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NEUTRON-INDUCED FISSION-CROSS-SECTION MEASUREMENTS AND CALCULATIONS OF SELECTED TRANSPLUTONIC ISOTOPES†

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The neutron-induced fission cross sections of ^{242m}Am and ^{245}Cm have been measured over an energy range of 10^{-4} eV to ~20 MeV in a series of experiments at three facilities during the past several years. The combined results of these measurements, in which only sub-milligram quantities of enriched isotopes were used, yield cross sections with uncertainties of approximately 5% below 10 MeV relative to the ^{235}U standard cross section used to normalize the data. We summarize the resonance analysis of the ^{242m}Am(n,f) cross section in the eV region. Hauser-Feshbach statistical calculations of the detailed fission cross sections of ^{235}U and ^{245}Cm have been carried out over the energy region from 0.1 to 5 MeV and these results are compared with our experimental data.

[^{242m}Am(n,f), ^{245}Cm(n,f), fission cross section measurements, $E_n = 10^{-3}$ eV to 20 MeV; ^{235}U(n,f), ^{245}Cm(n,f), Hauser-Feshbach statistical calculations, E_n = 100 keV to 5 MeV]

Introduction

A series of measurements of the neutron-induced fission cross sections of ^{242m}Am and ^{245}Cm have been carried out at three facilities during the past several years (see Table I). These measurements span an energy range of 10^{-4} to $2\times10^{+7}$ eV. In this report we summarize the results of the low energy ^{242m}Am resonance analysis and then outline the results and comparisons of our high energy (MeV) measurements with previously reported data. Hauser-Feshbach statistical calculations of the fission cross section over the energy region of the fission neutron spectrum are discussed and results of these calculations for the ^{235}U and ^{245}Cm fission cross sections are presented.

Table I. Summary of experimental measurements on the neutron-induced fission cross sections of ^{242m}Am and ^{245}Cm at various facilities since 1977.

YEAR	FACILITY	ISOTOPE	ENERGY RANGE (eV)
1977	LLNL-Linac†	^{242m}Am	10^{-2} - $2\times10^{+7}$
1979	LLNL-Linac	^{242m}Am	10^{-4} - $2\times10^{+7}$
		^{245}Cm††	10^{-4} - $2\times10^{+7}$
1980	LANL-WNR‡	^{242m}Am	~10^{+6} - ~10^{+7}
		^{245}Cm	~10^{+6} - ~10^{+7}
1980	LLNL-ICT‡‡	^{242m}Am	14.1 MeV
		^{245}Cm	14.1 MeV

†Livermore 100-MeV Electron Linac, Ref. 1.
††Livermore 100-MeV Electron Linac, Ref. 2.
‡Los Alamos Weapons Neutron Research Facility
‡‡Livermore Insulated Core Transformer Accelerator

Experimental Techniques

The experimental techniques of the 1977 and 1979 Linac measurements, as well as preliminary statistical analysis of the low energy resonance parameters of both ^{242m}Am and ^{245}Cm, have been reported previously.[1,2] The ^{242m}Am and ^{245}Cm fission samples used in these measurements were prepared by the LLNL Nuclear Chemistry Division and were electroplated on 0.05mm thick hemispherically shaped ionization chambers. As the samples were extremely radioactive with α-decay, the hemispherical geometry helped to differentiate between the α-decay and fission signals by limiting the path-length of alphas and fission fragments through the gas in the fission chambers. Table II gives the pertinent data on the fission samples used.

