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Modeling and Fission Cross Sections for Americium

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Modeling and Fission Cross Sections for Americium

Final Report for the LANL Contract

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May 23, 2005

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Summary

This is the final report of the work performed under the LANL contract on the modeling and fission cross section for americium isotopes (May 2004 - June 2005). The purpose of the contract was to provide fission cross sections for americium isotopes with the nuclear reaction model code EMPIRE 2.19.

The following work was performed:

- Fission calculations capability suitable for americium was implemented to the EMPIRE-2.19 code.
- Calculations of neutron-induced fission cross sections for ^{239}Am to ^{244g}Am were performed with EMPIRE-2.19 for energies up to 20 MeV. For the neutron-induced reaction of ^{240}Am, fission cross sections were predicted and uncertainties were assessed.
- Set of fission barrier heights for each americium isotopes was chosen so that the new calculations fit the experimental data and follow the systematics found in the literature.

References for the present contract:

BNL account No.	86737
BNL project title:	Modeling and Fission Cross Sections for Americium
BNL principal investigator	Pavel Obložinský, NNDC, oblozinsky@bnl.gov
Period of time	May 2004 - June 2005
LANL purchase order No.	03533-001-04 3C
LANL technical representative	Mark Chadwick, T-16, chadwick@lanl.gov and Eric Pitcher, T-16, pitcher@lanl.gov

I. STATEMENT OF WORK

The statement of work (SOW) specifies technical tasks to be performed under the present project. This statement, formulated by Mark Chadwick, LANL, is reproduced below in full.

Tasks

1. Extend BNL EMPIRE fission modeling code so as to be able to predict fission cross sections for americium isotopes.
   - Implement barrier penetration model for americium isotopes.
   - Literature search, using BNL's online nuclear databases, of experimental data on americium isotope fission cross sections for 240 - 243Am; literature search for fission probability data, based on indirect transfer reactions.
   - Assessment of double-humped fission barrier parameters and fission level density.
2. Perform nuclear model predictions of n + ^{240}Am fission cross sections, for energies from 1 keV to 20 MeV, based on BNL EMPIRE modeling code.
   - Assess uncertainties in predicted cross section (no fundamental data measurements exist for ^{240}Am), due to uncertainties in input parameters.
   - Compare EMPIRE code predictions with those from LANL analysis, for an independent validation of LANL work.
3. Issue Report on this work to LANL, by June 1st, 2005

II. INTRODUCTION

Recently, americium isotopes have been subject for a growing interest in defense related research and energy production field. Both weapons grade (with 0.5 % of plutonium-241 [1]) and power reactor grade plutonium (with 13.9 % of plutonium-241 in the MOX fuel [2]) contain some plutonium-241, which decays into americium-241 by emitting a beta particle. Since americium-241 has a longer half-life (432 years) than plutonium-241 (14.4 years), it builds up as plutonium-241 decays. The gamma radiation from americium-241 decay, which is far stronger than that from plutonium-239, also builds up with the age of the plutonium sample. Americium might then slowly change the chemistry of the plutonium metal and modify the global reaction of an actinide assembly (americium isotopes contribute about 10 % to the radiotoxic inventory in spent nuclear fuel [3]). Furthermore, stockpile stewardship, DOE's program for certifying the long-term safety and performance of the enduring stockpile without underground nuclear testing, has heightened the importance of assessing and predicting the long-term behavior of actinides. Finally, actinides such as uranium, neptunium, plutonium, and americium are the major contributors to the long-term radioactivity of nuclear waste currently targeted for the proposed Yucca Mountain repository in Nevada.

These applications call for new evaluations for the neutron-induced cross sections, especially fission, which in the case of americium were not given much attention. Most of the direct measurements on fission cross sections on americium isotopes were done in the late seventies and early eighties, except for ^{242m}Am and ^{243}Am, for which the most recent measurements were carried out in 1997 and 2004, respectively [4, 5]. But for these two latter isotopes, the measurements are not in agreement with other experimental data and differ by up to 50 % in the case of ^{242m}Am and up to 20 % for ^{243}Am. Recently, new indirect measurements of fission probabilities were performed using the "surrogate" technique exploiting (^{3}He,df) and (^{3}He,tf) reactions on stable targets [6]. Comparison with directly measured values shows that the surrogate results agree within 10 % of systematics uncertainties.

New evaluations for the americium isotopes are being performed at the Theoreti-

cal Division at the Los Alamos National Laboratory (T-16). This effort relies on the pre-equilibrium-statistical model code GNASH [7, 8]. The present work provides for cross-check of these calculations. The nuclear reaction model code EMPIRE 2.19 [9] has been used at the National Nuclear Data Center (NNDC) at the Brookhaven National Laboratory to calculate neutron-induced fission cross sections for the americium 239 through 244.

In this report, we collect existing experimental data and evaluations, and compare them against new calculations with the EMPIRE 2.19 code. In the case of ^{239,240,242g,244g}Am, for which there are almost no direct experimental data or evaluations, EMPIRE 2.19 calculations are compared with existing simulations or indirect measurements. The present report is organized as follows. First, a literature is presented for the neutron-induced fission data existing in the literature and in the EXFOR library. Then the EMPIRE-2.19 fission model is succinctly presented. The next parts of this report present the calculations of fission cross section up to 20 MeV for the americium isotopes with the EMPIRE-2.19, compared to the literature experimental data as well as simulations and data from evaluated libraries. In the last part of this report, the fission barrier heights from the EMPIRE-2.19 calculations are compared to the systematics of the fission barrier heights found in the literature.

III. LITERATURE SEARCH

A. Fission barrier heights for ^{239}Am to ^{244g}Am

In the following we list relevant references with a short explanation of the method to obtain the fission barrier parameters.

- Ref. [10]: V. M. Maslov, IAEA Report, INDC(BLR)-013 (1998)
  Adopted fission and total level densities modeling along with fission barrier parameters are presented to describe neutron-induced fission cross section data of, among others, americium isotopes. The data for incident neutron energies from 10 keV up to the fission threshold were analyzed. Saddle asymmetries relevant to the Shell Correction Method from Strutinsky-type theory calculations were used.

- Ref. [11]: H. Weigmann and J. P. Theobald, Nucl. Phys. **A 187**, 305 (1972)

A combined analysis of data from narrow intermediate structure in near-barrier fission cross sections and from shape-isomer half-lives is tried in a very phenomenological model. Among the results, one observes a marked even-odd effect in the $\hbar\omega$ values, whilst the barrier heights do not show a significant effect of this type.

- Ref. [12]: V. M. Kupriyanov, G. N. Smirenkin, B. I. Fursov, Yad. Fiz. **38**, 281 (1984).
  The calculations were carried out on the basis of a systematics developed in the framework of the two-humped barrier model of fission and a statistical description of the average decay widths of the compound nucleus.

- Ref. [13]: B. B. Back, O. Hansen, H. C. Britt and J. D. Garrett, Phys. Rev. **C 9**, 1924 (1974).
  Fission probability distributions were measured by (t,pf), (t, alpha f), (t,t'f), (p,p'f), (3He,df), and (3He, alpha f) reactions with bombarding energies of 15, 16, 20, 22.5, 24, and 24 MeV, respectively. The results are analyzed with a statistical model which involves resonant penetration of the double-humped fission barrier. Estimates of the heights and curvature of the two peaks in the fission barrier are obtained for, among other, americium isotopes and these values are compared with various theoretical calculations.

- Ref. [14]: T. Ohsawa, Y. Shigemitsu, M. Ohta and K. Kudo, Journ. of Nucl. Sci. and Techn. **21**, 887 (1984)
  Fission cross sections of 24 actinides were analyzed in terms of the double-humped fission barrier model to deduce the barrier heights.

- Ref. [15]: J. E. Lynn, AERA-R-7468, (1979)
  Barrier heights were extracted from measurements on (d,pf), (t,pf) reactions and neutron-induced fission. Level densities were considered energy-independent with empirical description of the parameters for the estimation of the transition state spectra below the pairing gap for odd mass nuclei.

B. Fission cross section data for ^{239}Am to ^{244g}Am

The references for the experimental data on fission cross section for the americium isotopes are presented in the next sections, together with the EMPIRE calculations. In Table I, these references are classified as function of the type of experiment.

TABLE I: Classification of the references as function of the type of experiment for different americium isotopes.

