PHYSICAL REVIEW VOLUME 125, NUMBER 4 FEBRUARY 15, 1962

Neutron-Induced Fission of Pu^{241}†

H. L. SMITH, R. K. SMITH, AND R. L. HENKEL
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico
(Received October 9, 1961)

The neutron-induced fission cross section for Pu^{241} was measured at neutron energies between 0.25 and 21 Mev. The fission excitation function is similar to other even-odd fissionable nuclei and shows a cross section of 1.68±0.08 barns at 3 Mev. A rough correlation is developed from fission systematics which allows predictions of 3-Mev cross sections. U^{235} fission cross section values between 10 and 21 Mev (previously unpublished) are also tabulated.

INTRODUCTION AND GENERAL METHOD

NEUTRON-INDUCED fission cross section information for Pu^{241} is of considerable value in the understanding and design of reactors having plutonium in the fuel. In addition, the fission cross section of such a high mass isotope is of theoretical interest for the understanding of the fission process, as well as of practical use as an aid in the estimation of cross sections not yet determined.

Two plutonium foils were prepared by depositing chemically-purified Pu^{241} on platinum plates. The quantities of the various nuclides on the foils were determined by several methods: by mass spectrometry and the measurement of the α-activity growth rate of the daughter Am^{241} (both checked by α-pulse analysis), by thermal neutron-induced fission counting, and by spontaneous fission counting. For the determination of the neutron-induced fission excitation function, the foils were irradiated by neutrons of known energies produced by the $T(p,n)He^3$, $D(d,n)He^3$ and $D(t,n)He^4$ reactions. At each neutron energy, the fission fragments from a Pu^{241} foil and a standard U^{235} foil (mounted back-to-back) were counted in an ionization chamber filled with a krypton-carbon dioxide mixture. The Pu^{241} cross section was then calculated using the ratio of the counting rates, the number of atoms on the foils, and the cross section of U^{235}.

EXPERIMENTAL METHOD

Foil Preparation and Analysis

The Pu^{241} was obtained from the Oak Ridge National Laboratory. Mass spectrometric analyses made at ORNL and at this laboratory immediately after chemical purification agreed very well. The data are given in Table I. In addition, Pu^{238} was present to the extent of 2.4% of the total plutonium α-activity, an amount too low for detection by mass spectrometry ($\sim 10^{-3}$ atom percent).

The foils were prepared by electrostatic spraying of plutonium nitrate in alcohol upon 2-in. polished platinum disks 0.005 in. thick, using a technique similar to that described by Carswell and Milsted.[1] With the apparatus used, the diameter of the deposit is usually about $\frac{3}{8}$ in.; however, the mount was moved mechanically during the spraying to enlarge the deposit to 1 in. The plutonium was then ignited to the oxide.

The amount of Pu^{241} on the foils was measured by several methods:

(1) Growth of Am^{241}. The samples were α-counted daily for about a month in a low-geometry zinc sulfide scintillation counter of known geometry. Analysis of the growth curves was used together with the half-lives of Am^{241} (458 years) and Pu^{241} (13.26 years) to calculate the mass of Pu^{241} on the foils. The α-pulse analysis of the foils confirmed the assumption that Am^{241} was the only α emitter growing in.

The foils were counted several months later when 3–4% of the Pu^{241} had decayed and the total α activity was about ten times the initial activity. These data gave masses consistent to within 1% with those calculated from the earlier α-counting determinations.

(2) Fission comparison with standard foils. This technique involved comparison of fission counting of a standard foil with an unknown foil in a flux of thermal neutrons. The Pu^{241} foils were counted in a fission counter in the Los Alamos Water Boiler back-to-back with standard foils of Pu^{239} and U^{235}. The ratios of fission counting rates for U^{235}/Pu^{241}, Pu^{239}/Pu^{241}, and U^{235}/Pu^{239} were measured. Using these ratios, the number of atoms on the standard foils, and thermal fission cross sections with correction factors for effective neutron temperature as compiled by Westcott,[2] the number of atoms of Pu^{241} was calculated.

