27
2/2/84
(10)

(1)
I-13113

DR 2096-6

UCID--19980

DE84 006264

Nuclear Modeling of the $^{239}\text{Pu}(n,xn)$
Excitation Function

M. Blann and R. M. White
Experimental Physics Division
Lawrence Livermore National Laboratory
University of California
Livermore, CA 94550

January 1984

This is an informal report intended primarily for internal or limited external distribution. The opinions and conclusions stated are those of the author and may or may not be those of the Laboratory.

Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore Laboratory under Contract W-7405-Eng-48.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Nuclear Modeling of the $^{239}\text{Pu}(n,xn)$ Excitation Function

M. Blann and R. M. White
Experimental Physics Division
Lawrence Livermore National Laboratory
University of California
Livermore, CA 94550

ABSTRACT

Calculated excitation functions for the $(n,2n)$ and $(n,3n)$ reactions on ^{239}Pu are presented. In these calculations precompound plus compound decay models with experimentally deduced fission probabilities are used. A range of results are presented indicating the uncertainty in the model calculations. Experimental measurements which would constrain the range of modeled result are suggested.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

I. INTRODUCTION

The measurement of the $^{239}\text{Pu}(n,2n)$ excitation function provides a great experimental challenge. Several groups have spent years on the problem, some without achieving definitive results. Measurements have been published by the Aldermaston group,¹ and the cross section in the region of 14 MeV incident neutron energy has been measured by R. Lougheed and collaborators.²

It is desirable to supplement experimental data with calculated results based on nuclear modeling. A sophisticated treatment has recently been performed by E. Arthur.^{3,4} In this report we present a calculation which is more empirical than Arthur's, yet in many ways similar. We will try not so much to present a single calculation, but a range of results spanning uncertainties in modeling input. We will also suggest, in the context of our calculations, experimental measurements which will further constrain the range of modeled results.

II. EXPERIMENTAL RESULTS USED IN CALCULATIONS

The calculations to be performed should be constrained to agree with what is known experimentally about the relevant Pu isotopes. Available data include measured and evaluated $^{239}\text{Pu}(n,f)$ fission cross sections for neutron energies from thermal to 20 MeV. We will use the recently re-evaluated results of Howerton and White.⁵ The confidence level is high for these results ($\pm 3\%$) up to neutron energies of ~ 15 MeV; above 15 MeV experimental results are far less certain.

Our model calculations require us to know the fission probabilities of all relevant Pu isotopes versus excitation energy. We define the fission probability, $P_f(E)$, as the fraction of an ensemble of a given isotope at some excitation energy E which de-excite by fission rather than by other decay modes such as neutron evaporation or photon emission. The $^{239}\text{Pu}(n,f)$ cross section gives us information on the fissioning of the compound nucleus ^{240}Pu . The fission cross section has also been measured for $^{238}\text{Pu}(n,f)$ at neutron energies up to 10 MeV.⁶ These data give us information and provide experimental constraints for de-excitation of the ^{239}Pu compound nucleus.

We are not aware of similar measurements for compound nuclei $^{238,237}\text{Pu}$. However, fission probabilities versus excitation energy have been measured for these isotopes using transfer reactions to excite compound nuclei at known excitation energies.⁷ We refer to Ref. 7 for details of the experimental measurement; we will show results later on in this work.

These data (Refs. 5-7) will provide the basis of experimental constraint for our calculations, which require fission probabilities (rather than cross sections) for first chance fission of all relevant Pu isotopes. This means that fission cross sections must be divided by a compound nucleus cross

section in order to generate probabilities. These results will represent first chance fission only in an excitation range in which fission cannot compete following the re-emission of a neutron. At these points nuclear modeling must enter the calculation. The following sections therefore deal with steps in modeling required to reach the final results, the $^{239}\text{Pu}(n,2n)$ and $(n,3n)$ excitation functions.

III. REACTION CROSS SECTIONS

Conversion of $^{239,238}\text{Pu}$ fission cross sections to $^{240,239}\text{Pu}$ (compound nucleus) fission probabilities requires division by the compound nucleus cross section appropriate to each nucleus and incident neutron energy. Calculations of total reaction cross sections for $^{239}\text{Pu} + n$ were performed by Arthur using a deformed optical potential, with corrections via coupled channel calculations for direct reactions to low lying rotational levels.^{3,4,6} The code used in our calculations has a spherical optical model routine, which uses a global parameter set, described in some detail in Ref. 9.

