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COLLECTED REPORTS ON FISSION CROSS SECTION
OF U²³⁷ IN THERMAL NEUTRON, INTERMEDIATE
NEUTRON, AND DEGRADED FISSION NEUTRON
SPECTRA

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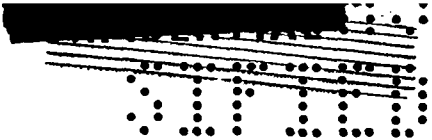
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PHYSICS AND MATHEMATICS
(M-3679, 15th edition)

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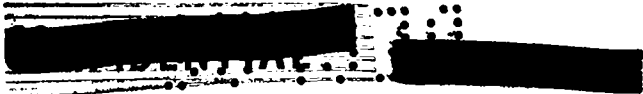
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PHYSICS AND MATHEMATICS
 (M-3679, 15th edition)

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ABSTRACT

Part 1

The neutron fission cross section of U^{237} has been measured in a thermal neutron spectrum and in a somewhat degraded fission spectrum. The fission cross section for thermal neutrons is found to be < 2 barns; the ratio of the fission cross section of U^{237} to that of U^{235} in the degraded fission spectrum is found to be $0.476 \pm 15\%$ which corresponds to $\bar{\sigma}_f$ in this spectrum equal to 0.66 ± 0.10 barns.

Part 2

The average neutron fission cross section of U^{237} has been measured in a neutron energy range extending from approximately 100 ev to fission spectrum. The average fission cross section in this spectrum is found to be 0.70 ± 0.07 barns.

Part 3

The low thermal fission cross section for U^{237} (< 2 barns) indicates that the excitation function for fission probably shows an effective threshold. If the excitation function is like all other heavy element ($Z \gtrsim 90$) neutron fission excitation functions, it will exhibit a region of approximate constancy starting at a neutron energy of 0.5 to 1 Mev above its effective threshold and extending to a neutron energy in the neighborhood of 5.5 Mev. A hypothetical excitation function for neutron

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fission of U^{237} is suggested which has a roughly constant plateau value of 0.6 to 0.8 barns. The plateau value would be reached at < 200 kev neutron energy if there is no maximum in the excitation function larger than the plateau value. The effective threshold for fission would occur at some higher energy if such a maximum exists.

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PART 1

THE THERMAL NEUTRON SPECTRUM AND TOPSY NEUTRON
SPECTRUM FISSION CROSS SECTIONS OF U²³⁷

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1.1 INTRODUCTION

Sufficient U^{237} to permit a good measurement of the thermal neutron fission cross section, if this cross section were of the order of 50 barns or greater, was obtained in late 1952 at this laboratory.

The fission cross section of U^{237} was measured at the Omega Site Water Boiler of the Los Alamos Scientific Laboratory by Cowan, Knobeloch, and Warren, members of the LASL radiochemistry group under R. W. Spence. The result was negative. Using three times the standard deviation of the fission counting rate measured over several weeks, the investigators set an upper limit to the U^{237} fission cross section of 28 barns.

Investigators at Argonne¹ reported that they had obtained in 1952 a fission cross section for U^{237} of 46 ± 8 barns. Therefore, the Los Alamos result was not fully reported until a more careful study could be made of the stability of the fission counters used in these measurements over the long periods of time necessary to complete them, and until the measurements could be repeated. This study of the behavior of the fission-counters was made in 1953 and plans were laid for more refined measurements of the fission cross section of U^{237} at thermal and higher neutron energies on larger samples of U^{237} expected to become available in 1954. Since no pile neutron spectrum peaking in the several kev region was found, members of LASL Group P-2 advanced the idea of the boron-covered fission counter described in Part 2 and assumed responsibility for the fission cross section measurement at an intermediate energy. Groups J-11 and W-2 collaborated to measure the fission cross section at

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thermal energy and in a degraded fission spectrum in the Topsy assembly at Pajarito site.

Preliminary measurements at thermal and Topsy energies were made on a preliminary sample prepared early in 1954. The thermal fission cross section was found to be less than 10 barns. The sample in the Topsy assembly became contaminated and the measurement was not completed. Because Pajarito assemblies are not coated with non-fissile protective layers and a very serious counter contamination problem exists, the original small fission counter designed for Group J-11 by John Larkins of Group H-4, which was otherwise completely satisfactory, was redesigned by Jarvis to minimize the contamination problem.