(3) Spontaneous fission counting. The spontaneous fission counting rate of the Pu^{240} and Pu^{242} in the

TABLE I. Mass spectrometric analysis of Pu^{241}.

Mass	Atom percent ORNL	LASL	Average
239	1.40	1.40	1.40
240	2.28	2.30	2.29
241	96.10	96.10	96.11
242	0.20	0.20	0.20

† Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] D. J. Carswell and J. Milsted, J. Nuclear Energy 4, 51 (1957).

[2] C. H. Westcott, Chalk River Laboratory Report CRRP-787, 1958 revision (unpublished).

TABLE II. Mass of Pu^{241} on the foils.

Method	Pu^{241} (μg) Thick foil	Thin foil
Growth of Am^{241}		
(1) from initial rate	599	74.9
(2) after finite decay of Pu^{241}	601	75.0
Fission comparison	595	76.4
Spontaneous fission counting (not included in average)	670	···
Average	598±18	75.4±2.3

larger sample was low (~28 fissions/hour), necessitating long counting times. The fission rates were used in conjunction with spontaneous fission half-lives[3] and the mass spectrometric analysis to calculate the amount of Pu^{241}. The thin foil was not used because of its very low counting rate. The weight obtained for the thick foil by this method is not inconsistent with those from other methods, but in view of the low abundance of the fissioning isotopes which resulted in a statistical uncertainty larger than 10%, this number is not considered in the average.

The data are presented in Table II. The first three determinations in the table give a mean deviation of 0.23% from the average for the thick foil and 0.9% for the thin foil. A consideration of the possible errors in half-lives, cross sections, and counter efficiencies leads to the assignment of a ±3% uncertainty in the mass determinations.

Measurement of the Cross Section

Fission Counting

The ratio of the neutron-induced fission cross section of Pu^{241} to that of U^{235} was measured by placing a U^{235} foil and a Pu^{241} foil back-to-back in a 4-in. diameter spherical ionization chamber and counting the fissions from both foils while irradiating with neutrons of different energies. The foils were of 1-in. diameter and were located about 5 in. from the neutron source. A number of errors were minimized by alternately rotating the chamber 180° so that first one and then the other foil was closer to the neutron source and averaging the two sets of data.

Because of the high β-disintegration rate of Pu^{241} (2.7×10^9 dis/sec from the thick foil) there was some question as to whether a conventional ionization chamber could be used to detect the fission events. However, by filling the chamber with gas at 30 mm absolute pressure and running it as a dE/dx device as suggested by Nyer,[4] good discrimination against the β pile-up was achieved. This technique also gives better discrimination against α-particle pulses than that obtained when the ionization chamber is operated for full fragment energy loss.

The amplification system consisted of a preamplifier with a gain of approximately 70 and an amplifier with a gain of 2000. Pulses were counted by duplicate scalers operated in parallel, and pulse height distributions were taken with two 100-channel analyzers. The pulse height distributions of the fission pulses served as a check on the performance of the chamber and amplification system, and were also used to correct for the contribution due to low-energy pulses.

Neutron Production

The range of neutron energies from 0.25 to 4.5 Mev was obtained from the $T(p,n)He^3$ reaction, from 5 to 8.5 Mev the neutrons were produced by the $D(d,n)He^3$ reaction, and from 12 to 21 Mev by the $T(d,n)He^4$ reaction. Gas targets, 1 or 3 cm in length, similar to

TABLE III. Neutron-induced fission cross sections of Pu^{241} and U^{235}.[a]