Until 1977, the only high energy data on ^{242m}Am were those of Seeger *et al.*[3] and Bowman *et al.*[4] In 1977 Browne *et al.*[1] measured the ^{242m}Am(n,f) cross section to 20 MeV. We repeated this measurement in 1979 (see Table I) with a new sample and in the same experiment measured the ^{245}Cm(n,f) cross section using two different samples (see Table II). Additional measurements of the ^{242m}Am(n,f) and ^{245}Cm(n,f) cross sections were subsequently carried out at the Los Alamos Weapons Neutron Research (WNR) facility (see Table I) to verify the Livermore Linac results in the ~1 to ~10 MeV region. Both facilities provided a 'white' source of neutrons and standard time-of-flight techniques were used to determine incident neutron energies. For these high energy measurements and for thermal energies, the cross sections were measured relative to the cross section of ^{235}U. In the resonance region, the neutron flux shape was measured with thin lithium glass detectors and the relative cross sections obtained were then normalized to the thermal data. An overlap check with the high energy data, measured independently with respect to ^{235}U, was then made in each case. Table III summarizes the errors for the ^{242m}Am(n,f) and ^{245}Cm(n,f) final data sets relative to the ENDF/B-V ^{235}U(n,f) cross section used to normalize these data.

Table II. Masses of ^{242m}Am and ^{245}Cm used in 1979 and 1980 measurements.†

ISOTOPE	SAMPLE	MASS (μg)	STATISTICAL	SYSTEMATIC
^{242m}Am	#1	207.3	± 1.0%‡	——
^{245}Cm	#1	195.3	± 0.4%	~ 2%
	#2	201.0	± 0.5%	~ 2%

Note: While these samples were isotopically enriched to >99%, there existed isotopic contamination which gave rise to spontaneous fission backgrounds which were random in time and could therefore be subtracted as constant backgrounds. *The spontaneous fission rates were* approximately as follows:
^{242m}Am #1 ~ 3.2 spontaneous fissions/sec
^{245}Cm #1 - 0.75 " " "
^{245}Cm #2 - 9.0 " " "

†The 1977 linac measurement (see Table I) used a mass of ~800μg of ^{242m}Am.
‡This number represents the total (statistical plus systematic) error on the mass assay of ^{242m}Am.

Resonance Analysis of ^{242m}Am(n,f)

Results of a Breit-Wigner sum-of-single-level

†Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48.

Table III. Summary of statistical and systematic† errors for ^{242m}Am(n,f) and ^{245}Cm(n,f) cross section measurements at the Livermore Linac.

STATISTICAL ERROR

Energy	^{242m}Am	^{245}Cm
Thermal	0.5%	0.7%
1 keV	3.5%	3.5%
1 MeV	0.5%	1.1%
14 MeV	2.0%	4.5%

SYSTEMATIC ERROR

	^{242m}Am	^{245}Cm	^{235}U
Mass	± 1%	±2 %	± 2%
Eff.‡	± 1%	$^{+1.5\%}_{-2.5\%}$	± 1%

†The known systematic errors evolve from the measured masses of ^{242m}Am, ^{245}Cm, and the ^{235}U 'standard' as well as from the efficiency of the corresponding fission chamber of each sample.
‡Estimated uncertainty in efficiency from fission pulse-height distribution and calculation of percent of fission fragments lost in sample deposit.

analysis of 48 fission resonances in the ^{242m}Am(n,f) cross section up to 20 eV are summarized in Ref. 1. These data show a complicated structure with many levels and a lack of any obvious interferences. This implies a large number of Bohr transition states (fission channels) open for this nucleus since the level-level interference is essentially nonexistent in the case of many channels. This is further supported by the fact that the distribution of fission widths for these levels follows a general chi-square distribution having at least 10 degrees of freedom. It is therefore more meaningful to fit these data with a sum of single levels than with a multilevel (R-matrix) approach which allows for interference between levels and which would require many channels per level to fit these data.

The complicated resonance structure in the ^{242m}Am(n,f) cross section in some regions between 1 and 20 eV requires a 'synthesis' of enough levels to fit the magnitude of the cross section while at the same time preserving its shape. Therefore, the average level spacing, <D>, which we determined to be 0.4 eV, is a maximum value since we have almost certainly missed narrow resonances in this region. When compared with even-even fissioning systems, the ^{243}Am nucleus should be expected to have a reduced average level spacing because the unpaired proton allows population of additional intrinsic excitations without the expenditure of energy necessary to first break a nucleon-nucleon pair. Because of the higher level density, the probability of the first resonance occurring lower in neutron energy is more likely. The first observed resonance in the ^{242m}Am(n,f) cross section[5] occurs at E_n=0.178 eV. The low energy of this first resonance together with the fact that it also has a very large reduced neutron width gives ^{242m}Am the largest thermal fission cross section known.