A sample of highly bombarded uranium ($\sim 160 \mu\text{g.}$) was prepared later by Group J-11 for the measurements described in detail in this report. Fission cross section measurements were made on this sample by Groups J-11, W-2, P-2, and investigators at the Argonne National Laboratory.

1.2 EXPERIMENTAL TECHNIQUE

1.2.1 Preparation of the Sample

In April, 1954, some target material was dissolved in nitric, hydrochloric, and perchloric acids, and the major fraction of the activity was recovered in about 50 ml. of 6N nitric acid solution. The initial solution measured several roentgens per hour of gamma activity at a few inches distance and was handled throughout early chemistry with long tongs

behind 1 to 2 in. of lead.

The uranium was separated from this sample by precipitation in approximately 5 mg. of $\text{La}(\text{OH})_3$ from the 6N HNO_3 solution taken to pH 10 with freshly made NH_4OH . The uranium was then purified by the Tracerlab carrier-free procedure, described in a private communication to this laboratory. The procedure consists of repeated cycles of $\text{Fe}(\text{OH})_3$ scavenging from excess Na_2CO_3 in solution, precipitation of the uranium in $\text{La}(\text{OH})_3$ from the carbonate-free solution, and extraction of $\text{UO}_2(\text{NO}_3)_2$ in hexone from an $\text{Al}(\text{NO}_3)_3$ saturated solution.

After three complete cycles of decontamination, the sample was recovered from hexone in 2 ml. of 0.1N HNO_3 , boiled down to free it of hexone, and made up to 3.00 ml. in 6N HNO_3 in a volumetric flask. This sample was designated Sample A. A 0.1 ml. aliquot was diluted to 25 ml. and designated Sample B. A 0.1 ml. aliquot of this dilution was again diluted to 25 ml. (Sample C) and an assay was made on two 1 ml. aliquots of Sample C for the U^{237} content.

Aliquots of Sample A were withdrawn and electroplated as UO_2 , ignited to U_3O_8 , on platinum foils of the proper sizes for the 2-1/2 in. inside diameter fission counters used with the Water Boiler at Omega Site, for the boron-covered fission counter used by Group P-2, and for the small fission counter used in the Topsy assembly at Pajarito Site. After electrolysis, the U^{237} left in the supernates was counted and the recovery calculated for the Water Boiler samples. This technique for calculating recovery was not successful for the Topsy sample.

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The samples taken for fission cross section measurements were designated as follows:

U ²³⁷ -A	0.500 ml. from 3.00 ml. total, used for Group P-2 measurement
U ²³⁷ -B	0.500 ml. used for thermal neutron measurement
U ²³⁷ -C	0.500 ml. used for duplicate thermal neutron measurement
U ²³⁷ -D	0.300 ml. used for Pajarito measurement in Topsy reactor
U ²³⁷ -E	0.500 ml. sent to Argonne National Laboratory

1.2.2 Thermal Fission Counting

Design for Counting Equipment

The development of comparison fission counters for use by the radiochemistry group began prior to Operation Sandstone. The design status as of 1948 is reported in the radiochemistry summary of Sandstone results.²

The equipment used to measure fissions at the Water Boiler consists in general of a double ion-chamber to each side of which are connected two parallel electronic channels, each composed of a preamplifier, amplifier, scaler and register. In this way a duplicate check of the fission counts from each half of the chamber is obtained.

