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Total Cross Section of ^{242}Pu Between 0.7 and 170 MeV

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Various evaluations of the neutron cross sections of ^{242}Pu lead to widely different predictions of bulk neutronics properties such as critical mass. These evaluations also show rather different behavior of the energy dependence of the total cross section. We have measured the total cross section. We have measured the total cross section of ^{242}Pu from 0.7 to 170 MeV to a statistical accuracy of $\approx$ 0.5% below 6 MeV, using 8 g of high purity material and the WNR pulsed neutron facility. Recent evaluations by Madland and Young and by Lagrange and Jary are found to be reasonably consistent with the data obtained. Best agreement, however, is found by using the simple prescription

$$\sigma_T(^{242}\mathrm{Pu}) = \sigma_T(^{238}\mathrm{U}) + [\sigma_T(^{239}\mathrm{Pu}) - \sigma_T(^{235}\mathrm{U})]$$

The remarkable accuracy of this description for ^{242}Pu suggests that it could be extended to other deformed actinides for which inadequate amounts of material exist for direct measurements of σ_T in the MeV region, as an evaluation constraint.

Introduction

The problem of nuclear data evaluation for transplutonium isotopes is difficult because the experimental data are in most cases limited or nonexistent. The evaluator relies on optical and statistical model calculations, the parameters for which are obtained by extrapolation from those of the more common fissionable materials, or as universal best fits. Frequently, such extrapolations are done subjectively and can lead to markedly different evaluated data sets. The situation that existed for ^{242}Pu prior to 1978 is perhaps typical. There were several evaluations in existence at that time: 1) Dunford and Alter[1] carried out calculations for the ENDF/B system in 1967; this was the basis of ENDF/B IV. 2) The 1973 evaluation by Caner and Yftah[2] was incorporated into the Karlsruhe KEDAK evaluation. 3) Another U.S. evaluation was that of Howerton,[3] done in 1976 for the Lawrence Livermore system ENDL. 4) A third U.S. effort was that of Mann and Schenter[4] of the Hanford Engineering Development Laboratory (HEDL) for the preliminary (1977) version of the special actinide file for ENDF/B. 5) Finally, in 1977, Lagrange[5] carried out an evaluation of ^{242}Pu at Bruyères-le-Châtel. Certain of these evaluations showed large differences in some of the partial cross sections, in particular, in the inelastic scattering and the use of a continuum distribution rather than discrete levels for the final states. Such differences in treatment have little effect when the amount of ^{242}Pu present in the system is dilute, but they can lead to pronounced effects in properties of bulk material. Calculation[6] of the critical mass for three of these evaluations gave 58, 170, and 73 kg for ENDF/B-IV, ENDL, and HEDL-77, respectively.

In 1978, two additional evaluations of ^{242}Pu became available, by Madland and Young for ENDF/B-V, also known as LASL-78 and by Lagrange and Jary.[8] These evaluations represented a substantial improvement over the earlier work, but they too did not agree in detail as shown in Fig. 1. Again, it is the optical model parameterization that appears to be responsible for the difference. Lagrange's general approach[9] to obtaining optical-model parameters may be of interest. He requires parameters to fit three sets of measurements: 1) total cross sections, 2) s- and p-wave strength functions at low energies, and 3) angular distributions of elastic plus inelastic scattering to the lowest states. For ^{242}Pu, the required data did not exist.

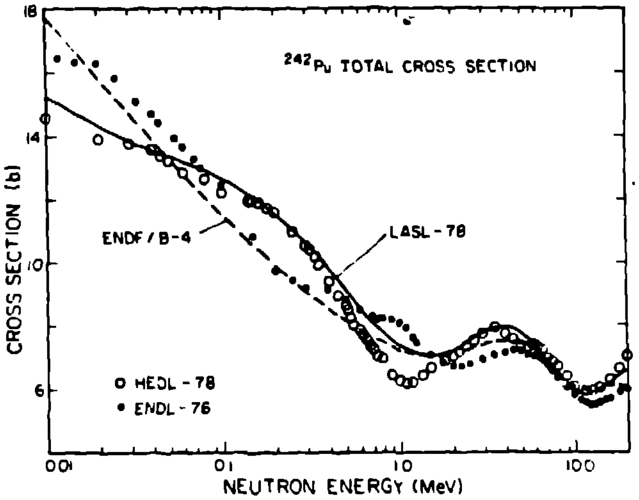

Fig. 1. Comparison of evaluated ^{242}Pu total cross sections from 10 to 20 MeV.

