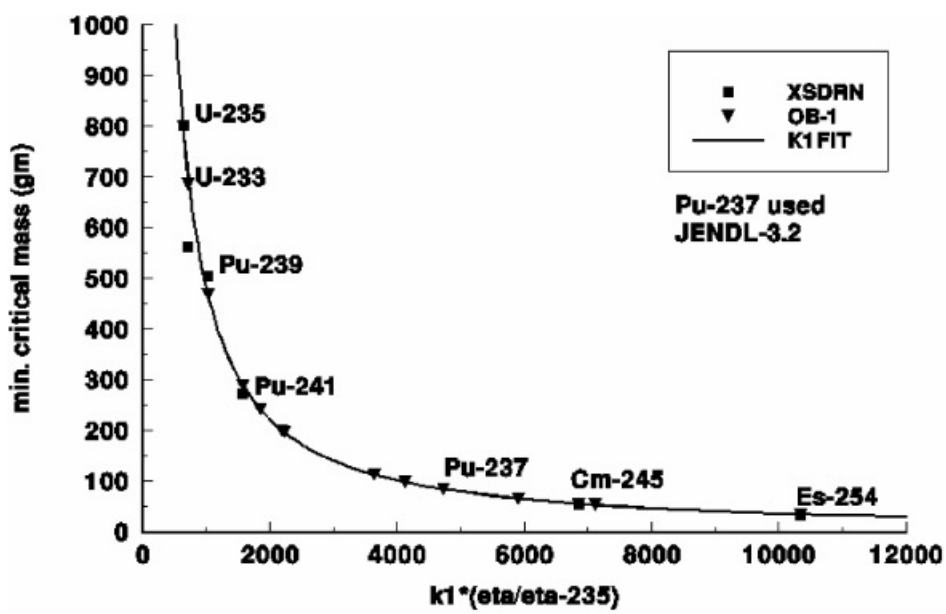

Fig. 2. Minimum critical masses, linear-linear plot. Values are listed in Table IV.

III. UNCERTAINTIES IN MINIMUM CRITICAL MASS

The OB-1 method has been extended to calculate the uncertainties in the mcm for 12 different nuclides. The only change is to perform calculations for the perturbed cross sections corresponding to a one-standard-deviation increase in the fission and capture cross sections and nubar. The calculation of the uncertainties is done for each parameter separately. The total uncertainty is obtained by "adding by squares" (the square root of the sum of the squares of each contribution). This neglects any possible cross-section correlations.

As an example, consider ^{241}Pu fission with an uncertainty of 1%. This uncertainty corresponds to a factor of 1.01 in σ_F. The perturbed value is $\sigma_F = \sigma_F \times$ factor.

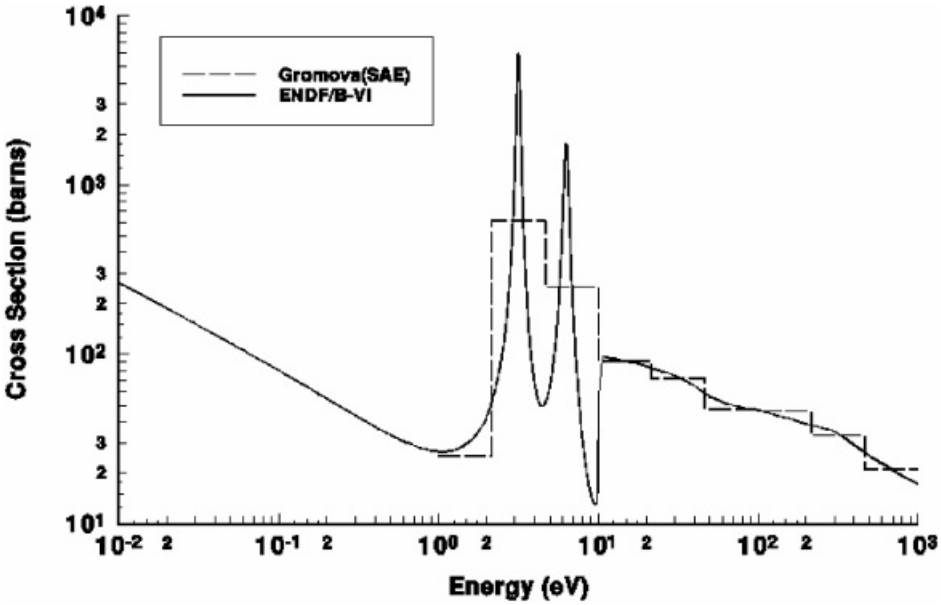

Fig. 3. The ^{236}Pu fission cross section from ENDF/B-VI, Release 7 evaluation.

Next, calculate the perturbed value of the parameter xk_1. Get the perturbed value of mcm from Eq. (1) using the perturbed value of xk_1: $\delta(\text{mcm}) = \text{mcm} - \text{mcm (perturbed)}$. Then, convert the value of δ to percent. Also do this for nubar and capture; in the extended method we need to use the capture cross section (not the absorption cross section) for calculating the uncertainties. Then, to get the total change, calculate the square root of the sum of the squares of the three contributions.