The chamber itself is fabricated from a rectangular Textolite block 3-1/2 in. wide, 3-1/2 in. high, and 6 in. long (Fig. 1.1). This block is cut in half horizontally, hinged at one end and fitted with a locking device at the other (Fig. 1.2). Each half of the block has its own ion

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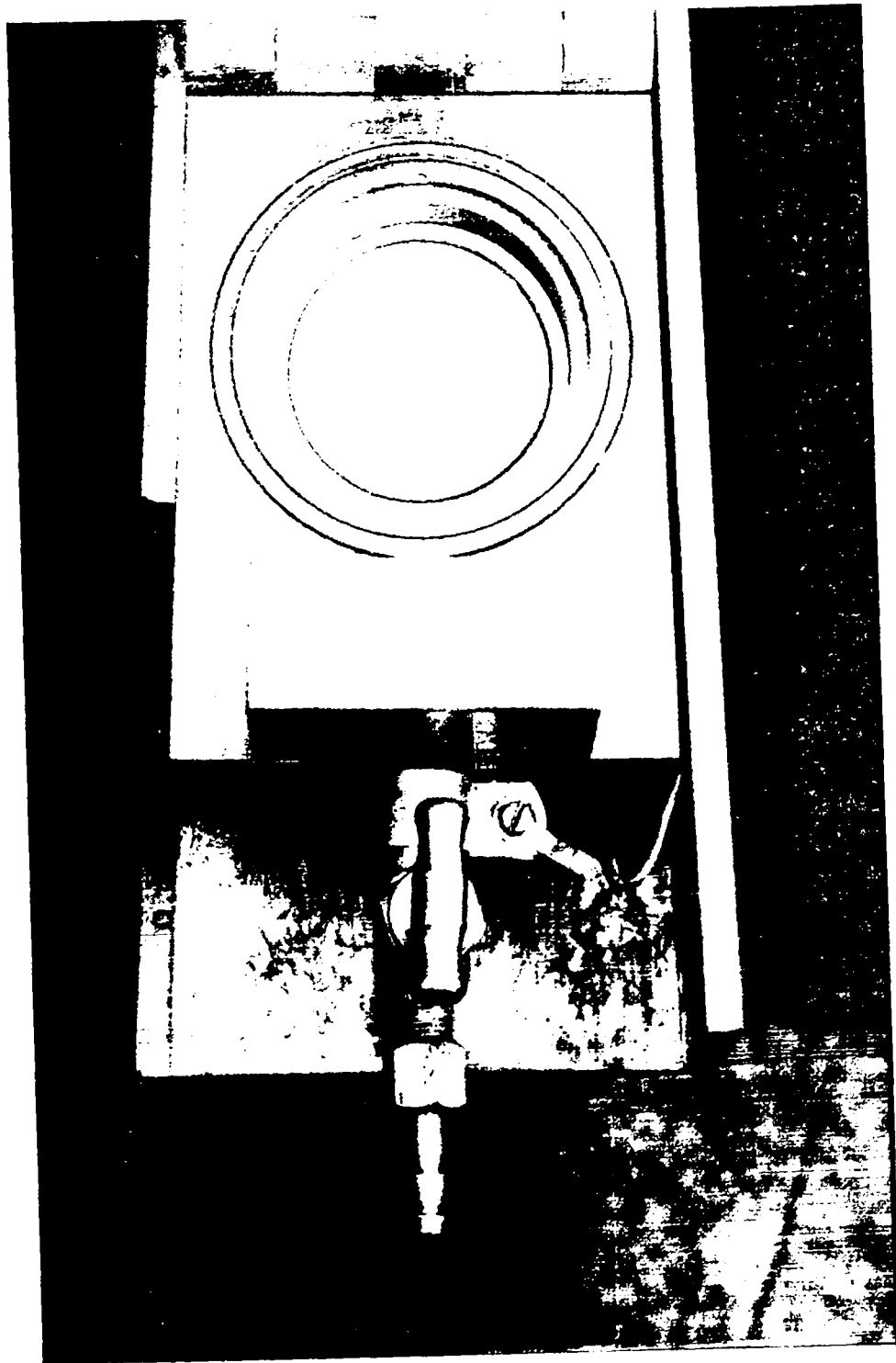


Fig. 1.1 - One chamber of thermal neutron comparison fission counter.