Data Comparison - High Energy Region

The high energy portions (10 keV to 10 MeV) of the 1977 and 1979 Linac measurements of the ^{242m}Am(n,f) cross section are plotted for comparison in Fig. 1. These were independent experiments using different samples of ^{242m}Am and the agreement in these two data sets is excellent. Also included in Fig. 1 are the recently published results of Fomushkin *et al.*[6]

The high energy portion (10 keV to 10 MeV) of the 1979 Linac measurement on the ^{245}Cm(n,f) cross section is given in Fig. 2 along with the previously measured ^{245}Cm(n,f) data of Moore and Keyworth[7] which were derived from a nuclear explosion used as a pulsed neutron source. From ~30 eV to 100 keV their data are in very good agreement with the present measurement and above 100 keV their data agree fairly well in shape with the present results.

Because of the poorer statistical quality of the 'white' neutron source data above 10 MeV, an

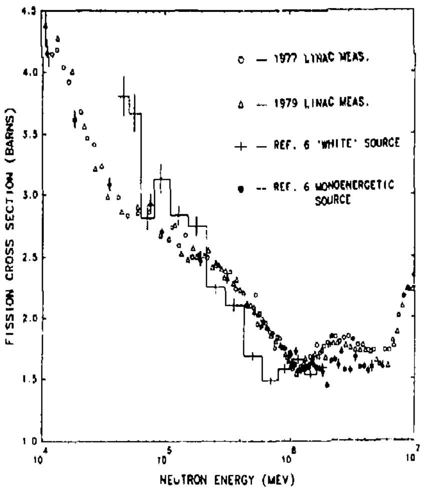

Figure 1. Comparison of the 1977 and 1979 Linac measurements of the ^{242m}Am(n,f) cross section. Also included are the recent data of Fomushkin *et al.*[6]

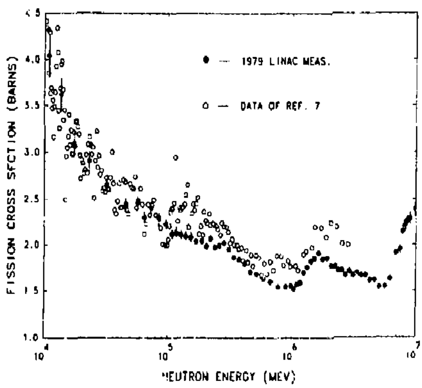

Figure 2. Comparison of our 1979 Linac measurement of the high energy ^{245}Cm(n,f) cross section with the data of Moore and Keyworth.[7]

independent monoenergetic measurement, using the same fission chambers, was conducted at the LLNL-ICT facility (see Table I). At the ICT, a pulsed 400-keV deuteron beam was used with a tritiated target to produce an intense flux of neutrons via the ^{3}H(d,n)^{4}He reaction. Collimating the neutron flux at 90° to the incident deutron beam produced a 14.1 MeV neutron beam of minimum energy spread. Time-of-flight techniques were employed to enhance the signal-to-background (spontaneous fission) ratio. The values obtained for the fission cross sections (relative to ^{235}U) at 14.1 MeV, along with the 14.8-MeV results of the ^{242m}Am(n,f) measurement of Ref. 6, are given in Table IV. As can be seen in the table, the 14-MeV cross section data for ^{242m}Am(n,f) are in good agreement. For both ^{242m}Am and ^{245}Cm these data also agree with our Linac measurements within the experimental uncertainty.