Reaction cross sections vs. neutron energy calculated from these two sources are compared in Fig. 1. Experimental measurement of the reaction cross sections by MacGregor et al.¹⁰ are also shown. The ratio of the cross sections is shown in the lower part of the figure; agreement is quite good in the 13-16 MeV region, and in agreement with the experimental results. Disagreement at the 10% level exists for neutron energies of ~5 to 9 MeV.

For most of the results presented in this work we used the reaction cross sections due to Arthur to convert the fission cross sections to fission probabilities (240 and ^{239}Pu). We used the reaction cross sections from the spherical optical model in calculating the (n,xn) excitation functions, since this was the most convenient for our code (Alice/Livermore). Had the cross sections due to Arthur been used consistently throughout, final results would scale linearly as the ratio of deformed to spherical cross sections, i.e. the resulting (n,xn) cross sections would be 5-10% lower in the 4-12 MeV neutron region.

Some evidence favors larger reaction cross sections in the 4-12 MeV than those given by the deformed optical potential region, e.g. the experimental point of MacGregor et al. at 8 MeV neutron energy. Also, the measured $^{238}\text{Pu}(n,f)$ cross sections of Ref. 6 exceed the cross sections we estimate from the deformed potential reaction cross sections above 7 MeV neutron energy, whereas they must of course be less. For these reasons we will present one set of results for which the spherical optical model reaction cross sections were used throughout. This will illustrate the manner in which uncertainties in reaction cross sections are 'leveraged' into larger uncertainties in final calculated excitation functions.

IV. MODEL CALCULATIONS

A. General Discussion

The calculations performed used a modified version of the nuclear reaction code ALICE/LIVERMORE 82.¹¹ There are three relevant parts of this code with respect to the goals of this work:

- (1) Following interaction of the incident neutron with the ^{239}Pu target nucleus, one or more prompt (inelastic) neutrons may be emitted. The time scale for this process is very short compared with the equilibration time and with the collective oscillation period required for nuclear fission. The neutrons emitted in this process are on the average higher in energy than those evaporated later from the equilibrated system. The model used in the ALICE code for this calculation is referred to as a precompound decay model¹²; in particular the geometry dependent hybrid model^{13,14} was used, with global default parameters, i.e. no parameter adjustment.
- (2) The evaporation model due originally to Weisskopf¹⁵ is used in the evaporation stages of the calculation, to give the energy spectrum of the several (up to four) successively emitted neutrons. Default parameter sets were used for these calculations, i.e. no parameters were varied from those provided by the code.

- (3) For heavy elements such as Pu, fission competes with particle evaporation when both are possible. Ordinarily the fission probability is calculated in ALICE using the Bohr-Wheeler approach¹⁶ in a statistical fashion.

The physics models contained in the ALICE code, along with detailed descriptions of the input, and comparisons with a broad range of data, are to be found in the literature.^{11,17} Two major modifications were made prior to performing the calculations of this work. First, the ALICE code uses a 1 MeV mesh size pointwise calculation. The code was modified to accept variable mesh size in order to reduce uncertainties due to integration steps. All results to be presented here have been performed using a 0.1 MeV mesh size.

The second modification was to use tabular $P_f(E)$ for each relevant isotope [^{240,239,238,237}Pu] rather than statistically calculated results. This permits more direct input of experimentally constrained $P_f(E)$ values than is given by the statistical input of the original code. There are still uncertainties in the fission probabilities where they must be extrapolated beyond the first chance fission only region. We will return to this point shortly.

B. Comparisons of Particle Spectra for Non-fissile Systems

If the calculation we perform uses good fission probabilities and reaction cross sections, and if it predicts good precompound and evaporation spectra, it should also yield good predictions for the (n,xn) excitation functions. Many comparisons of precompound + evaporation spectra were presented in Ref. 9. We reproduce the experimental and calculated

$^{93}\text{Nb}(n,xn)$ spectrum for 14 MeV incident neutron energy from Ref. 9 as our Fig. 2. We also show recent $^{197}\text{Au}(p,n)$ spectra¹⁷ plus calculated results as an example of a heavy element at a different excitation energy (Fig. 3). All calculations are absolute and un-normalized. Based on these comparisons we expect that the spectra for $^{239}\text{Pu}(n,xn)$ are satisfactorily predicted by our code; however we would emphasize that the ability to compare our calculated spectra with experimental results of spectra from Pu targets would greatly increase our ability to eliminate uncertainties in the modeled excitation functions.