Neutron energy (Mev) ± energy spread	$\sigma_f(Pu^{241})/\sigma_f(U^{235})$ (±5%)	$\sigma_f(U^{235})$ (barns) (±5%)	$\sigma_f(Pu^{241})$ (barns) (±6%)
0.12 ±0.03	1.39	1.75	2.43
0.155±0.035	1.31	1.62	2.11
0.27 ±0.03	1.40	1.41	1.98
0.40 ±0.06	1.40	1.30	1.82
0.43 ±0.10	1.26	1.28	1.61
0.528±0.094	1.40	1.23	1.72
0.75 ±0.31	1.39	1.28	1.64
1.00 ±0.23	1.39	1.26	1.75
1.50 ±0.20	1.43	1.29	1.85
2.01 ±0.15	1.37	1.32	1.82
2.45 ±0.15	1.33	1.30	1.73
3.01 ±0.13	1.29	1.24	1.62
3.01 ±0.13[b]	1.29	1.24	1.60
3.50 ±0.12	1.31	1.20	1.58
4.0 ±0.11	1.33	1.20	1.59
4.49 ±0.10	1.31	1.19	1.56
5.00 ±0.24	1.40	1.16	1.62
5.43 ±0.34	1.43	1.13	1.61
6.02 ±0.28	1.38	1.16	1.60
6.30 ±0.08	1.38	1.30	1.79
6.52 ±0.25	1.32	1.40	1.85
6.54 ±0.08	1.35	1.42	1.92
7.00 ±0.23	1.28	1.63	2.09
7.42 ±0.21	1.25	1.74	2.17
8.01 ±0.19	1.26	1.82	2.29
8.22 ±0.06	1.26	1.84	2.32
8.53 ±0.06	1.27	1.87	2.37
12.4 ±0.3	1.21	1.86	2.25
14.4 ±0.6	1.18	2.20	2.60
17.0 ±0.5	1.15	2.30	2.64
18.0 ±0.4	1.16	2.25	2.61
19.0 ±0.3	1.17	2.20	2.57
20.0 ±0.25	1.17	2.20	2.57
21.0 ±0.20	1.21	2.25	2.72

[a] See section on uncertainties to understand how one assigns the errors in this data tabulation.
[b] Thin-foil data.

[3] F. R. Barclay, W. Galbraith, K. M. Glover, G. R. Hall, and W. J. Whitehouse, Proc. Phys. Soc. (London) **A67**, 646 (1954).
[4] W. E. Nyer, Los Alamos Scientific Laboratory Reports, LAMS-938, 1950 (unpublished) and LA-994, 1949 (unpublished).

that described by Nobles[5] but with the cooling chamber removed, were filled with deuterium or tritium gas to provide neutron sources. Targets had either nickel or molybdenum windows. Various combinations of window materials, target lengths, and gas pressures were chosen to minimize experimental time while keeping the neutron energy spread as low as practicable (see Table III for tabulated energies and energy spreads).

Background Measurements

Backgrounds caused by the spontaneous fission of Pu^{240} and Pu^{242} and the fissions caused by extraneous neutrons from the machine (current not passing through the target) were found to be negligible. The gas target was evacuated and backgrounds taken at 3.5 and 18.0 Mev. These vacuum backgrounds, which measure effects due to extraneous neutrons, were very similar for the Pu^{241} and U^{235} and were less than 1% at both energies, so that the counting rate ratio was not appreciably affected. No measurement was made of the thermal neutron and scattered neutron backgrounds, but in other such measurements these backgrounds were found to be negligible even when one sample was thermally fissile and the other was not.

CALCULATIONS AND RESULTS

It can be shown that the ratio R of the fission counting rates of the plutonium and uranium foils is given by the expression

$$R=\Big(\sum_{i=240}^{242}(N_i\sigma_i)+N_{\mathrm{Am}}\sigma_{\mathrm{Am}}\Big)\Big/\sum_{j=234}^{238}(N_j\sigma_j),$$

where i indicates plutonium isotopes and j indicates uranium isotopes. N = number of atoms of a substance; σ = cross section in barns of the particular isotope; Am = americium 241.