Isotopes	Surrogate + (^3He,xf)	Linac White Neutron Source	Mono-energetic neutron source	Van De Graaff (d,d), (p,t)	Cyclotron and Time-Of-Flight
^{239}Am	[16]				
^{240}Am	[6, 16]				
^{241}Am	[6]	[17–23]	[24]	[25]	
^{242g}Am	[6]		[24]		
^{242m}Am	[6, 16]	[26–30]	[24, 26]		
^{243}Am		[17, 18]	[24]	[4, 31–34]	[5]
^{244g}Am	[6, 16]				

The references containing information for the fission barrier heights are presented in Tables II to IV for each americium isotopes.

IV. NUCLEAR REACTION MODEL CODE EMPIRE-2.19

A. Overview

Detailed description of the EMPIRE nuclear reaction model code can be found elsewhere [35, 36]. Its latest version, EMPIRE-2.19, was released in March 2005. EMPIRE-2.19 includes extensions of fission modeling that allows to predict fission cross sections for americium isotopes (see next chapters).

EMPIRE includes various nuclear models, designed for calculations in a broad range of energies and incident particles. The version used in the present calculations (EMPIRE-2.19)

includes coupled channels model (ECIS) [37], quantum-mechanical multistep direct (ORION +TRISTAN) [38] mechanism, two implementations of the phenomenological preequilibrium exciton model (PCROSS and DEGAS), Monte Carlo preequilibrium hybrid model (HMS) emission, and the full-featured Hauser-Feshbach model with width fluctuation correction. For the purpose of this work, EMPIRE 2.19 was extended to include advanced modeling of the fission process. This formalism makes use of the optical model approach to fission and calculates transitions through a double-humped fission barrier with imaginary potential in the second well. A comprehensive library of input parameters, largely based on the RIPL-2 library covers, nuclear masses, optical model parameters, ground-state deformations, discrete levels and decay schemes, level densities, moments of inertia, and gamma-ray strength functions.

B. Fission calculations with EMPIRE

Fission calculations are selected in EMPIRE-2.19 with the option FISSHI = 0. Neutron-induced fission calculation is a new feature of EMPIRE-2.19 with an advanced fission formalism for multi-chance fission. One can select to describe the incident neutron through a single-, double- or triple-humped fission barrier model, starting from sub-barrier excitation energies up to about 200 MeV. For multi-humped barrier, an expression for the fission probability is derived in the frame of the optical model for fission. In the case of double-humped barrier, as selected here, the expression is generalized for the case of multi-modal fission. This fission model can provide good description of the experimental data and reasonably good predictive power for the determination of fission barrier parameters.

a) Fission barriers

The optical model for fission considers the possible transmission mechanisms using a complex potential to describe the uni-dimensional multi-humped fission barrier, see Fig. 1 and next section.

b) Transmission mechanism

In the case of excitation energy below both humps V_1 and V_2, the incoming neutron can

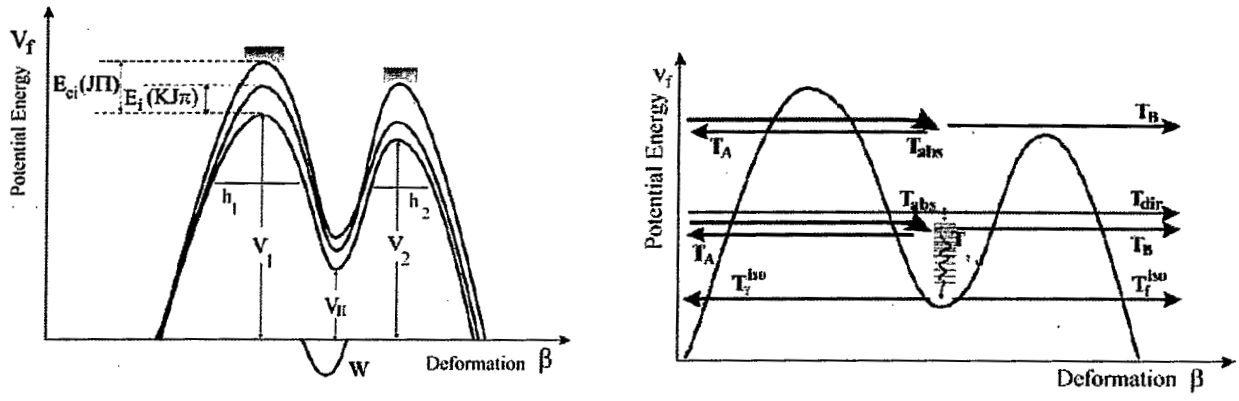

FIG. 1: Left: Double-humped fission barrier used in multi-modal calculations. V_1 and V_2 are the barrier heights, h_1 and h_2 the barrier widths. E_i is defined in Eq. (2). Right: Transmission mechanisms through a double-humped fission barrier (see text for details).

be transmitted directly through the barrier or can be absorbed in the isomeric well. The fraction absorbed in the isomeric well can:

- be re-emitted in the fission channel (T_B in Fig. 1 right)
- return back to a class I state (T_A in Fig. 1 right)
- undergo gamma-transition to the isomeric state (T_γ in Fig. 1 right).

c) Decay probabilities

The most general expressions used in EMPIRE for the fission probability calculation are derived from the procedure of Back [13] for multi-modal fission. It uses a statistical model which involves resonant penetration of the double-humped barrier.

d) Parameters used in for the calculations

- The optical model for fission is selected, and can be applied for the multi-humped barriers at any excitation energy, but requires complete information about the complex fission potential (option FISOPT = 1)
- The heights and widths are retrieved from an internal library where one can store the desired barrier parameters (option FISBAR = 1).
- The transition state spectrum has a discrete part. The maximum number of rotational band-heads states is 30. Each band-head is characterized by its excitation energy, its spin

projection on the symmetry axis and its parity. The widths of the parabolas for a given barrier associated to a certain transition state is defined in the input file. By default, there are four discrete states and their inertial parameters are in agreement with the RIPL-2 recommendations (option FISDIS = 1).

- The level densities at saddle points are calculated using the same EMPIRE-specific dynamical approach as used for normal states (option FISMOD = 0).

The following models were used for the calculations of fission of americium isotopes:

- Coupled Channels (ECIS)

  The coupled channels method was used for calculation of total, elastic and reaction cross sections including inelastic scattering to collective levels. All transmission coefficients for neutron emission were calculated within the Coupled Channels approach using ECIS03 code (option DIRECT = 2).

- Width Fluctuations (HRTW)

  The width fluctuation effects were treated in terms of the HRTW model proposed by H. M. Hofmann *et al.* [39] (option HRTW = 2).

- Exciton Model with cluster emission (PCROSS)

  The PCROSS module was used to improve fast neutron capture and to add the capability of predicting preequilibrium emission of clusters in terms of the Iwamoto-Harada model (option PCROSS = 1.4 for ^{239,240,241,242g,242m}Am).

- Monte Carlo Preequilibrium Model (HMS)

  The Hybrid Monte-Carlo Simulation (HMS) approach was used to predict preequilibrium emission of nucleons from ^{243,244}Am (option HMS = 1).

- Level Density Model

  The EMPIRE-specific level densities, BCS + Fermi gas with deformation-dependent collective effects, adjusted to experimental a values and to discrete levels were used for all isotopes (option LEVDEN = 0).

C. Implementation of Barrier Penetration Model for americium isotopes

The fission barrier parameters are one of the most important parameters considering the shape of the fission cross section and its absolute values. The real part of the barriers, associated with the discrete transition states, are parameterized as a function of the quadrupole deformation using smoothly joined parabolas (see Fig. 1):

$$V_i(\beta) = E_{fi} + (-1)^i \frac{1}{2}\mu\hbar^2\omega_i^2(\beta - \beta_i)^2, \tag{1}$$

where $i = 1$ for a single barrier, runs from 1 to 3 for a two-humped barrier, and from 1 to 5 for a three-humped one. The energies E_{fi} represent maxima of V_i in odd regions (humps) and minima in even regions (wells), β_i are the corresponding abscissae, the harmonic oscillator frequencies ω_i define the curvature of each parabola and μ is the inertial mass parameter, assumed independent of β and approximated by the semi-empirical expression $\mu \approx 0.054A^{5/3}MeV^{-1}$, where A is the mass number.