Table IV. Results and comparison of Livermore-ICT measurement of ^{242m}Am(n,f) and ^{245}Cm(n,f) cross sections at E_N=14.1 MeV and ^{242m}Am(n,f) cross section of Ref. 6 at E_N=14.8 MeV.

ISOTOPE	PRESENT WORK σ(n,f)	ERROR Stat.	ERROR Sys.	REF.6 σ(n,f)	ERROR Stat.
^{242m}Am	2.41 barns	5.1%	~5%	2.31 barns	2.2%
^{245}Cm	2.55 barns	2.8%	~5%	—	—

Calculation of the Fission Cross Section

Our present effort involves calculating these fission cross sections over the energy region of the fission neutron spectrum (~100 keV to ~5 Mev). In this energy region many of the fissile actinides have 'macroscopic' structure clearly seen in the $^{235}U(n,f)$ and $^{245}Cm(n,f)$ cross sections shown below. Bjørnholm and Lynn[8] have determined trends in fission barrier parameters over the actinide region. We have written a statistical code, FISCAL, utilizing their results, to investigate this structure as well as the general shape and magnitude of the fission cross section in this energy region.

For neutron-induced reactions, proceeding through channel x, we write the cross section as follows:

$$\sigma_{n,x}(E) = \sum_{J^\pi} \sigma_{cn}^{J^\pi}(E) \frac{T_x^{J^\pi}}{\sum_c T_c^{J^\pi}}$$

where $\sigma_{cn}^{J^\pi}(E)$ is the compound nucleus formation cross section (for a given J^π) written:

$$\sigma_{cn}^{J^\pi}(E) = \pi\lambda^2 g_J \sum_{L,S} T_{L,S}^{J^\pi}(E)$$

and J^π is the total compound nucleus angular momentum and parity, S is the channel spin (neutron+target) and L is the orbital angular momentum. For the neutron transmission coefficients we used the Moldauer form[9] suggested in Bjørnholm and Lynn. T_X is the total transmission coefficient for the decay of the compound nucleus through channel x and $\sum T_c$ is the sum of the total transmission coefficients for decay through all channels. The modes of decay important to this work are (n,n), (n,n'), (n,γ), and (n,f). Above a few hundred keV the competing decay modes are mainly the inelastic and fission channels. The total transmission coefficient for elastic/inelastic scattering is written:

$$T_n^{J^\pi}(E) = \sum_{i,L,S} T_{L,S}^{J^\pi}(E_{CM}-\varepsilon_i) + \sum_{I,L,S} \int_{E_d}^{E_{CM}} T_{L,S}^{J^\pi}(E_{CM}-\varepsilon)\rho(\varepsilon,I^\pi)\, d\varepsilon$$

where E_{CM} is center-of-mass energy, ε_i is the energy of the i^{th} discrete level in the residual nucleus, and $\rho(\varepsilon,I^\pi)$ is the level density (used above the known discrete levels) of the residual nucleus. For the gamma-decay channel we used the total transmission coefficient given in Ref. 8. For the energy region considered here, the total transmission coefficient for fission is given by:

$$T_f^{J^\pi} = \frac{T_A^{J^\pi} \cdot T_B^{J^\pi}}{T_A^{J^\pi} + T_B^{J^\pi}}$$

where A and B are the inner and outer barriers of the double-humped fission barier and

$$T_A^{J^\pi} = \sum_i \frac{1}{1+e^{-2\pi(E_x-E_A-\varepsilon_i)/\hbar\omega_A}} + \int_{E_d}^{\infty} \frac{\rho(\varepsilon,J^\pi)}{1+e^{-2\pi(E_x-E_A-\varepsilon)/\hbar\omega_A}}\, d\varepsilon$$

where the sum is over discrete barrier levels (fission channels) and the integral is the contribution from higher levels. E_A is the barrier height, E_X is the excitation of the compound nucleus and a similar expression holds for barrier B.