In order to provide an evaluation constraint for the cross sections of ^{242}Pu, we undertook to measure the total cross section at WNR, a pulsed neutron time-of-flight facility that produces spallation neutrons by using pulses of 800-MeV protons from LAMPF.

Experimental Summary

Experimental conditions for the measurement are summarized in Table I. Details of the measurement technique are described in a separate contribution to this conference[10], so that only items particular to the ^{242}Pu measurement will be covered here. The sample was a 15-mm-long, 6-mm-D cylinder of ^{242}Pu metal whose isotopic composition is given in Table II.

The sample thickness was determined by measuring the density and average diameter; the cylinder had been broken from a longer cylinder such that its length was not uniform and did not lend itself to an accurate measurement. The density was determined by weighing the sample in air and in bromobenzene, C_6H_5Br, whose density was calibrated by comparing it to that of

Table I. Experimental conditions for the ^{242}Pu total cross section measurement at WNR.

Repetition rate	1250 to 1500 pulses/sec
Flight path	31.78 m
Target	Water-cooled Ta, no moderator
Proton pulse width	0.16 ns
Neutron pulse width	~ 1.5 ns
Channel width	1 ns
Overall resolution	~ 1.8 ns or 60 ps/m
Sample temperature	~ 300 K (ambient)
Sample thickness	0.0760 x 10^{24} atoms/cm^2
Sample diameter	6.08 mm
Beam diameter at sample	5.08 mm
Detector	10 cm D x 5 cm NE-110
Beam intensity variation	12-15%
Micropulse contamination	0-4%
Data collection time	67 hours

distilled water at a known temperature. The density of the ^{242}Pu metal cylinder was determined to be 19.885 g/cm^3, which can be taken as evidence that there had been no significant nonuniformities introduced in the chill-casting preparation of the cylinder. The average diameter was determined by using micrometer calipers, after carefully filing off the casting ridges. We assign a conservative estimate of 1% as the systematic uncertainty associated with the sample thickness determination.

The sample was contained in a copper can whose end windows had a total thickness of 0.0130 x 10^{24} atoms/cm^2; the ^{242}Pu transmission measurement was carried out by using a matched blank copper can with window thickness of 0.0129 x 10^{24} atoms/cm^2. The sample, and reference samples of ^{235}U metal and natural carbon, were placed in a sample changer between two 102-mm-long collimators of depleted uranium, having a circular aperture of 5.08 mm diameter. The positioning of the sample was checked by ^{60}Co radiography prior to the measurement. Backgrounds were typically less than 1% and were measured by plugging the ^{238}U post collimator with a Ta rod that was 180 mm long. Samples were cycled at 10 to 20 minute intervals; data collection time for the ^{242}Pu sample and its blank, including backgrounds, amounted to 67 hours in three separate runs over a three-week period. Data were collected in four pulse-height windows, using detector bias settings for recoil protons ranging from 300 keV to 4 MeV. For neutron energies above 25 MeV, only the highest bias data were used; at lower energies, data in all four pulse-height windows were summed. The ^{235}U and carbon reference samples gave satisfactory agreement (within 1%) with measurements by Schwartz et al.[11] and Auchampaugh et al.,[12] respectively.

Table II. Plutonium isotopic composition of cast electrorefined ^{242}Pu metal.
(Casting 242-1, 141 g Pu)

Pu Mass No.	Atom %
236	$\leq 3 \times 10^{-9}$ (a)
238	0.00043 (a)
239	0.082 (b)
240	0.011 (b)
241	< 0.002 (b)
242	99.91 9b)

(a) Radiochemical analysis

(b) Mass spectrometer analysis

The data obtained were box-averaged in bins ranging from 5 to 99 one ns time channels; the energies listed in Table III correspond to that of the center channel in the box average. The data are plotted in Fig. 2; the solid curve drawn through the data below 20 MeV is not an eyeguide but our simple prescription of calculating the total cross section for ^{242}Pu. This was done before the data had been processed and is discussed in the next section.