C. Fission Probabilities

The new ENDL evaluation for $^{239}\text{Pu}(n,f)$ and reaction cross sections calculated by Arthur were used to obtain the fission probability of the ^{240}Pu compound system. These results are shown in Fig. 4. For the fission probability of the ^{239}Pu compound system, $^{238}\text{Pu}(n,f)$ cross section measurements have been reported for neutron energies up to 10 MeV.⁶ Smooth curves were drawn through these data and values selected at 1 MeV intervals were divided by the corresponding calculated reaction cross sections to give fission probabilities. These results are shown in Fig. 5.

For $^{238,237}\text{Pu}$, fission probabilities were measured experimentally (as opposed to experimental measurement of fission cross sections and conversion to probabilities as we have done for $^{240,239}\text{Pu}$). These results are shown in Figs. 6 and 7.

The fission probabilities to be entered into our calculations must be for first chance fission only. Therefore we use the experimentally measured or deduced results in the energy region corresponding to first chance fission

only; for higher excitation energies we must extrapolate these values, thereby introducing some uncertainty into our results. Note however that we still have the experimental constraint that the sum of the various fission contributions must sum to the total fission observed for the $^{239}\text{Pu}(n,f)$ excitation function (see Fig. 4).

The fission probabilities which we show in Fig. 4 are not really the results needed as input for our calculations, for we have divided fission cross sections by modeled reaction cross sections to generate those probabilities. Really we need to use fission probabilities which result from division of the fission cross sections by the equilibrated compound nucleus cross sections. The compound nucleus cross sections will be less than the reaction cross sections, since a part of the reaction cross sections will be depleted by the fast precompound decay mechanism prior to forming equilibrated compound nuclei. We have therefore applied small ($\leq 15\%$) corrections for this effect in the fission probabilities used as input for the calculations to be presented.

The first chance fission probabilities so deduced are shown by curves in Figs. 4-7. Beyond the first chance only region for $P_f(E)$ we must extrapolate for first chance only results. The $P_f(E)$ should not show a precipitate change in value, but rather a smooth variation with energy. We have therefore made three different assumptions on the extrapolated behavior of $P_f(E)$, which are shown as broken lines in Figs. 4-7:

- (a) The $P_f(E)$ reach a peak value and are thereafter constant with increasing energy,
- (b) the $P_f(E)$ show a very gradual decrease with energy beyond the estimated peak value,

- (c) the $P_f(E)$ show a fairly steep decline beyond the estimated peak values.

Quantitative illumination of the qualitative commentary of (a) - (c) above may be seen in the broken curves of Figs. 4-7.

How well the $P_f(E)$ sets satisfy the constraint of the total $^{239}\text{Pu}(n,f)$ excitation function is shown in Figs. 8-10. The agreement with $P_f(E)$ is generally equally good for all three sets of $P_f(E)$ corresponding to sets (a) - (c) with set (c) giving slightly better agreement with experimentally deduced results in the 6-14 MeV region.

C. Results of Excitation Function Calculations

Calculated $^{239}\text{Pu}(n,2n)$ excitation functions using parameter sets (a) - (c) for $P_f(E)$ are shown in Fig. 11; $(n,3n)$ excitation functions are shown in Fig. 12. Also shown in Figs. 11 and 12 are experimental results reported in Refs. 1 and 2. The calculated results were all performed prior to comparison with experimental results, and were in no way constrained or parameterized to force agreement with experimental results. The five points for ^{238}Pu from Ref. 1 were measured by direct neutron counting in a scintillator; the three points between 13.7 and 14.8 MeV due to R. Loughheed² were measured by activation.

If an average result is drawn through the three excitation functions shown, all results are within $\pm 10\%$ of the central result. We consider it unproductive to push the calculations beyond this level of uncertainty. In our judgment this is the noise level of the present model.

We mentioned earlier that the reaction cross sections based on a deformed optical potential were lower in the 6-12 MeV neutron energy region than results of the spherical optical model. We therefore recalculated $P_f(E)$ experimental values for $^{240,239}\text{Pu}$ based on the spherical optical model results shown in Fig. 1. These results are shown in Figs. 13 and 14. The revised $P_f(E)$ used as input in the calculations are also shown in these figures. There was no change in the $^{238,237}\text{Pu}$ results, since these were measured experimentally as fission probabilities rather than cross sections. The resulting total $P_f(E)$ for $^{239}\text{Pu}(n,f)$ from this set of calculations is shown in Fig. 13. The $^{239}\text{Pu}(n,2n)$ and $(n,3n)$ excitation functions resulting from these calculations are shown in Figs. 11 and 12.