Calculation of the $^{235}U(n,f)$ Cross Section

To test our code and to explore the sensitivity of the calculated cross section to the various input parameters, we chose to start with the best-known fission cross section, $^{235}U(n,f)$, and have used the recent evaluation of Poenitz[10] as our standard reference data. For the inelastic channel, 93 (discrete) levels[11,12] up to 1 MeV in excitation were included for the residual ^{235}U nucleus. Not all of these levels (particularly above 500 keV) have known spin and parity but a plot of the spin distribution of the known (and tentatively known) spins yielded a spin dispersion coefficient consistent with σ=4. Therefore, levels of unknown spin were assigned spins consistent with a spin dispersion coefficient σ=4. Above 1 MeV, a level density representation of the Gilbert-Cameron constant temperature form,[13]

$$\rho(E,I^\pi) = C(2I+1)e^{-(I+\frac{1}{2})^2/2\sigma^2} e^{E/\theta}$$

was employed. Values for C, θ, and σ were taken from Ref. 8 as were the parameters for the gamma channel which employed the same form.

For the fisson channel, the barrier reference parameters, E_A=5.63 MeV, $\hbar\omega_A$=1.04 MeV, E_B=5.53 MeV, $\hbar\omega_B$=0.6 MeV, of Ref. 8 were used. The fission barrier level densities were represented by the constant temperature form given above, with several temperature regions (see. Ref. 8) employed for each barrier. However, in order to calculate the detailed cross section, it was found necessary to replace the 'shape' of the barrier level densities represented by the several constant temperature regions with a level density function which maintained that general shape and which had a continuous first derivative.[14] The resulting calculation for the $^{235}U(n,f)$ cross section is plotted in Fig. 3. No width fluctuation correction, which will reduce the cross section below ~1 MeV in the fission channel,[15] is yet incorporated in this calculation.

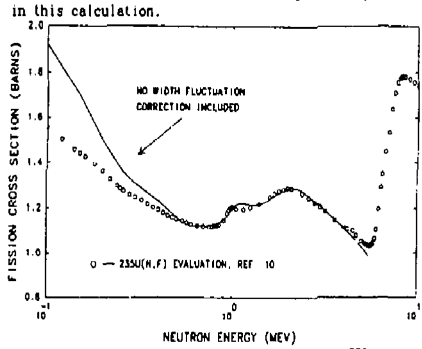

Figure 3. Hauser-Feshbach calculations of the $^{235}U(n,f)$ cross section compared to the evaluation of Poenitz.[10]

The step in the $^{235}U(n,f)$ cross section between 800 keV and 1 Mev is about 8% and, while the exact shape is not well known, the fact that this 'macroscopic' structure really exists is clearly seen in the various fission data sets on ^{235}U (see for example Ref. 10). This structure appears in the calculation because the decreasing fission cross section up to ~1 MeV is governed mainly by the increasing inelastic scattering cross section which is competing with fission. The shape of the increasing inelastic cross section with energy is governed by the increasing number of levels in the residual ^{235}U nucleus to which the neutron can inelastically scatter. Therefore, if the fission cross section is to be calculated correctly in this region, it is very important to represent the discrete levels in ^{235}U up to 1 MeV as complete as possible. From ~1.5 to ~5 MeV, the level density in the residual ^{235}U nucleus appears to be represented satisfactorily by a constant temperature form using the parameters of Bjørnholm and Lynn. However, if our representation of the discrete levels is reasonable, then there exists structure in the level density in the 1 MeV region of excitation in ^{235}U in order for our discrete representation to 'tie onto' the constant temperature form which fits the data at higher energies. This structure is seen, to a greater or lesser degree, in the level density representations of Bjørnholm and Lynn in the 1-3 MeV region of excitation above the fission barriers in ^{238}U. Without this structure in the level density our *calculation would not reproduce the detailed $^{235}U(n,f)$* cross section.