The discrete transition states are rotational levels built on vibrational or non-collective band-heads, characterized by a given set of quantum numbers (angular momentum J, parity π and angular momentum projection on the nuclear symmetry axis K) with excitation energies

$$E_i(J, K, \pi) = E_{fi} + \epsilon_i(K, \pi) + \frac{\hbar^2}{2I_i}[J(J+1) - K(K+1)], \tag{2}$$

where $\epsilon_i(K, \pi)$ are the excitation energies of the band-heads, and $\hbar^2/2I$ are inertial parameters (the Coriolis term for $K = 1/2$ is neglected). To each transition state associated is a parabolic barrier with the height $E_i(J, K, \pi)$ and the curvature $\hbar\omega_i$. The transition state spectrum consists of a discrete part below a certain energy E_{ci} and a continuum described by the level density functions $\rho_i(E, J, \pi)$.

The negative imaginary potential iW is introduced in the deformation range corresponding to the second well for both double- and triple-humped barriers to simulate damping of the class II vibrational states, hence, the absorption of the incoming flux in this well (see Fig. 1). The strength W depends quadratically on deformation and increases with the excitation energy

$$W(\beta) = -\alpha[E - V(\beta)] \tag{3}$$

The input parameter α, which controls the strength of the imaginary part of the fission potential, should be chosen to fit the width of the resonances in sub-barrier fission cross section and to be consistent with physical values for the transmission coefficients at higher energies.

The dynamic approach to the level densities is specific to the EMPIRE code. It takes into account collective enhancements of the level densities due to nuclear vibration and rotation. The formalism uses the super-fluid model below critical excitation energy (when the EMPIRE specific parametrization of the level density parameter is selected) and the Fermi gas model above. Differently from other similar formulations, the latter one accounts explicitly for the rotation induced deformation of the nucleus, which becomes spin dependent. The deformation enters level densities formulas through moments of inertia and through the level density parameter a that increases with increase in the surface of the nucleus. Assuming that the prolate nuclei rotate along the axis perpendicular to the symmetry axis the explicit level density formulas reads

$$\begin{aligned}\rho(E,J,\pi) &= \frac{1}{16\sqrt{6\pi}}\left(\frac{\hbar^2}{\Im_{\|}}\right)^{\frac{1}{2}} a^{1/4} \sum_{K=-J}^{J}\left(U-\frac{\hbar^2K^2}{2\Im_{eff}}\right)^{-\frac{5}{4}} \\ &\exp\left\{2\left[a\left(U-\frac{\hbar^2K^2}{2\Im_{eff}}\right)\right]^{\frac{1}{2}}\right\}. \end{aligned} \tag{4}$$

In the case of the oblate nuclei which are assumed to rotate parallel to the symmetry axis we have

$$\begin{aligned}\rho(E,J,\pi) &= \frac{1}{16\sqrt{6\pi}}\left(\frac{\hbar^2}{\Im_{\|}}\right)^{\frac{1}{2}} a^{1/4} \\ &\sum_{K=-J}^{J}\left(U-\frac{\hbar^2\left[J\left(J+1\right)-K^2\right]}{2|\Im_{eff}|}\right)^{-\frac{5}{4}} \\ &\exp\left\{2\left[a\left(U-\frac{\hbar^2\left[J\left(J+1\right)-K^2\right]}{2|\Im_{eff}|}\right)\right]^{\frac{1}{2}}\right\}, \end{aligned} \tag{5}$$

where a is a level density parameter, J is a nucleus spin and K its projection, E is the excitation energy and U is the excitation energy less pairing (Δ). The effective moment of inertia $\Im_{eff}$ is defined in terms of the perpendicular $\Im_{\|}$and parallel $\Im_{\perp}$moments through the difference of their inverses

$$\frac{1}{\Im_{eff}} = \frac{1}{\Im_{\|}} - \frac{1}{\Im_{\perp}} \tag{6}$$

The following parameters have been adjusted compared to the initial fission input file created by EMPIRE for the first run.

- V_i - maxima (and minima) of the real part of the fission potential
- $\hbar\omega_i$ - the associated barrier's width
- a_j - the polynomial coefficients adjusting level densities at saddles (factor of $\rho_i(E, J, \pi)$ in Eq.(5)).

In Tables II to IV, the fission barrier parameters (V_i, $\hbar\omega_i$) and the a_j coefficient from the literature are compared to the parameters chosen for the present calculations for each americium isotope.

The parameters referred as "This work" were used as input values for the present calculations. They were obtained by fitting the experimental data presented in the next section. In the case of ^{243}Am(n,f) (or ^{244*}Am), two separate sets of experimental data exist. Both were fitted with the EMPIRE calculations with, mainly, different values for the barrier heights. In the following, the fission cross sections are calculated up to 20 MeV, considering the (n,f), (n,n'f) and (n,2nf) reactions.

V. EMPIRE PREDICTIONS FOR ^{240}Am(n,f)

A. Fission cross section

As ^{240}Am has a short half-life ($\simeq$ 55 h.), only one direct measurement exists [16]. There is only one direct measurement at 3 MeV by H. C. Britt *et al.* [16]. Another source of experimental data is from the indirect measurements of (n,f) reaction from ^{240}Pu(^{3}He,*df*) reaction [6] (known as surrogate technique). Different simulations exist in the literature, from 0.3 MeV to 5 MeV [12, 41–43]. Numerical values of the EMPIRE 2.19 calculations can be found in Appendix A.

Results of the present calculations compared with the data from the literature are shown in Fig. 2 from $E_n = 1$ keV to 20 MeV. At the date of this report, the evaluation of ^{240}Am(n,f) from LANL is not yet available. The EMPIRE calculation closely follow the surrogate data and is also in good agreement with the other simulations up to 5 MeV. At higher energy, the

TABLE II: Parameters for ^{239*,240*,241*}Am

^{239*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
6.00	0.8		5.40	0.5				[10]
6.47			4.49	0.5				[11]
6.48			5.22					[12]
6.5			5.2					[16] (Exp.)
6.15	0.8	0.80	4.5	0.49	0.80	2.15	1.00	This work
^{240*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
6.10	0.6		6.00	0.4				[10]
6.78			4.86	0.4				[11]
6.23			5.09					[12]
6.0			5.1					[16] (Exp.)
6.55	0.8	0.50	5.90	0.49	0.30	2.15	1.00	This work
^{241*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
6.00	0.8		5.35	0.5				[10]
6.57			4.72	0.48				[11]
6.33			5.31					[12]
6.5			5.4					[16] (Exp.)
6.35	0.3	0.50	5.10	0.49	0.30	2.15	1.00	This work

second and third chance fission become important and the total fission cross section increases up to almost 2.8 MeV. As no evaluation or calculation exist for neutron energy higher than 5 MeV, the barrier heights for ^{240*}Am are obtained from the ^{239}Am(n,f) reaction.

TABLE III: Parameters for ^{242*,243*}Am

^{242*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
6.32	0.6		5.78	0.4				[10]
6.49			4.72	0.38				[11]
6.35								[40] (Exp.)
6.61	0.8		5.56	0.55				[14]
6.5	0.8		5.7	0.52				[15] (Exp.)
6.02			5.12					[12]
5.9			5.4					[16] (Exp.)
6.30	0.6	0.60	5.88	0.39	0.30	2.12	1.00	This work
^{243*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
6.40	1.0		5.05	0.5				[10]
6.08			4.40	0.47				[11]
6.17	0.6		4.75	0.42				[14]
6.25	0.65		5.65	0.45				[15] (Exp.)
6.08			5.38					[15] (Exp.)
6.11			5.32					[12]
6.3			5.4					[16] (Exp.)
5.80	0.4	0.40	5.55	0.4	0.40	2.80	1.00	This work

B. Estimate of uncertainties

The uncertainties associated to the fission cross section for ^{240}Am can be estimated in different manner. The following approximations were made in the present work:

1. The most sensitive parameters are considered, namely the barrier heights V_1 and V_2 for each compound nucleus ^{241*}Am, ^{240*}Am and ^{239*}Am. The other parameters, the barrier width $\hbar\omega$, level densities, are not taken into account. This is mainly because

TABLE IV: Parameters for ^{244*}Am. Two different sets of parameters are presented here which correspond to the two distinct groups of experimental data.