Although less than 1% of the Pu^{241} had decayed to Am^{241} at the time of the measurements, a correction was made for the americium contribution above its fission threshold, using cross-section values proportional to the Pu^{241} cross section normalized at 3 Mev to the $Z^{4/3}/A$ relationship mentioned by Allen and Henkel.[6] The correction was made by adding an appropriate weight of Am^{241} to the Pu^{241} weight.

The Pu^{239} cross section came from Smith, Henkel, and Nobles,[7,8] and the Pu^{240} cross section up to 8 Mev came from work by the same group.[8] The Pu^{240} cross section above 8 Mev was estimated by assuming the same shape as the Pu^{241} cross section. For the Pu^{242} cross section up to 7.5 Mev, an excitation function recently measured by Simmons *et al.*[9] was normalized to the absolute determination (0.1–1.6 Mev) of Butler.[10] Above 7.5 Mev the cross section was estimated by assuming the same general shape as for Pu^{241}.

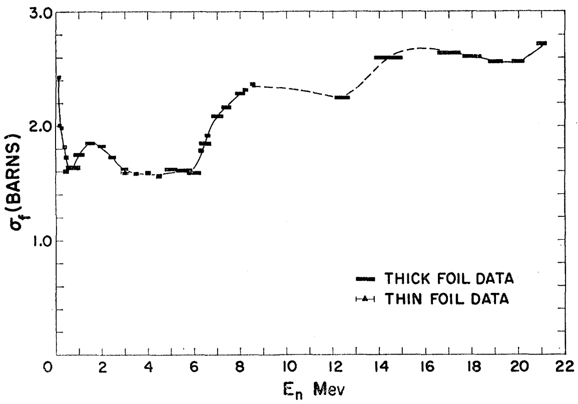

FIG. 1. Neutron-induced fission cross section of Pu^{241}.

The approximation was made that all the uranium on the standard foil was U^{235}. This introduces an error of less than 0.1%, since the uranium was 99.85% U^{235}, with less than 0.1% each of U^{234}, U^{236}, and U^{238}. A correction of approximately 1% was applied to the data between 8 and 8.5 Mev because of the low-energy neutrons from the D(d,np)D reaction.[11]

Table III presents the cross section data for Pu^{241}, taken mostly with the thick foil. A measurement at 3 Mev using the thin foil is included in the table. The thin-foil value agrees with that of the thick foil within the standard deviation of the statistical uncertainty. Figure 1 is a plot of the Pu^{141} cross section vs neutron energy. The general shape of the curve is consistent with the concept of n fission, for the region from 1 to 6 Mev and n, n' fission for the region from 0.9 to 13 Mev and n, $2n$ fission for 15 to 20 Mev. The dip at about 0.6 Mev probably indicates a resonance in the inelastic neutron cross section.

Since the absolute cross section of Pu^{241} depends directly upon the U^{235} cross section and the U^{235} numbers were obtained from various sources, all U^{235} values used are also tabulated. The sources of these numbers are the following: Between 0.1 and 10.0 Mev, cross sections[12] were read from the curves in LA-2114. This report includes more recent data[8] than those tabulated by Allen and Henkel.[6] The U^{235} cross-section values above 10 Mev were determined at the same time and using the same technique as 2–10 Mev data previously

[5] R. A. Nobles, Rev. Sci. Instr. **28**, 962 (1957).

[6] D. W. Allen and R. L. Henkel, *Progress in Nuclear Energy*, edited by R. A. Charpie *et al.* (Pergamon Press, New York, 1958), Vol. 2, Ser. 1.

[7] R. K. Smith, R. L. Henkel, and R. A. Nobles, Bull. Am. Phys. Soc. **2**, 196 (1957).

[8] R. L. Henkel, R. A. Nobles, and R. K. Smith (private communication).

[9] J. E. Simmons, R. B. Perkins, and R. L. Henkel (private communication).