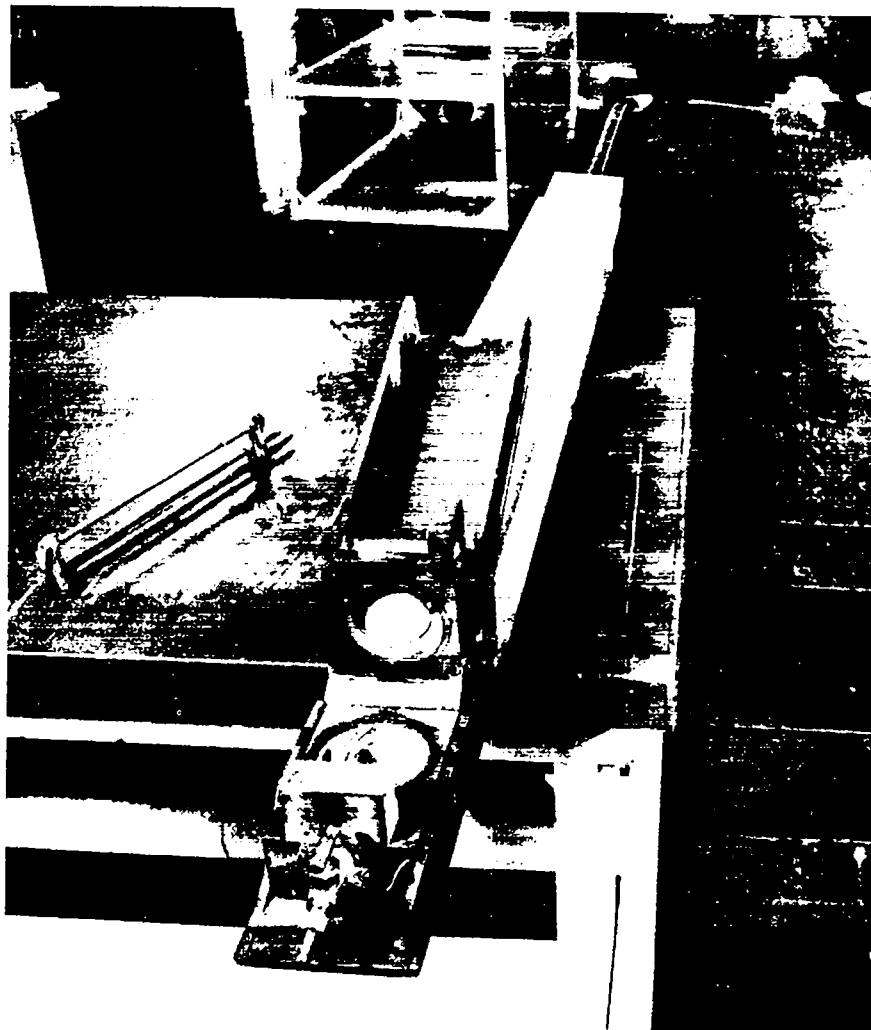


Fig. 1.2 - Thermal neutron comparison fission counter.

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chamber, which is a cylindrical hole 2-1/4 in. in diameter and 1/2 in. deep. It is cut so that when the two Textolite halves are closed, the two holes are lined up. The two samples to be comparatively counted are plated on 5 mil platinum discs 1 in. in diameter, which in turn are mounted on chrome-plated mounting plates 2-1/2 in. in diameter and placed back to back. Each sample faces half of the double ion chamber and acts as an electrode for its side of the chamber. The Textolite halves are closed and locked, and the chamber is filled with argon to a pressure of about 10 in. of mercury above atmospheric.

The chamber as described above is built into the end of a paraffin filled lucite case 4 in. square and about 4 ft. long. Coaxial cables carry the signal through the paraffin shielding to a junction box at the far end of the case. The junction box protrudes from the pile after the case has been inserted into the thermal column.

Four identical electronic circuits are used, two for each side of the chamber, to amplify and record the fission signals. Each of the circuits consists of an Atomic Instrument Company preamplifier, Model 205-B; an Atomic Instrument Company linear amplifier, Model 204-C; an Sc-3 scaler built by LASL Group CMR-7, and finally, a Veeder-Root register. All circuits are interconnected in such a way that one "counts" switch turns on all the scaling units at the same time.

Power for the entire system is supplied by an electric motor-generator, set to refine and screen out as much electronic noise as possible from the normal 110 v. house power line.

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Taking of Data

Before any data were taken for the U^{237} thermal fission cross section experiment, the system was calibrated by connecting a precision pulser with a pulse height of about 0.3 mv. to the input of all four preamplifiers. All four discriminators were set to record as nearly the same as possible.