This set of calculations is in reasonably good agreement with the constraint of the total $^{239}\text{Pu}(n,f)$ fission probability from 9 to 15 MeV neutron energy. From 6 to 8 MeV the calculated $P_f(E)$ is somewhat lower than the experimentally deduced value. Corrections for this discrepancy would not change results by over 10%. Experimental results which would increase confidence in these model calculations would be activation yields at $E_n = 7$ and 11 MeV, and precompound spectral yields at several energies such as 14 and 18 MeV. Some feel for the believability of these modeled excitation functions may be achieved by comparisons with similar calculations for $^{235}\text{U}(n,2n)$ and $^{238}\text{U}(n,2n)$ excitation functions. These results are shown in Figs. 15 and 16

using reaction cross sections based on the spherical optical model, and based on the spherical optical model reduced by the ratios in Fig. 1 to estimate deformed potential results. Many experimental measurements have been reported for these excitation functions,^{1,18-21}, as indicated in Figs. 15 and 16 [not all $^{238}\text{U}(n,2n)$ experimental results have been shown in Fig. 16]. The comparisons of Figs. 15 and 16 suggest that our calculations are least trustworthy in the first 3 MeV above the threshold, and best in the high energy region. The threshold behavior suggests that a more sophisticated treatment of level densities at low residual excitations should be entered into the ALICE computer code. These comparisons suggest caution in using our modeled $^{239}\text{Pu}(n,2n)$ results below ~ 9 MeV neutron energy, and therefore suggest also that experimental measurements in the threshold to 9 MeV region are highly desirable. Careful scrutiny on total non-elastic cross sections are also essential to the model calculation, and this is an area in which our model results may also be improved.

We present a best estimate of the $^{239}\text{Pu}(n,2n)$ calculated excitation function resulting from the present calculations in Fig. 17, and give numerical results in Table 1. The experimental results of Ref. 1 of $^{239}\text{Pu}(n,2n)$, as shown in Figs. 11 and 17, are questionable as to accuracy.

The $^{239}\text{Pu}(n,2n)$ excitation function at $\epsilon_n \geq 15$ MeV is largely a measure of the precompound calculation and is not sensitive to the fission cross sections. The $(n,3n)$ excitation function, however, is extremely sensitive to fission cross sections in this energy region, and since the evaluated results are somewhat uncertain here, the calculated results must also be considered to be highly uncertain.

III. ACKNOWLEDGEMENTS

The authors of this report greatly appreciate several enlightening discussions with Dr. E. Arthur. We also appreciate data supplied by Dr. Arthur which was used in the calculations reported herein, as noted.

Table 1. Modeled $^{239}\text{Pu}(n,2n)$ Excitation Function

Incident Neutron Energy (MeV)	Calculated (n,2n) Cross Section (mb)
6.0	2.2
6.5	14
7.0	130
7.5	215
8.0	265
8.5	290
9.0	320
9.5	330
10.0	325
10.5	325
11.0	325
11.5	330
12.0	335
12.5	330
13.0	320
13.5	285
14.0	250
14.5	220
15.0	190
15.5	165
16.0	145
16.5	130
17.0	116
17.5	106
18.0	97
18.5	92
19.0	86
19.5	82
20.0	79

References

1. D. S. Mather, et al., Report 072/72, AWRE, Aldermaston, U.K. (1972) unpublished.
2. R. Loughheed, LLNL, private communication (1983).
3. E. Arthur, LANL Report LA-9841-PR, (1983) unpublished.
4. E. Arthur, P. G. Young, D. G. Madland, and R. E. MacFarlane, LANL Report LA 9873-MS (1983) unpublished.
5. R. J. Howerton and R. White, UCID-19973 (1984) unpublished.
6. C. Budtz-Jorgensen, H. Knitter and D. L. Smith, Proc. Int. Conf. Nucl. Data Sci. Technol., Antwerp, Belgium, Sept. 1982, Ed. K. N. Böckhoff (D. Reidel Publ. Co., Boston) 206.
7. A. Gavron, H. C. Britt, E. Konecny, J. Weber and J. B. Wilhelmy, Phys. Rev. C13, 2374 (1976); and A. Gavron, private communication (1982).
8. E. Arthur, private communication (1983).
9. M. Blann and H. K. Vonach, Phys. Rev. C 28, 1475 (1983).
10. M. H. MacGregor, R. Booth and W. P. Ball, Phys. Rev. 130, 1471 (1963).
11. M. Blann and J. Bisplinghoff, LLNL Report, UCID-19614 (1982) unpublished.
12. Energy and Technology Review (Sept. 1983); M. Blann, Ann. Rev. Nucl. Sci. 25, 123 (1975).
13. M. Blann, Phys. Rev. Lett. 27, 337 (1971), 700E.
14. M. Blann, Phys. Rev. Lett. 28, 757 (1972).
15. V. F. Weisskopf, and D. H. Ewing, Phys. Rev. 57, 472 (1940).
16. N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939).
17. M. I. Svirin and S. S. Prokhorov, INDC [CCP], -208/L, IAEA translated from Nuclear Constants 4 (48) 53 (1982).
18. J. Frehaut, A. Bertin and R. Bois, Nucl. Sci. and Eng. 74, 29 (1980).
19. L. R. Veaser and E. P. Arthur, Proc. Int. Conf. Neutron Physics and Nuclear Data for Reactors, Harwell, U. K. (1978) 1054.
20. H. Karius, A. Ackermann and W. Scobel, J. Phys. G: Nucl. Phys. 5, 715 (1979).
21. Data summarized in ENDL data file.