[10] D. K. Butler, Phys. Rev. **117**, 1305 (1960).

[11] L. Cranberg, A. H. Armstrong, and R. L. Henkel, Phys. Rev. **104**, 1639 (1956).

[12] R. L. Henkel, Los Alamos Scientific Laboratory Report LA-2114, 1957 (unpublished). This report contains the same data on U^{235} as reference 6 with reference 8 data on U^{235} added.

reported,[7] but have only recently been released for publication.[13]

The standard error in the absolute value of the Pu^{241} cross section has been estimated to be 6% by combining a number of individual uncertainties. The amount of Pu^{241} on the foil is uncertain to 3%. The same U^{235} standard foil was used to measure both the Pu^{241} and U^{235} cross sections reported in Table III above 10 Mev, thus some errors associated with the U^{235} data and foil above 10 Mev are eliminated from this measurement. The uncertainty associated with the U^{235} foil is a basic uncertainty of about 4% of the flux measurement. Below 10 Mev a 5% uncertainty should be associated with the flux measurement. (The U^{235} standard foil was calibrated for neutron flux measurement by comparison with a polyethylene foil in a neutron telescope.[14] The statistical error varied from 1 to 1.5%.) Other possible sources of error are the corrections made necessary by the impurities in the plutonium, self-absorption, etc., which together would probably amount to about 2%. Taking an rms average of these main source of error, a value of 6% is obtained.

DISCUSSION OF FISSION SYSTEMATICS

The average of values of the Pu^{241} fission cross section between 2 and 5.5 Mev obtained from these measurements is 1.64 barns. This is about 0.2 barn above the value predicted by the σ_f vs $Z^{4/3}/A$ relationship mentioned in the literature.[6] Values at 2.5 and 14.6 Mev are 30% and 22% higher than those obtained by Kazarinova *et al.*[15] Between 0.8 and 1.7 Mev the data are 6% higher than recent work at Argonne by Butler.[16]

The best-known cross sections[6,9,17–22] of a number of nuclides (Table IV) were used to look for a simple relationship between first plateau values of fission cross sections as a function of Z and A. Using an IBM 704 computer, it was found that the plateau values for a dozen isotopes varied linearly with the function $(Z^2/A^{3/2})^n$ (Z=nuclear charge and A=number of nucleons in the compound nucleus), where n could be varied from about -2 to $+4$ and the function still fit the data within the experimental uncertainty. σ_f was found to vary as

$$\sigma_f(\text{barns}) = -39.031 + 17.231\, Z^2/A^{3/2}. \qquad (1)$$

TABLE IV. Average neutron-induced fission cross sections from 2 to 5.5 Mev.

Target isotope	σ_f average 2–5.5 Mev	Reference No.
Ra^{226}	0.001	17
Th^{232}	0.133	18
Pa^{231}	1.28	18
U^{238}	0.542	6
U^{236}	0.8455	18
U^{235}	1.206	6
U^{234}	1.54 (to 4 Mev only)	19
U^{233}	1.745	6
Np^{237}	1.572	20
Pu^{242}	1.23	9[a]
Pu^{241}	1.64	(This work)
Pu^{240}	1.586	22
Pu^{239}	1.91	6

[a] Data of reference 9 normalized to data of reference 21 at 1.5 Mev.

It is interesting that although full freedom was given to the powers of Z and A, the data determine the ratio of the power of Z to the power of A to be 1.351 ± 0.034. The case of $n=1$ is shown in the following curve (Fig. 2). The $Z^{4/3}/A$ function previously reported[6] is one of many such solutions. However, the often used fissionability parameter Z^2/A is not a solution.

The values of the fission cross sections shown in Fig. 2 were obtained by averaging the fission cross section from 2 to 5.5 Mev except for Pa^{231} and U^{234}, where values at 3.0 Mev were plotted. The Pu^{241} cross section of 1.64 barns is about 0.2 barn above the value predicted by Eq. (1). No explanation is proposed for this discrepancy.