Calculation of the $^{245}Cm(n,f)$ Cross Section

For the initial calculation of the $^{245}Cm(n,f)$ cross section, we maintained the same level density parameters as in the $^{235}U(n,f)$ calculation and used the barrier parameters, E_A=5.7 MeV, $\hbar\omega_A$=1.04, E_B=4.2 MeV, $\hbar\omega_B$=0.6 MeV, of Bjørnholm and Lynn for ^{246}Cm. With these parameters, we calculated the cross section to be in qualitative agreement with the magnitude of the measured cross section (~1.7 barns) in the MeV region. However, the detailed shape was completely wrong. We should not expect the level densities used for the ^{235}U calculation to fit the detail of the $^{245}Cm(n,f)$ cross section. Since less is known about the discrete levels up to ~1 MeV in ^{245}Cm than in ^{235}U, we allowed the shape of the level density in the residual ^{245}Cm nucleus to vary while maintaining the same fission barrier level densities as used in the ^{235}U calculation. The height of the outer barrier, E_B, in ^{246}Cm was also changed to 4.5 MeV to fit the measured cross section better. The results of the final calculation are shown in Fig. 4 with no width fluctuation correction yet included in the code. As *in the case for the $^{235}U(n,f)$ calculations, we could* only fit the $^{245}Cm(n,f)$ data by allowing some structure in the level density representation of the residual ^{245}Cm nucleus at an excitation energy of ~1 to 2 MeV.

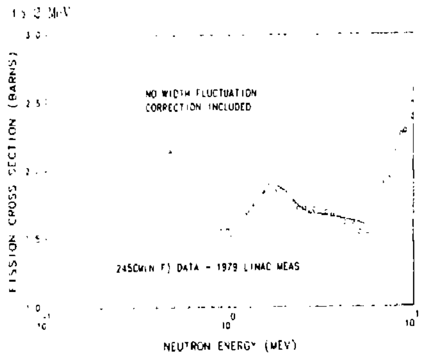

Figure 4. Hauser-Feshbach calculation of the $^{245}Cm(n,f)$ cross section compared to our 1979 Linac measurement.

Conclusions

We have completed measurements of the ^{242m}Am and ^{245}Cm fission cross sections from thermal energies to ~20 MeV in a series of experiments at several facilities. These measurements were accomplished to good accuracy with sub-milligram quantities of isotopically enriched ^{242m}Am and ^{245}Cm.

Hauser-Feshbach calculations have been carried out which reasonably represent the detailed ^{235}U and ^{245}Cm fission cross sections in the energy range from 1 to 5 MeV. Our calculations do not yet include a width fluctuation correction nor do they include a second chance fission calculation. We have been guided in this calculational effort mainly by parameterizations of Bjørnholm and Lynn. Our calculations indicate that, in order to reproduce the detailed cross section, we require level density functions which have continuous first derivatives and which contain some structure in both the residual and compound nucleus in the lower energy region of excitation.

While we conclude from these calculations that the 'macroscopic' structure seen in the 1 to 2 MeV region of these fission cross sections is most likely caused by structure in the level densities, we cannot positively rule out the possibility of this structure entering through the fission (barrier) transmission coefficients. If indeed this were the case, i.e., that the macroscopic structure were really intermediate structure, our present method of *calculating the total fission transmission would not* produce it. It seems unlikely that such intermediate structure should be present at an excitation of 2 to 3 MeV above the fission barriers.

There exist two important areas in which our calculations are being improved beyond the existing limitations discussed above. The first is in the calculation of the neutron transmission coefficients and the compound nucleus formation cross section. Neutron transmission coefficients calculated from a deformed optical potential which will fit the total cross sections over a range of actinides and over the energy region of interest here should improve the energy dependence of these coefficients and give a more physically realistic compound reaction cross section. The second area of improvement and probably the more important one is to calculate correctly the level density (intrinsic and collective) to at least a few MeV both at ground state deformation for the target nucleus and at deformations corresponding to the two barriers in the compound nucleus. It is essential that these density calculations be included if we attempt to use statistical calculations to accurately predict those cross sections for which measurements are not now feasible.

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