^{244*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
6.25	0.7		5.9	0.53				[10]
6.06			4.48	0.41				[11]
6.15								[40] (Exp.)
6.42	0.8		5.49	0.55				[14]
6.37	0.8		5.57	0.52				[15] (Exp.)
5.76			5.08					[12]
6.20	1.00	0.50	5.95	0.47	0.70	2.80	1.00	This work
6.07	1.00	0.70	5.89	0.47	0.73	2.80	1.00	This work
^{245*}Am								
V_1 (MeV)	$\hbar\omega_1$ (MeV)	a_1	V_2 (MeV)	$\hbar\omega_2$ (MeV)	a_2	V_i	$\hbar\omega_i$ (MeV)	Reference
5.76			5.19					[12]
5.55	1.00	0.70	5.00	0.47	0.70	2.80	1.00	This work

the influences are of the second order compared to that of the barrier heights (for $\hbar\omega$), or because one parameter is strongly correlated to the other (for the level densities).

2. To evaluate the variation of V_i for ^{241*}Am, the experimental data are taken into account only when more than one measurement by energy bin exists: at $E_n = 3$ and 4 MeV. At these energies, two measurements are reported. First the mean values and standard deviation are calculated (7 % at 3 MeV and 6 % at 4 MeV). Then, V_1 is modified to reproduce the bounderies defined by the standard deviations defined at 3 and 4 MeV. We have found that V_1 and V_2 can independently change by $\pm$ 2 % and (+4 %,-3 %), respectively.

3. In the case of ^{240*}Am and ^{239*}Am (compound nuclei for the second and third chance fission of ^{240}Am(n,f)), the four parameters (V_1^{240*}, V_2^{240*}) and (V_1^{239*}, V_2^{239*}) are assumed to vary with the same amplitude and together with V_1^{241*} and V_2^{241*} for ^{241*}Am.

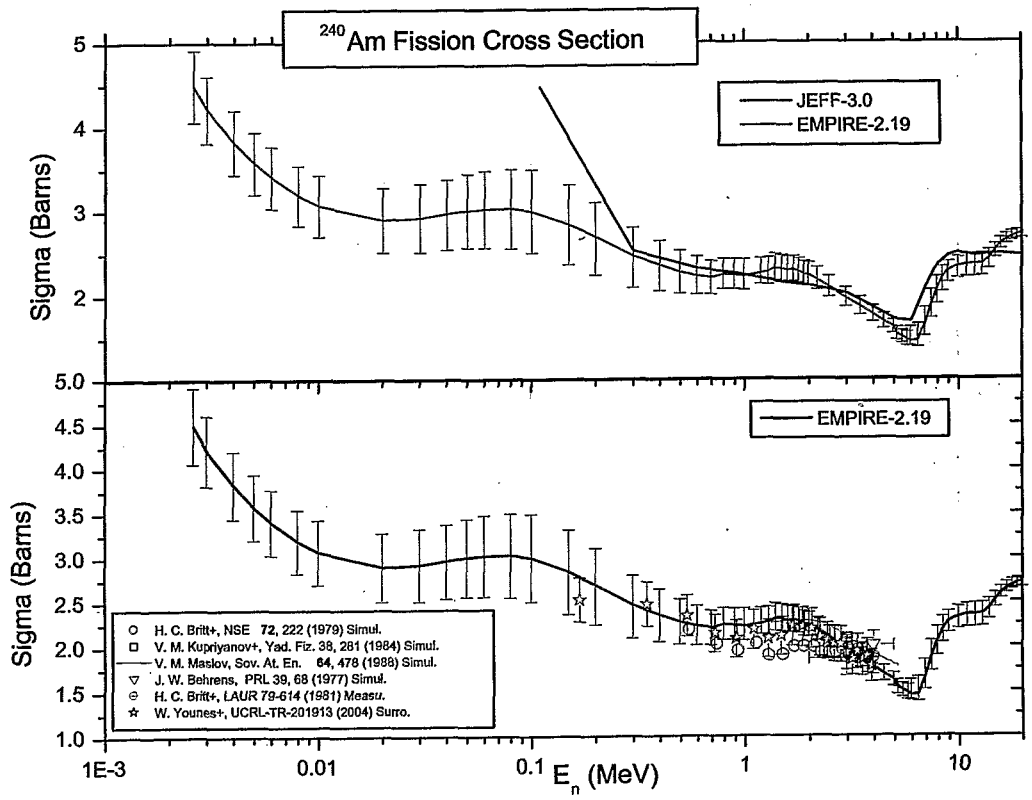

FIG. 2: Calculated neutron-induced fission cross section for ^{240}Am with EMPIRE 2.19 compared to measurements and simulations. Error bars show uncertainties in calculated cross sections as estimated in the present work. See text for references.

The total variation of the V_i parameters was as follow:

For negative variation (a higher barrier height decreases the cross section):

$$\text{Max of} \begin{cases} (V_1^{241*}+2\%)+\ (V_1^{240*}+2\%)+(V_2^{240*}+4\%)+\ (V_1^{239*}+2\%)+(V_2^{239*}+4\%) \\ (V_2^{241*}+4\%)+\ (V_1^{240*}+4\%)+(V_2^{240*}+4\%)+\ (V_1^{239*}+2\%)+(V_2^{239*}+4\%) \end{cases}$$

For positive variation (a lower barrier height increases the cross section):

$$\text{Min of} \begin{cases} (V_1^{241*}-2\%)+\ (V_1^{240*}-2\%)+(V_2^{240*}-3\%)+\ (V_1^{239*}-2\%)+(V_2^{239*}-3\%) \\ (V_2^{241*}-3\%)+\ (V_1^{240*}-3\%)+(V_2^{240*}-3\%)+\ (V_1^{239*}-2\%)+(V_2^{239*}-3\%) \end{cases}$$

The results are presented in Fig. 2 and the numerical values can be found in Appendix A.

Our calculation of the uncertainties is strongly linked to the models used in EMPIRE. For instance, if one uses HMS or DEGAS instead of PCROSS, different set of values for the cross sections and the uncertainties would be obtained. In the future, estimation of uncertainties will be obtained using a more general approach based on Monte Carlo calculations. In such calculations, a larger number of parameters will be taken into account. A Monte Carlo module for covariances and uncertainties calculation is currently under development and will be included in EMPIRE in its next release.

VI. EMPIRE PREDICTIONS FOR OTHER AMERICIUM ISOTOPES

A. Americium 239

Because of its short half-life ($\simeq$ 12 h), cross sections on ^{239}Am are difficult to measure directly. There is no evaluations in the current version of the evaluated libraries (ENDF/B-VI.8, JEFF-3.1, or JENDL-3.3). Only one measurement was performed at 3 MeV by H. C. Britt *et al.* [16]. Different simulations exist in the literature, from 0.3 MeV to 5 MeV [12, 41–43]. Numerical values of the EMPIRE 2.19 calculations can be found in Appendix A. Results of the present calculations compared with the data from the literature are shown in Fig. 3 for E_n = 100 keV to 20 MeV. A good agreement is obtained between 1 and 5 MeV. For neutron energies lower than 1 MeV, the present calculation is lower than the result from Ref. [42], but comparable to Ref. [41]. For neutron energies higher than 5 MeV, no previous calculation exists. The slow increase for the ^{240*}Am in the energy range is comparable to the behavior of the fission cross section for ^{242*}Am (see paragraph VI B).

B. Americium 241

The knowledge of the neutron-induced fission for ^{241}Am is different than for ^{239}Am(n,f) or ^{240}Am(n,f). As its half-life is about 430 years, measurements were done more extensively up to 20 MeV [14, 17–25], with three simulations [41, 44, 45]. Because of the large number of measurements and the good agreement between them, the americium 241 can be considered as a standard to fix the fission barrier heights and the level density parameter a_i for the excited nuclei ^{242*}Am and ^{241*}Am from the first and second chance fission.