An independent method for predicting a correlation of "fissionability" with Z and A has been used by others including Huizenga,[23] Halpern,[24] and Hyde.[25] In their approach, the important quantity which is a measure of the ability for a nucleus to fission is the energy difference between the fission barrier and the neutron binding energy $(B_f - B_n)$. Values of σ_f on the first plateaus or values of Γ_f/Γ_T plotted as a function of $(B_f - B_n)$ show a smooth correlation.[25] (The Γ symbols refer to average level widths taken over many compound nuclear levels.)

It is of interest to show that Z^n/A^m and $(B_f - B_n)$ are very similar functions. Halpern[26] has expanded the

[13] Absolute fission cross-section measurements were made with a U^{235} foil and a CH_2 foil, mounted back-to-back and irradiated in a neutron flux of variable energy using the $T(p,n)He^3$, $D(d,n)He^3$, and $T(d,n)He^3$ reactions as neutron sources. Fission fragments were detected in a parallel-plate ionization chamber and proton recoils were counted by a coincidence detector using two proportional counters and a crystal scintillator. By measuring the counting rate ratios of the foils and using the well-known cross section for (n,p) scattering, the fission cross sections were obtained. Corrections were made for the $D(d,np)D$ neutrons in the neutron energy region between 8 and 10 Mev. Neutron flux determinations had uncertainties of about 3%. The uncertainties in fission cross section values are about 5%.

[14] S. J. Bame, Jr., Eugene Haddad, J. E. Perry, Jr., and R. K. Smith, Rev. Sci. Instr. **28**, 997 (1957).

[15] M. I. Kazarinova, Tu. S. Zamiatnin, and V. M. Gorbachev, Atomnaia Energiia **8**, No. 2, 139 (1960).

[16] D. K. Butler (private communication).

[17] R. A. Nobles and R. B. Leachman, Nuclear Phys. **5**, 211 (1958).

[18] R. L. Henkel, Los Alamos Scientific Laboratory Report LA-2122, 1957 (unpublished).

[19] R. W. Lamphere, Phys. Rev. **104**, 1654 (1956).

[20] H. W. Schmitt and R. B. Murray, Phys. Rev. **116**, 1575 (1959).

[21] D. K. Butler, Phys. Rev. **117**, 1305 (1960).

[22] R. L. Henkel, R. A. Nobles, and R. K. Smith (private communication).

[23] J. R. Huizenga and R. Vandenbosch, *Nuclear Reactions* [North Holland Publishing Company, Amsterdam (to be published)], Vol. 2.

[24] I. Halpern, Ann. Rev. Nuclear Sci. **9**, 245 (1959).

[25] E. K. Hyde, University of California Radiation Laboratory Report UCRL-9065 (unpublished).

[26] I. Halpern (private communication).

values of B_f and B_n in series functions of Z and A. From measured spontaneous fission half-lives he arrives at the fission barrier $B_f=B(46-Z^2/A)$ where B is evaluated to be 0.60 ± 0.40 by using known fission thresholds. The neutron binding energy is given by $B_n=6+\gamma(Z-Z_a)$ Mev, where Z_a is the Z of the most stable nuclide for a given A. For fissionable nuclei the ratio of Z_a/A is approximately 1/3. This corresponds to the slope of the line of maximum stability over the range $A=226$ to $A=242$. Thus,

$$B_n=\gamma Z-\tfrac{1}{3}\gamma A+\text{const} \quad (\gamma=;.80\pm0.04 \text{ to fit experimental data in the fission region}),$$

$$B_f-B_n=C-BZ^2/A-\gamma Z+\tfrac{1}{3}\gamma A. \qquad (2)$$

Halpern then assumes $\sigma_f=f(Z^m/A^n)$, and shows that

$$m/n=1.40\pm0.1. \qquad (3)$$

This is in agreement with $m/n=1.351\pm0.034$ obtained empirically by the authors.