Theoretical Estimate of σ_T for ^{242}Pu

It is well known that the 1-20 MeV total cross sections of all the actinide nuclei show very nearly the same features: a size resonance in the region of 4 MeV, and another about 20 MeV. The positions and widths of these resonances depend only weakly on deformation, on nucleon number, on whether the nucleons are paired, etc. The best-known total cross sections are probably those of ^{235}U, ^{238}U, and ^{239}Pu; the ENDF/B-IV evaluations for these three are based primarily on measurements by Schwartz et al. If all four of the actinides under consideration here (i.e., ^{242}Pu plus the "big three") are rigidly deformed, then an adequate description of the total cross section of ^{242}Pu should be given by using that of ^{238}U and correcting for the extra four nucleons by adding the ^{239}Pu-^{235}U total cross section difference. We assume

$$\sigma_T(^{242}\mathrm{Pu}) = \sigma_T(^{238}\mathrm{U}) + [\sigma_T(^{239}\mathrm{Pu}) - \sigma_T(^{235}\mathrm{U})] \qquad (1)$$

This description gives the solid curve plotted in Fig. 2; the data from which it was generated were taken from ENDF/B-IV.

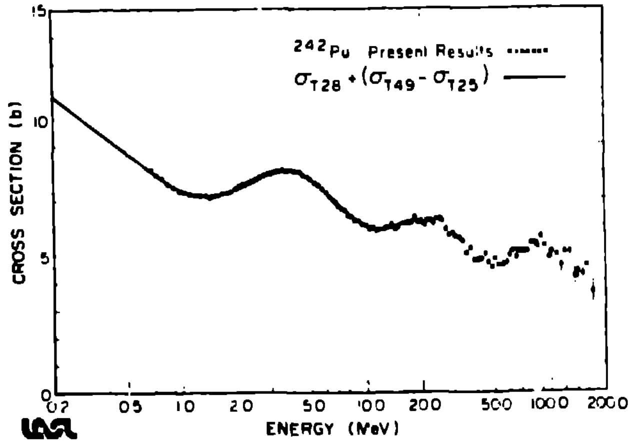

Fig. 2. Present results for the total cross section of ^{242}Pu. The solid line was obtained from equation 1 using ENDF/B-IV data.

It may be noted that if the deformation or any other variable affecting the shape and magnitude of the total cross section vary in such a way that a first-order Taylor's series expansion in A is an adequate description in this region of A and Z, then we also expect Eq. 1 to give an accurate representation. For example, let us use the variable-moment-of-inertia (VMI) model[13,14] to estimate the change in deformation for the ^{242}Pu-^{238}U difference, and for the ^{239}Pu-^{235}U difference. Using the data from the current Nuclear Data Sheets for these four isotopes,[15-18] we can calculate the parameters I_0, the moment of inertia for the nonrotating system, and C, the stiffness parameter of the VMI model, for various common bands. One finds that in going from ^{238}U to ^{242}Pu, I_0 increases by 5 ± 1%, whereas in going from ^{235}U to ^{239}Pu, I_0 increases by 7.3 ±1.6%. While the uncertainties are overlapping, the analysis based on the VMI model suggests that the deformation correction contained in the ^{239}Pu-^{235}U cross section difference may be slightly too large. This is, however, not substantiated by the data of Fig. 2. In the region of 1.5 MeV, the smooth curve of Eq. 1 is very slightly but systematically above the measured data, and from 2.5 to 3 MeV the smooth curve is slightly below the measurements. The implication is that the observed size resonance near 4 MeV is slightly wider than the predicted shape, and that the actual ^{242}Pu nucleus is perhaps slightly more deformed than Eq. 1 would suggest. The prescription given by Eq. 1 is, however, remarkably accurate for ^{242}Pu. The value of chi-square for this fit is nearly a factor of 10 lower than that for the best of the ^{242}PU evaluations.[7,8] We suggest that the prescription could usefully be extended to other actinide total cross sections, as an evaluation constraint. Using these ^{242}Pu data and the correction obtained from the ^{239}Pu-^{235}U difference gives the total cross sections for ^{246}Cm; using the ^{239}Pu evaluation as the initial values would give data for ^{243}Cm, etc. Careful measurements on a few relatively abundant heavy nuclides could provide total cross section data useful for evaluation work between 1 and 100 MeV for essentially all the transactinide isotopes of interest to various applied programs.