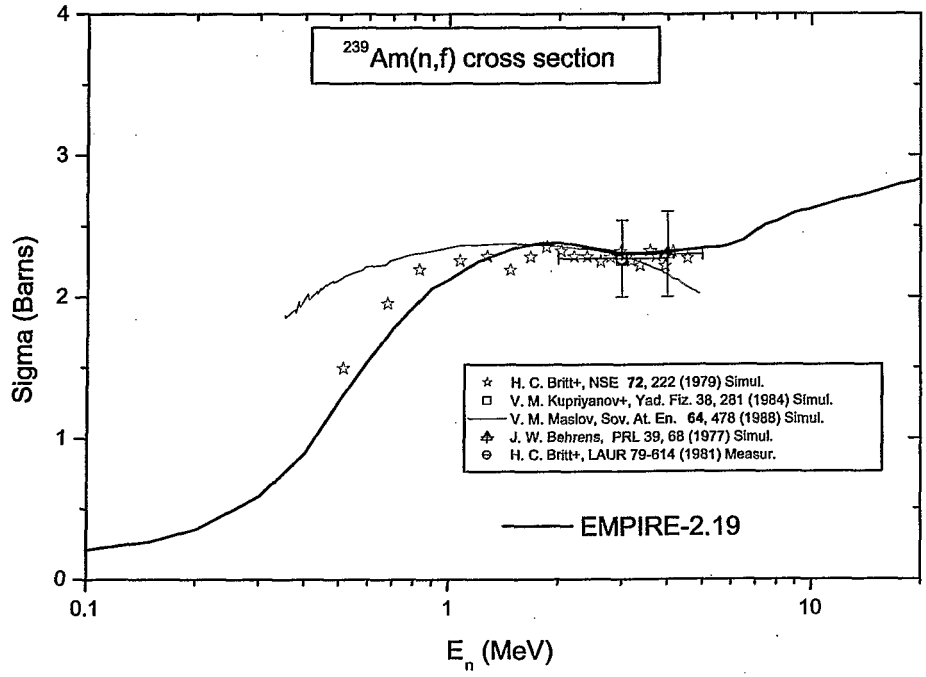

FIG. 3: Calculated neutron-induced fission cross section for ^{239}Am with EMPIRE 2.19 compared to measurements and simulations. See text for references.

At least three evaluated libraries contain data for the fission cross section of ^{241}Am: JEFF-3.1 and ENDF/B-VI.8 and the preliminary version of ENDF/B-VII [46]. All these libraries and the EMPIRE 2.19 calculation are in good agreement up to 15 MeV, but differences appear for higher energies with experimental data and between evaluated libraries. Results of the present calculations compared with the data from the literature are shown in Fig. 4.

C. Americium 242g and 242m

The calculations with EMPIRE 2.19 for the americium 242g and 242m are using the same input files, with a different energy state for the nucleus at rest (ground state for ^{242g}Am and 48.60 keV for ^{242m}Am).

The knowledge of the fission cross section of the ^{242g}Am is poor because of its short half-life ($\simeq$ 16 h). Except for one direct measurement at 14.8 MeV [24], there exists only indirect

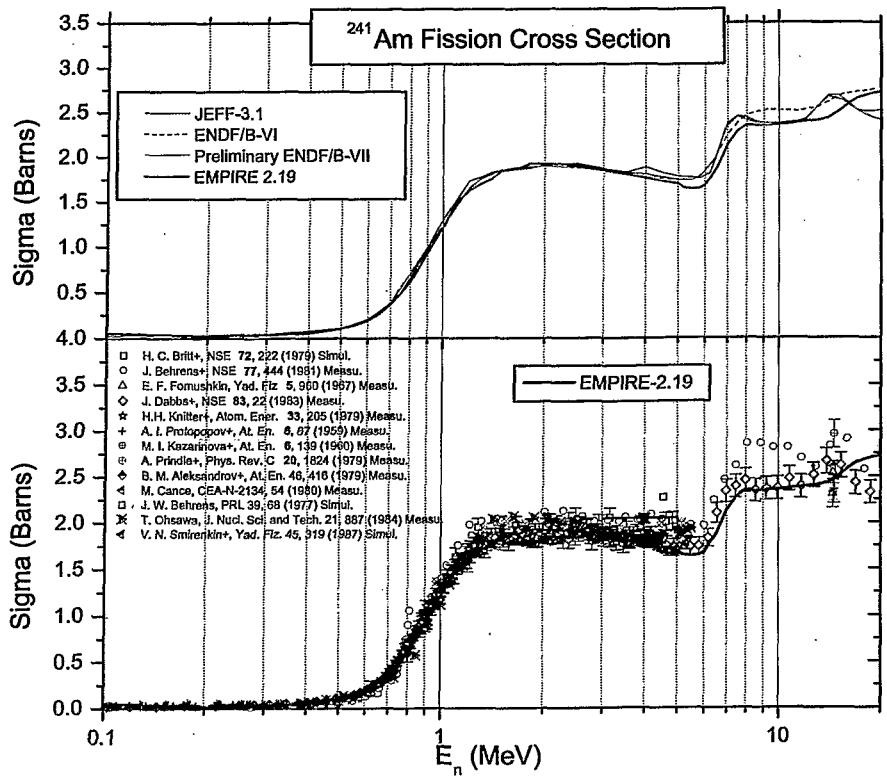

FIG. 4: Calculated neutron-induced fission cross section for ^{241}Am with EMPIRE 2.19 compared to measurements and simulations. See text for references.

measurement by surrogate method (^{242}Pu(^{3}He,df)) up to 5 MeV [6]. The JENDL-3.3 and JEFF-3.1 libraries contain evaluation for the fission cross section of ^{242g}Am that are almost identical. Our calculation presented in Fig. 5 is closer to the experimental data than the previous evaluation. But at high energy ($E_n > 12$ MeV), the EMPIRE 2.19 calculation is strongly increasing compared to JENDL-3.3, being by 0.5 barns higher compared to the measurement at 14.8 MeV.

Because of its longer half-life ($\simeq$ 141 y), more direct measurements exist for ^{242m}Am [16, 24, 26–30] along with one simulation [41]. The experimental data are nevertheless scattered and do not agree, except for the region between 2 and 4 MeV. From the parameters obtained in the case of ^{242g}Am, the EMPIRE calculation follow the surrogate data for ^{242m}Am(n,f) as

presented in Fig. 6. At high neutron energy ($E_n > 5$ MeV), the present calculation is higher than the three different measurements, and it is closer to the evaluated libraries. Numerical values from calculated cross sections are presented in Appendix A.

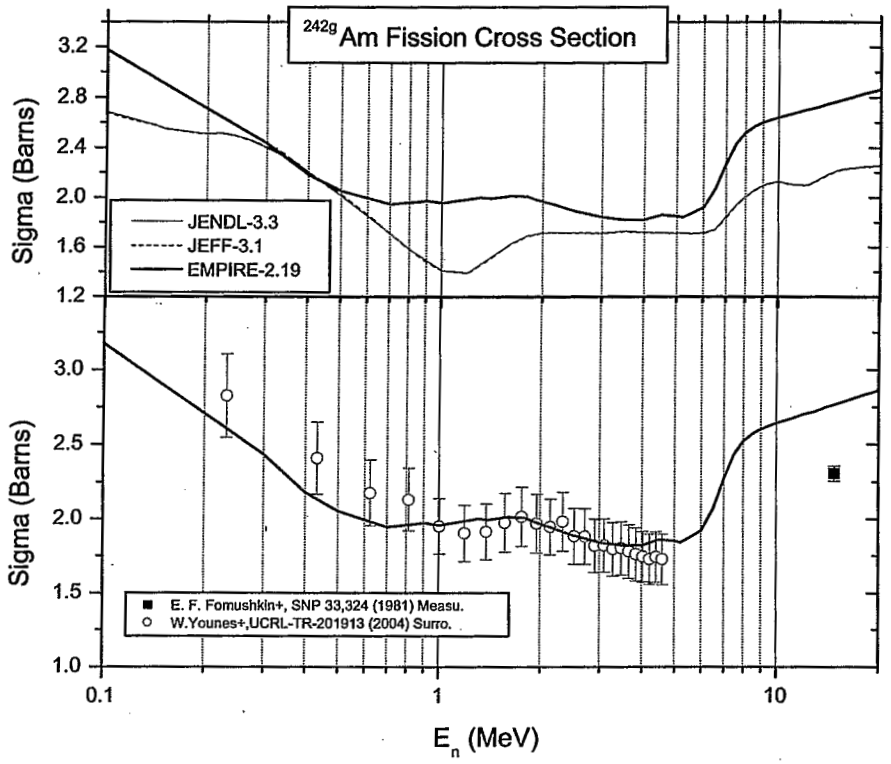

FIG. 5: Calculated neutron-induced fission cross section for ^{242g}Am with EMPIRE 2.19 compared to measurements and simulations. See text for references.