One can derive a function for σ_f by starting with Halpern's expressions

$$B_f-B_n=c-\beta Z^2/A-\gamma Z+\tfrac{1}{3}\gamma A, \quad \text{[Eq. (2) reference 26)},$$

and

$$\Gamma_f/\Gamma_n=Ne^{(B_n-B_f)/T}, \quad \text{(reference 24)} \qquad (4)$$

where T is the nuclear temperature and N is a slowly varying function which is approximately 1. Since the compound nucleus decays principally by fission and neutron emission, one has

$$\Gamma_T\cong\Gamma_f+\Gamma_n.$$

Substituting in Eq. (4),

$$\Gamma_T/\Gamma_f-1\cong N^{-1}e^{(B_f-B_n)/T}. \qquad (5)$$

First-order approximations will be used to simplify the expressions. Treating N as a constant and expanding $(B_f-B_n)/T$ in a power series, the following expression is obtained[27]:

$$\frac{\Gamma_T}{\Gamma_f}\cong C_1+C_2\frac{B_f-B_n}{T} \quad \text{or} \quad \frac{\Gamma_f}{\Gamma_T}\cong C_3+C_4\frac{B_f-B_n}{T}=\frac{\sigma_f}{\sigma_T}. \qquad (6)$$

[27] Since $(B_f-B_n)/T=[C-0.6(Z^2/A)+Z-\frac{1}{3}A]/T$ and Z and A vary about 7% in the region of radium to plutonium and T is some slowly varying function, $e^{(B_f-B_n)/T}$ can be expressed by the first two terms in a power-series treatment.

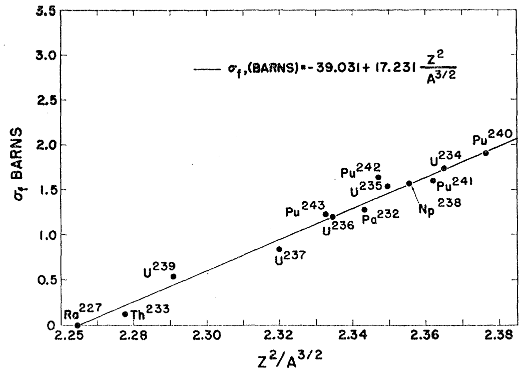

FIG. 2. First "plateau" values of fission cross sections versus $Z^2/A^{\frac{3}{2}}$.

For neutron energies between 2 and 5.5 Mev, where the fission data were averaged, σ_T has an average value of approximately 7.5 barns for U^{235} and Pu^{239}, so it is reasonable to treat σ_T as a constant. If the nucleus is excited a few Mev, T varies only a small amount and can also be treated as a constant. Thus,

$$\sigma_f=C_5+C_6(BZ^2/A+\gamma Z-\tfrac{1}{3}\gamma A) = C_5+C_7(Z^2/A+Z-\tfrac{1}{3}A). \qquad (7)$$

Using Ra^{226} and Pu^{239} data, one determines

$$\sigma_f=-20.83+0.4474(Z^2/A+Z-\tfrac{1}{3}A). \qquad (8)$$

The 12 data points fit this equation also, within experimental error.

Because Z and A vary only a few percent, it can be shown that Eq. (6) is equivalent to $\sigma_f=C_8+C_9(Z^2/A^n)$. For a particular value of σ_f, the derivatives of these two expressions can be set equal to zero and solved for n which gives $2/n=1.40\pm0.1$, which is in agreement with Eq. (2).

ACKNOWLEDGMENTS

The authors are deeply indebted to Patricia C. Stein, who prepared the foils, to Munson M. Thorpe for considerable help in the thermal fission analysis, to Lawrence F. Krenzien for the mass spectrometry of the plutonium, and to Jules S. Levin for help with the 704 analysis.