Acknowledgments

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Table III. Energy dependence of the total cross section of ^{242}Pu. The listed energies are those of the center channels of the bins over which a box average was performed. Cross section uncertainties listed are those due to counting statistics: a conservatively estimated 1% systematic uncertainty should be combined quadratically.

Energy (MeV)	σ_T (b)	$\delta\sigma_T$ (b)	Energy (MeV)	σ_T (b)	$\delta\sigma_T$ (b)	Energy (MeV)	σ_T (b)	$\delta\sigma_T$ (b)
0.676	8.12	0.05	5.51	7.54	0.04	29.4	5.76	0.12
0.726	7.94	0.04	5.80	7.37	0.05	30.8	5.68	0.12
0.783	7.78	0.04	6.08	7.27	0.05	32.2	5.61	0.12
0.844	7.58	0.03	6.31	7.08	0.05	33.7	5.46	0.12
0.908	7.48	0.03	6.54	7.01	0.05	35.3	5.04	0.12
0.975	7.31	0.03	6.79	6.96	0.05	37.1	5.23	0.12
1.041	7.28	0.03	7.06	6.76	0.05	39.0	4.77	0.12
1.111	7.21	0.03	7.35	6.68	0.05	40.8	4.83	0.14
1.182	7.16	0.03	7.66	6.60	0.05	42.5	4.84	0.14
1.256	7.15	0.03	7.98	6.41	0.05	44.3	5.03	0.14
1.332	7.15	0.03	8.33	6.37	0.05	46.2	4.70	0.14
1.410	7.08	0.03	8.70	6.21	0.06	46.3	4.51	0.14
1.492	7.14	0.03	9.10	6.22	0.05	50.5	4.83	0.15
1.576	7.17	0.03	9.52	6.09	0.05	52.8	4.58	0.15
1.665	7.23	0.03	9.95	6.03	0.06	55.4	4.65	0.14
1.758	7.25	0.03	10.37	5.90	0.06	58.1	4.71	0.15
1.855	7.29	0.03	20.83	5.99	0.06	60.6	4.96	0.18
1.956	7.40	0.03	11.31	5.86	0.06	62.8	5.15	0.18
2.06	7.53	0.03	11.83	5.88	0.06	65.1	4.86	0.18
2.17	7.56	0.03	12.39	5.90	0.06	67.6	5.15	0.18
2.28	7.61	0.03	12.98	6.00	0.07	70.2	5.12	0.18
2.39	7.67	0.03	13.62	6.10	0.07	73.0	5.14	0.18
2.50	7.75	0.03	14.31	5.9[illegible]	0.07	76.0	5.10	0.19
2.62	7.81	0.03	14.95	5.98	0.08	79.2	5.37	0.18
2.74	7.89	0.03	15.53	6.00	0.08	82.5	5.42	0.18
2.86	7.95	0.03	16.14	6.08	0.08	86.1	5.24	0.20
2.98	7.95	0.03	16.79	6.12	0.08	89.1	5.65	0.19
3.10	7.99	0.03	17.47	6.13	0.08	94.1	5.29	0.20
3.22	8.04	0.03	18.21	6.39	0.09	98.5	4.90	0.21
3.35	8.06	0.03	18.98	6.17	0.08	103.3	5.12	0.22
3.49	8.10	0.03	19.82	6.14	0.08	108.4	5.02	0.21
3.64	8.02	0.03	20.6	6.04	0.09	114.0	4.57	0.23
3.79	8.08	0.03	21.4	6.24	0.09	120.0	5.07	0.25
3.96	8.08	0.03	22.2	6.27	0.09	126.6	5.09	0.26
4.14	8.05	0.03	23.1	6.13	0.09	133.7	4.16	0.29
4.33	8.03	0.03	24.0	6.33	0.10	141.5	4.34	0.31
4.53	7.91	0.03	24.9	6.31	0.10	150.1	4.28	0.33
4.75	7.85	0.03	26.0	6.25	0.12	159.5	4.61	0.35
4.98	7.76	0.03	27.0	5.94	0.12	170.0	3.57	0.44
5.24	7.56	0.03	28.2	5.75	0.12			

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