D. Americium 243

Americium 243 has been extensively studied, because of its availability (longest half-life for the americium isotopes). There are 12 different measurements for $E_n > 0.1$ MeV [4, 5, 17, 18, 24, 31–34, 41, 47, 48] and three simulations [14, 16, 49]. The analysis of experimental data (taken directly from the publications or after their renormalization to the new standards) shows that there are noticeable systematic discrepancies in the energy range 1.5-6 MeV between two groups of measurements, separated by 0.2 barns. As noticed in

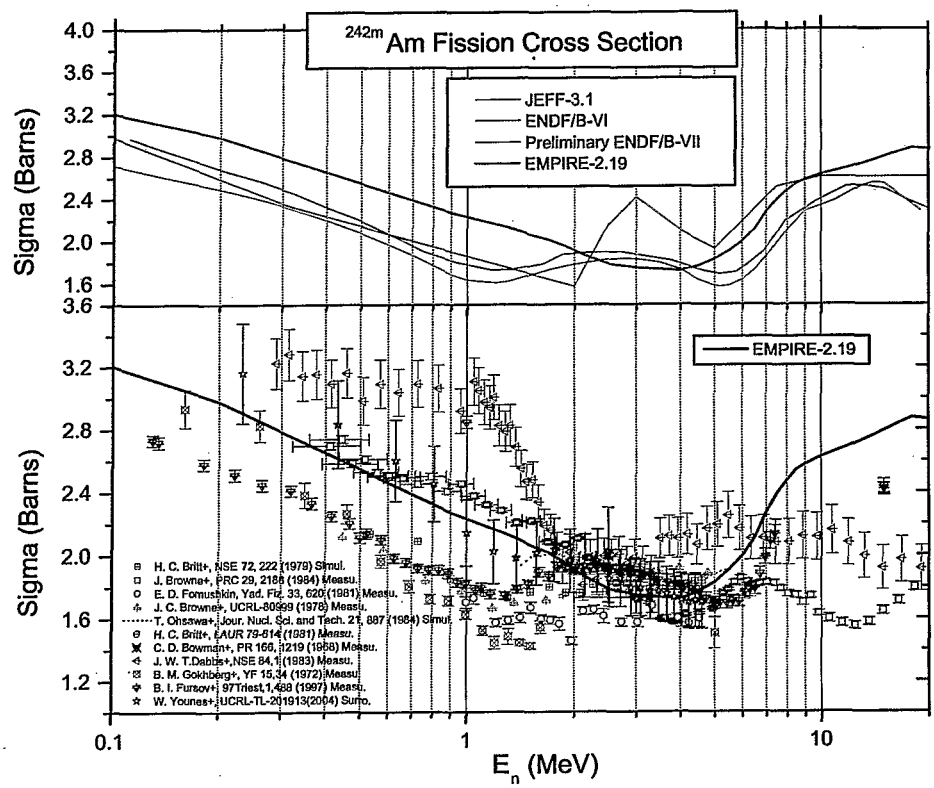

FIG. 6: Calculated neutron-induced fission cross section for ^{242m}Am with EMPIRE 2.19 compared to measurements and simulations. See text for references.

Ref. [50], the average cross section obtained with the ZEBRA reactor spectrum, calculated on the basis of the data from Refs. [32, 33] (lower set of experimental data), are in good agreement with the results of the integral experiment [51]. A similar calculation with the use of the data from Ref. [17] (higher set of experimental data) gives average cross sections higher than the results from the integral measurement by 15 %. Results of our calculations are presented in Fig. 7. The parameters (first and second barrier heights) were modified to reproduce the two sets of data and they are presented in Table III.

Concerning the evaluations, all the libraries (ENDF/B-VI and JEFF-3.1) where fission for americium 243 is included follow the lower set of data. A description of the barrier height systematics is given in section VII.

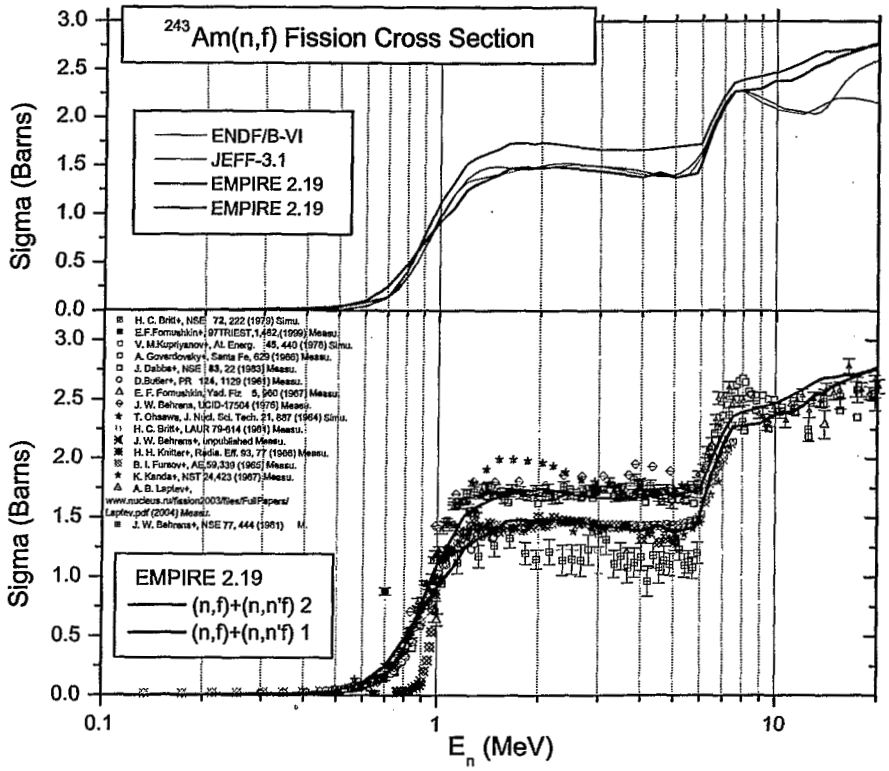

FIG. 7: Calculated neutron-induced fission cross section for ^{243}Am with EMPIRE 2.19 compared to measurements and simulations. See text for references.

E. Americium 244g

Because of its short half-life ($\simeq$ 10 h), direct measurements of the fission cross section of americium 244 are rare [41]. The surrogate method provides a set of indirect data from 0.1 to 5 MeV, in good agreement with the measurement and simulation from H. C. Britt [16, 41]. The present calculation (see Fig. 8) is also in good agreement with the other data, up to 5 MeV. For higher energies, as there is no measurement, the value of the second barrier height is obtained with a large uncertainty (see Fig. 10).

The two libraries, where evaluation of ^{244g}Am(n,f) are available, are almost identical and slightly different than the measurements and simulation from Refs. [6, 16, 41].

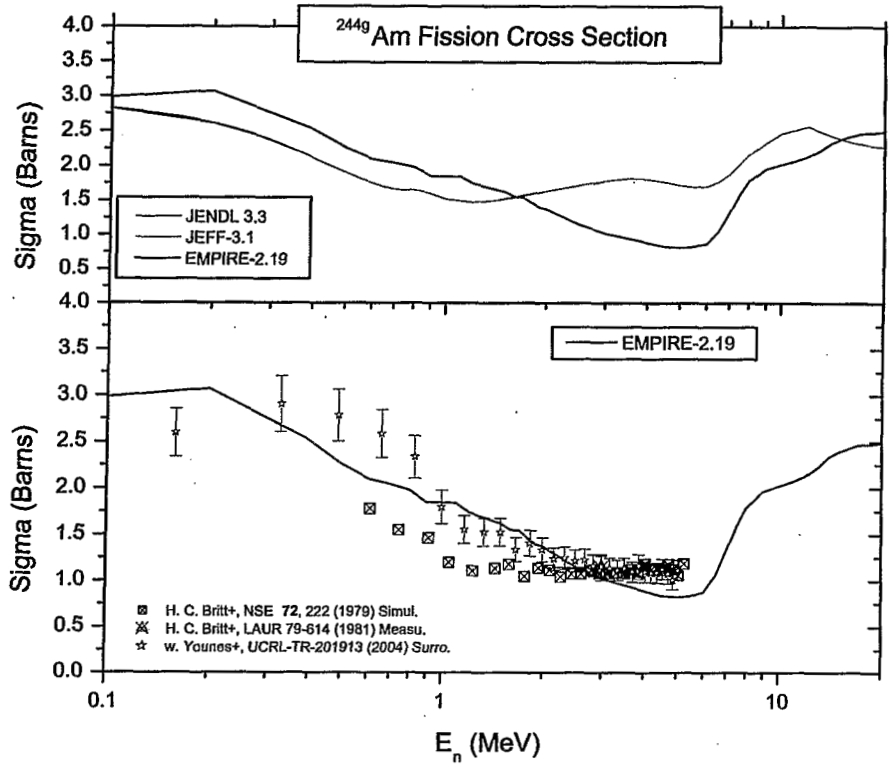

FIG. 8: Calculated neutron-induced fission cross section for ^{244g}Am with EMPIRE 2.19 compared to measurements and simulations. See text for references.