The cross-section uncertainties for capture and fission from Holden[6] and the estimated nubar uncertainties are used to determine the uncertainties in the mcm. The nubar uncertainties assumed in this work for ^{243}Cm, ^{245}Cm, ^{251}Cf, ^{253}Cf , and ^{254}Es are 4.0, 2.7, 11.1, 11.1, and 11.1%, respectively, obtained from Holden and Zucker.[7] The nubar uncertainty for ^{242m}Am is 0.75%, from Jaffey and Lerner.[8] The values for ^{233}U, ^{235}U, ^{236}Pu, ^{239}Pu, and ^{241}Pu are 0.3, 0.2, 4.0, 0.4, and 0.5%, respectively, assigned in this paper.[a] For ^{249}Cf a value of 1.75% was chosen.[9] It is very important to have the best possible value for the nubar uncertainty because it is usually the largest component of the total uncertainty. The fission and capture cross-section uncertainties from Holden[6] are larger than the corresponding values from Mughabghab[9] and appear to be more realistic. The uncertainties for nubar, fission, and capture are displayed in Table V.

[a]Nubar uncertainties for ^{233}U, ^{235}U, ^{236}Pu, ^{239}Pu, and ^{241}Pu are specified in this paper. The values for ^{233}U, ^{235}U, and ^{241}Pu are essentially the same as those found in Ref. 8; the value for ^{239}Pu is higher than Ref. 8; and a value for ^{236}Pu is not given in Ref. 8.

TABLE V

Cross Section Uncertainties*

Nuclide	Nubar	Fission[6]	Capture[6]
^{233}U	0.30 (Ref. 9)	1.90	4.25
^{235}U	0.20 (Ref. 9)	0.34	5.26
^{236}Pu	4.00 (Ref. 9)	19	19
^{239}Pu	0.40 (Ref. 9)	0.40	3.70
^{241}Pu	0.50 (Ref. 9)	1.00	2.70
^{242m}Am	0.75 (Ref. 8)	4.30	23.5
^{243}Cm	4.00 (Ref. 7)	3.24	7.70
^{245}Cm	2.70 (Ref. 7)	4.76	5.70
^{249}Cf	1.75 (Ref. 10)	5.90	6.00
^{251}Cf	11.1 (Ref. 7)	11.1	6.90
^{253}Cf	11.1 (Ref. 7)	18.5	10.0
^{254}Es	11.1 (Ref. 7)	11.1	10.7

*Values are in percent.

The results obtained (using the uncertainties in Table V) are given in Table VI (values are in percent). "SQRSS" is the square root of the sum of the squares of the nubar, fission, and capture components. The mcm and uncertainty values (in grams) are also shown. It is interesting to observe that for several of the nuclides the OB-1/XSDRN difference is less than the uncertainty in the XSDRN mcm resulting from the basic cross-section uncertainties given in Table V. This is true for ^{235}U, ^{242m}Am, ^{245}Cm, and ^{254}Es. The uncertainties used in this paper for the fission and capture cross sections could be too low for some nuclides. If we find that the various

TABLE VI

Change in Minimum Critical Mass due to Increase Corresponding to the Changes in Cross-Section Uncertainties*

Nuclide	Nubar	Fission	Capture	SQRSS[a]	Mass (g)
^{233}U	−0.66	−2.39	0.67	2.57	562 ± 15
^{235}U	−0.93	−0.49	1.68	1.98	800 ± 16
^{236}Pu	−11.3	−21.9	6.08	25.4	1100 ± 280
^{239}Pu	−1.32	−0.70	2.32	2.76	504 ± 14
^{241}Pu	−1.59	−1.64	1.46	2.71	273 ± 7.4
^{242m}Am	−2.16	−5.84	7.33	9.62	20.5 ± 2
^{243}Cm	−10.29	−3.93	0.99	11.0	200 ± 22
^{245}Cm	−7.15	−6.07	1.26	9.46	54.4 ± 5.1
^{249}Cf	−4.72	−7.99	1.99	9.49	64.9 ± 6.2
^{251}Cf	−26.4	−17.1	4.77	31.8	26.2 ± 8.3
^{253}Cf	−24.0	−17.7	0.21	29.8	52.7 ± 16
^{254}Es	−24.0	−11.4	0.23	26.6	32.4 ± 8.6

*Values are in percent.

[a]Cross-section correlations are not considered. SQRSS = square root of the sum of the squares of the nubar, fission, and capture components.

evaluations (e.g., ENDF/B-VI and JENDL-3.3) differ by more than the currently assigned uncertainty, there would appear to be something wrong. For the last column in Table VI, the masses for ^{243}Cm, ^{249}Cf, ^{251}Cf, and ^{253}Cf are from the OB-1 calculations; the masses for the other eight nuclides were calculated by XSDRN. The uncertainties for all nuclides are obtained using the OB-1 calculation as described in this paper. Thus, the last column of Table VI has the best values for the mcm and also credible values for the uncertainties in these values as determined in this paper.

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