Figure Captions

- Fig. 1 Upper: Reaction cross section (barns) versus neutron energy (MeV). Triangles are results of global spherical optical model from code ALICE; circles are results of a deformed potential optical model, reduced by calculated transitions to low-lying rotational levels (due to E. Arthur, Refs. 3, 4, 8). The closed circles with error bars are from Ref. 10. Lower: Ratio of spherical optical model reaction (total non-elastic) cross sections to deformed potential results (from upper figure) versus energy.
- Fig. 2 Calculated and experimental neutron spectra from 14.5 MeV neutrons incident on ^{93}Nb . The solid line is the result of the GDH model plus evaporation model; dashed line is hybrid + evaporation model, and dotted line is the pure evaporation yield.
- Fig. 3 Calculated and experimental neutron spectra from the $^{197}\text{Au}(p,n)$ reaction with 22.5 MeV incident protons. Closed circles are experimental results and open circles are GDH plus evaporation model predictions.
- Fig. 4 Fission probability versus neutron energy for $^{239}\text{Pu} + n$. The open circles represent evaluated cross sections from Ref. 5 divided by the reaction cross sections due to Arthur (Fig. 1). The closed circles represent three sets of first chance only fission probabilities which were used as input to our calculations.
- Fig. 5 Fission probability versus neutron energy for ^{239}Pu . The open points represent experimental fission cross sections from Ref. 6 divided by the $^{239}\text{Pu} + n$ reaction cross sections given in Fig. 1 (due to Arthur) under the assumption that $^{238}\text{Pu} + n$ cross sections are equal to the $^{239}\text{Pu} + n$ cross sections. Closed circles represent first chance only values used as input to our calculations.

- Fig. 6 Experimental and input value fission probabilities for ^{238}Pu . Open circles represent experimental results from Ref. 7; closed circles represent input to our calculation.
- Fig. 7 Experimental and input fission probabilities for ^{237}Pu . Points defined as in Fig. 6.
- Fig. 8 Experimentally deduced and calculated fission probabilities for $^{239}\text{Pu} + n$, including multiple chance fission. Open points are as in Fig. 4. Closed points represent the total fission probabilities resulting from our calculation using set (a) of first chance fission probabilities.
- Fig. 9 As in Fig. 8, using set (b) first chance fission probabilities.
- Fig. 10 As in Fig. 8, using set (c) first chance fission probabilities.
- Fig. 11 Calculated $^{239}\text{Pu}(n,2n)$ excitation functions with first chance fission probability sets (a,b,c). Set d is a recalculation based on spherical optical model reaction cross sections (Fig. 1). Experimental results of Refs. 1 and 2 are shown.
- Fig. 12 Calculated $^{239}\text{Pu}(n,3n)$ excitation function. Curves are as defined in Fig. 11.
- Fig. 13 Experimentally deduced and calculated $^{239}\text{Pu} + n$ fission probabilities based on spherical optical model reaction cross sections. The first chance only input result is shown as a dotted line.
- Fig. 14 As in Fig. 13 for $^{238}\text{Pu} + n$. Closed points represent values used as input into our calculations using spherical optical model reaction cross sections; open circles are experimentally deduced values using deformed optical potential reaction cross sections.

Fig. 15 Calculated and experimental $^{235}\text{U}(n,2n)$ excitation function. Solid line represents a calculation based on spherical o.m. reaction cross sections; dashed line represents results with reaction cross sections scaled as in the lower part of Fig. 1 to approximate results of a deformed optical potential.

Fig. 16 As in Fig. 15 for $^{238}\text{U}(n,2n)$ excitation function.

Fig. 17 Best estimate $^{239}\text{Pu}(n,2n)$ calculated excitation function, taken as an average of sets (a - c) with corrections to full use of deformed optical model reaction cross sections.

0305m/0029m

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13

Figure 14

Figure 15

Figure 16

Figure 17