VII. SYSTEMATICS OF THE FISSION BARRIER PARAMETERS

In order to study the systematics of barrier heights for the americium isotopes, the values used in the EMPIRE calculations are presented in Figs. 9 and 10. General trends found here are common to almost all actinides [14].

The values of the first barrier height V_1 increases as a general trend with increasing neutron number N up to $N = 145 - 146$, and it decreases as N goes further over 147. This value is very close to $N = 146 \simeq 148$ for which fission isomers have the longest half-lives. This indicates that the second well is deeper for the americium nuclei in this region. It is also reported that there is a subshell region around $N = 146$ [52], where the shell correction to the deformation energy surface deepens the second well and raises the height of the first

barrier.

Contrary the the first barrier height, the second barrier height V_2 is observed to decrease monotonically as a function of N and A, as presented in Fig. 10. The barrier height for $N = 150$ (or $A = 245$) was difficult to assess in the present study. This is because only one measurement is available at $E_n < 10$ MeV for the system ^{244}Am(n,f) [38] (determination of V_2 via the first chance fission) and different values of V_2 lead to equivalent results. Related uncertainty is shown in Fig. 10 for $N = 150$.

In general, the barrier heights for americium isotopes with odd-N are higher than those for even-N. The mean value of the barrier heights for odd-N nuclei is typically by $0.4 \simeq 0.7$ MeV higher than for even-N nuclei. This is in agreement with the value of $\simeq 0.5$ MeV deduced from the even-odd differences for other actinides [14].

In the case of ^{243}Am(n,f), two distinct experimental data sets exist that are separated in the energy range 1.5-6 MeV by 0.2 barns. Both sets were fitted with EMPIRE 2.19 using two different binomes for (V_1,V_2), shown as solid and dot lines in Figs. 9 and 10. The highest values of (V_1,V_2) correspond to the lower calculation (in black in Fig. 7) and the lowest values of (V_1,V_2) correspond to the highest calculation (in red in Fig. 7). One can expect that the values for (V_1,V_2) from the EMPIRE calculation could give an indication on the preferred set of measurement by comparing the barrier heights between the other americium isotopes or with the other values from the literature for ^{243}Am. From the first barrier height, the small difference in the two values of the EMPIRE calculations ($V_1^l = 6.06$ and $V_1^h = 6.20$ MeV) does not change enough the slope of V_1 as a function of N (Fig. 9) to modify the systematics. Furthermore, the mean V_1 obtained from the data in the literature (6.13 MeV) is between V_1^l and V_1^h.

Some indication for prefered datacomes from the comparison of the height of the second barrier V_2. It was noticed that V_2 is decreasing with N increasing. Following this trend, the value for the calculation corresponding to the lower set of data ($V_2^l = 5.85$ MeV) is in better agreement than the values fitting highest set of data ($V_2^h = 5.89$ MeV). Nevertheless, because of the small difference between V_2^l and V_2^h (less than 1 %), the present conclusion is a slight indication in favor of the highest set of experimental data. A strongest indication comes from the integral measurement with the ZEBRA reactor spectrum [51], which is in agreement with the highest set of data.

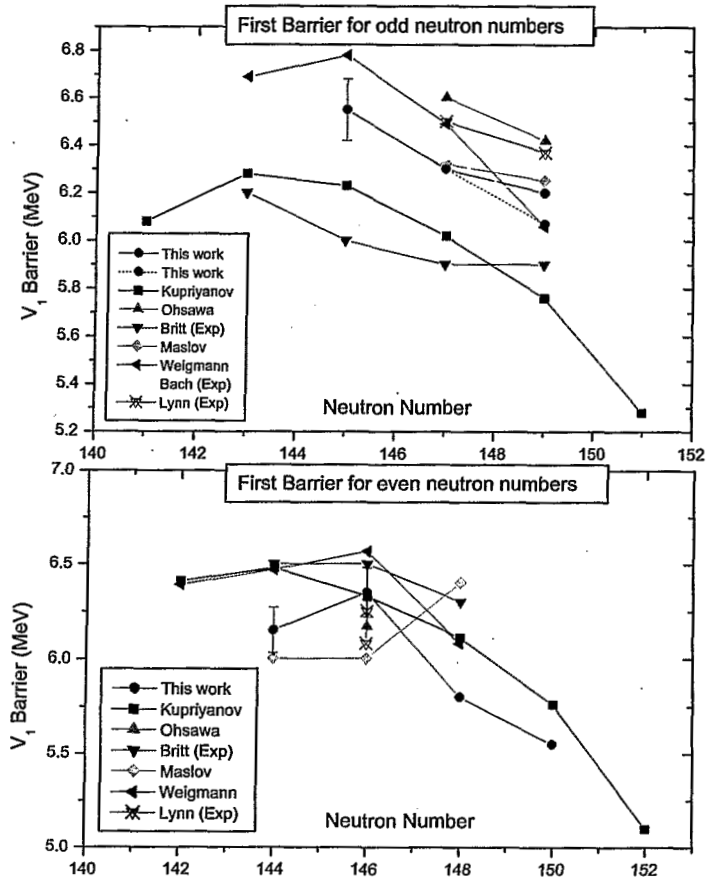

FIG. 9: Comparison of the first barrier height for the even and odd neutron number of americium isotopes. The uncertainties on the barrier heights for $N = 144$, 145 and 146 come from the estimation of uncertainties for ^{240}Am(n,f), see section V A. See references in the text.

VIII. CONCLUSION

The nuclear reaction code EMPIRE code was extended to account for advanced modeling of the fission process, incorporated in the new version 2.19. Calculations of fission cross sections were performed for ^{240}Am(n,f) from 1 keV to 20 MeV and for other americium isotopes from 100 keV to 20 MeV. Uncertainties in the case of ^{240}Am(n,f) were estimated for the fission cross sections and the barrier heights.

The fission barrier heights for the americium isotopes used in the EMPIRE calculations were compared to the values from the literature and they appear to fit the different systematics.

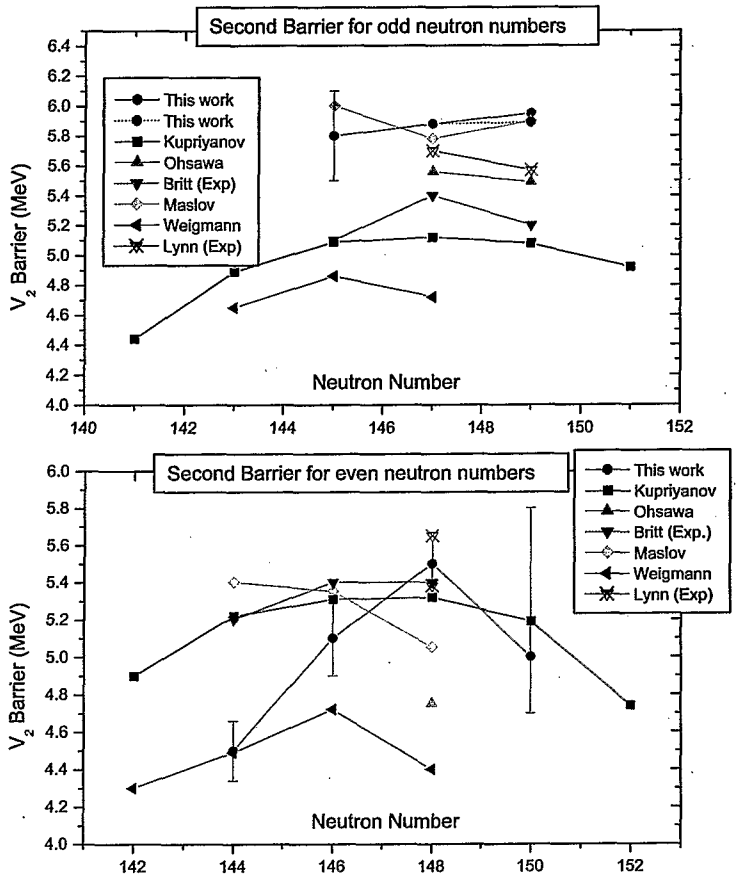

FIG. 10: Comparison of the second barrier height for the even and odd neutron number of americium isotopes. The uncertainties on the barrier heights for $N = 144$, 145 and 146 come from the estimation of uncertainties for ^{240}Am(n,f), see section V A. See references in the text.

Specific EMPIRE level densities were used, modified for the first and second barrier.

In the case of sparse experimental data or for disagreement between data, as for all the americium isotopes except ^{241}Am, we present values used in our calculations, according to the models taking into account in EMPIRE-2.19.

In the future, further investigations are needed in order to extend these calculations at higher neutron energies and to estimate the uncertainties of fission cross sections with a Monte Carlo approach.

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APPENDIX A: NUMERICAL FISSION CROSS SECTIONS CALCULATED WITH EMPIRE 2.19

TABLE V: Fission cross section for $E_n = 0.01$ to 0.08 MeV for ^{240}Am(n,f).

E_n MeV	^{240}Am(n,f) Barns	E_n MeV	^{240}Am(n,f) Barns
0.001	6.63	0.01	$3.07^{+0.36}_{-0.37}$
0.002	$4.93^{+0.42}_{-0.43}$	0.02	$2.90^{+0.38}_{-0.39}$
0.003	$4.22^{+0.38}_{-0.41}$	0.03	$2.92^{+0.40}_{-0.41}$
0.004	$3.83^{+0.37}_{-0.39}$	0.04	$2.97^{+0.40}_{-0.43}$
0.005	$3.58^{+0.36}_{-0.38}$	0.05	$3.01^{+0.43}_{-0.45}$
0.006	$3.41^{+0.36}_{-0.38}$	0.06	$3.02^{+0.45}_{-0.46}$
0.008	$3.19^{+0.35}_{-0.36}$	0.08	$3.03^{+0.46}_{-0.48}$

TABLE VI: Fission cross section for $E_n = 0.1$ to 4.0 MeV. In the case of ^{243}Am(n,f), two different calculations are presented to reproduce both sets of experimental data.

E_n	^{239}Am(n,f)	^{240}Am(n,f)	^{241}Am(n,f)	^{242g}Am(n,f)	^{242m}Am(n,f)	^{243}Am(n,f)		^{244}Am(n,f)
MeV	Barns	Barns	Barns	Barns	Barns	Barns		Barns
0.1	0.21	$3.00^{+0.49}_{-0.49}$	0.00	3.17	3.20	0.00	0.00	2.98
0.2	0.35	$2.69^{+0.41}_{-0.45}$	0.01		2.98	0.00	0.00	3.07
0.3	0.59	$2.46^{+0.33}_{-0.37}$	0.02	2.43			0.00	
0.4	0.89	$2.35^{+0.29}_{-0.30}$	0.04	2.18		0.02	0.01	2.52
0.5	1.27	$2.27^{+0.25}_{-0.25}$	0.10	2.05		0.04	0.03	2.26
0.6	1.56	$2.23^{+0.22}_{-0.22}$	0.19	1.99		0.10	0.04	2.10
0.7	1.78	$2.20^{+0.20}_{-0.19}$	0.35	1.94		0.25	0.14	2.04
0.8	1.93	$2.24^{+0.18}_{-0.17}$	0.61			0.48	0.39	1.98
0.9	2.06	$2.23^{+0.19}_{-0.16}$	0.92	1.97	2.27	0.72	0.78	1.85
1.0	2.13	$2.22^{+0.20}_{-0.16}$	1.20	1.96		0.92	1.12	
1.1	2.19		1.50			1.06	1.32	1.84
1.2	2.25	$2.28^{+0.18}_{-0.12}$	1.62			1.24	1.52	1.74
1.3	2.28	$2.27^{+0.17}_{-0.15}$	1.70	2.00	2.13	1.32	1.59	
1.4	2.31	$2.31^{+0.17}_{-0.14}$	1.75	1.99	2.10	1.37	1.64	
1.5	2.34	$2.30^{+0.16}_{-0.14}$	1.85		2.06	1.41	1.67	1.62
1.6	2.36	$2.29^{+0.16}_{-0.14}$	1.86	2.01	2.01	1.47	1.73	1.55
1.7	2.37	$2.30^{+0.16}_{-0.14}$	1.87			1.48	1.73	1.54
1.8	2.38	$2.27^{+0.16}_{-0.13}$	1.92	2.01	1.95	1.48	1.73	1.46
1.9	2.38	$2.24^{+0.15}_{-0.13}$	1.92	1.98		1.48	1.72	1.40
2.0	2.38	$2.23^{+0.14}_{-0.13}$	1.91			1.50	1.74	1.37
2.2	2.37	$2.17^{+0.14}_{-0.11}$	1.92	1.93	1.85	1.49	1.73	
2.5	2.34	$2.08^{+0.13}_{-0.12}$	1.90	1.89	1.78	1.47	1.69	1.15
3.0	2.31	$1.95^{+0.12}_{-0.10}$	1.85	1.84	1.75	1.44	1.66	1.02
3.5	2.31	$1.85^{+0.11}_{-0.10}$	1.80	1.82				
4.0	2.32	$1.77^{+0.10}_{-0.09}$	1.76	1.82	1.72	1.39	1.67	

TABLE VII: Fission cross section for $E_n = 4.0$ to 20.0 MeV.

E_n MeV	^{239}Am(n,f) Barns	^{240}Am(n,f) Barns	^{241}Am(n,f) Barns	^{242g}Am(n,f) Barns	^{242m}Am(n,f) Barns	^{243}Am(n,f) Barns	^{244}Am(n,f) Barns
4.5	2.33	$1.68^{+0.09}_{-0.08}$	1.72	1.86	1.77	1.43 1.68	0.83
5.0	2.34	$1.62^{+0.08}_{-0.08}$	1.70	1.85	1.83	1.38 1.70	0.82
5.2	2.35	$1.54^{+0.08}_{-0.08}$	1.69	1.84	1.86		0.82
5.5	2.35	$1.50^{+0.08}_{-0.07}$	1.64	1.87	1.90	1.38	0.83
5.8	2.36	$1.47^{+0.09}_{-0.07}$	1.64	1.90	1.96	1.41	0.86
6.0	2.37	$1.46^{+0.16}_{-0.06}$	1.67	1.92	1.99	1.54 1.72	0.87
6.5	2.40	$1.46^{+0.19}_{-0.08}$	1.86	2.07	2.11	1.88 1.99	1.05
7.0	2.46	$1.65^{+0.19}_{-0.14}$	2.12	2.27	2.27	2.12 2.21	1.32
7.5	2.51	$1.88^{+0.17}_{-0.16}$	2.27	2.43	2.39	2.27 2.36	1.58
8.0	2.53	$2.06^{+0.14}_{-0.17}$	2.35	2.52	2.47	2.28 2.39	1.78
8.5	2.56	$2.19^{+0.13}_{-0.16}$	2.33	2.57	2.54	2.29 2.41	1.87
9.0	2.59	$2.27^{+0.11}_{-0.13}$	2.34	2.60	2.57	2.30 2.43	1.96
10.0	2.62	$2.33^{+0.11}_{-0.14}$	2.35	2.64	2.62	2.33 2.47	2.02
11.0	2.65	$2.35^{+0.11}_{-0.14}$	2.37	2.67		2.39 2.52	2.07
12.0	2.68	$2.35^{+0.12}_{-0.14}$	2.38	2.70	2.69	2.45 2.58	2.13
13.0	2.70	$2.36^{+0.12}_{-0.14}$	2.40	2.72		2.53 2.64	2.22
14.0	2.72	$2.42^{+0.10}_{-0.12}$	2.43	2.75	2.76	2.57 2.68	2.33
15.0	2.74	$2.52^{+0.07}_{-0.09}$	2.50	2.77		2.62 2.69	2.39
16.0	2.76	$2.60^{+0.06}_{-0.07}$	2.58		2.82	2.71	2.43
17.0	2.78	$2.64^{+0.06}_{-0.05}$	2.64			2.69 2.70	2.46
18.0	2.80	$2.67^{+0.06}_{-0.04}$	2.68	2.83	2.87	2.73	2.47
19.0	2.81	$2.70^{+0.06}_{-0.05}$	2.70			2.74	2.48
20.0	2.83	$2.71^{+0.05}_{-0.05}$	2.71	2.86	2.92	2.77 2.77	2.49