The chamber was then inserted into port No. 1 of the south thermal column of the Water Boiler to a point just outside the cadmium curtain where the thermal neutron flux is $\sim 10^9/\text{cm}^2\text{-sec.-kw.}$ and the cadmium ratio is 200:1. The pile was operated at a power level of 25 kw.

It is known that with an over-all sensitivity of 0.3 mv. and with 300 v. applied to this type of chamber on the collecting plate, 98% or more of all the fissions that take place in the sample are counted. If it is assumed that the energy distribution of the fission fragments from both the sample and the standard is the same, then, since only the relative counting rate of the two sides of the chamber is of interest, the 2% of all fissions that are not counted have no effect on the result. There is, however, a constant small difference in the over-all efficiency of the two sides of the chamber which is taken into account in the final result.

Each $U^{235}\text{-}U^{237}$ sample from the best bombardment was counted several times per day, about 5 days per week over 5 weeks. Each count was of sufficient duration to insure at least 10^6 counts on either the sample or the U^{235} standard, whichever had the lower counting rate. This number

[REDACTED]

of counts corresponds to an expected statistical standard deviation of 0.14% in the ratio of the two sides.

The counting rate of any sample is, of course, a function of the neutron flux it sees. Both samples in a comparative counter are close enough together to insure their seeing almost identical fluxes. The flux, is, in turn, a function of the exact distance to which the chamber is inserted into the pile. The chamber was inserted to such a point that the counting rate of the stronger of the two samples was never greater than 80,000 counts per minute. The dead time of the system is 0.4 microsecond, so the coincidence correction is 0.05%. No coincidence corrections were made since the sample and standard counting rates were nearly the same and since the sample counting rate underwent negligible decay during the 5 weeks of counting.

A number of possible pitfalls in comparative fission counting of such samples as these can be present. First, the samples are subject to contamination, especially in the neighborhood of an enriched uranium reactor like the Water Boiler, and all possible precautions were taken to avoid such contamination. The practice of loading one sample and one U^{235} standard into each chamber and then locking the chamber shut for the duration of the experiment (except for opening the gas-filling cock) was adopted. Chambers must be reasonably gas-tight; at least, to such an extent that a positive argon pressure is maintained over four or five counting intervals (about 2 hours). Each chamber had to be charged with argon only once a day. If a chamber does lose its positive argon

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pressure, the discriminator plateau (plot of discriminator setting vs. counts per unit time) becomes steep over its entire span and no reliable data are obtained. Argon cylinders that contain other gas contaminants can also cause this trouble.

Before this experiment was conducted, the possibility of counting difficulties because of the presence of high beta activity directly in the chamber was investigated. It was found that 0.7 mc. of beta activity introduced on a foil had no measurable effect on counting statistics. The induced activity inside the counter appeared to be many-fold greater than the activity put on the foil. The initial activity of U^{237} on the samples finally measured was 10 mc.

The biggest difficulty encountered in comparative fission counting as done by this group is that presented by electronic noise. Should the chamber become grounded to the pile through contact of the gas-filling cock to the thermal column graphite, the pickup of noise associated with the automatic control of the reactor becomes intolerable. The operation of a.c. relays such as those used on the reactor cooler or the automatic rabbit for sample irradiations, to name two examples, cause very high spurious count rates and seriously affect the operation of our electronic equipment. The obvious corrective measure is to place metal shielding around the chamber to decrease the amount of noise picked up by the chamber. However, this solution results in large amounts of induced activities and further increases the already present hazard of beta and gamma activity to the personnel involved.

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The following precautions can be taken to keep the noise at a minimum: Shielded lines and condenser dampers on relays are used wherever possible; electric motor-generators are used to screen out any noise associated with the house power line; the contact area of the lucite chamber case with the pile graphite is kept at a minimum. It is, indeed, possible to operate at a negligible noise level and to recognize whenever noise is contributing to the counting rate. The noise source problems change from day to day and even hour to hour because different people with different electronic equipment utilize the available facilities of the Water Boiler. Fortunately, all four channels of the equipment are not affected by noise in exactly the same manner so that if a new source of noise comes into existence in the middle of a count, it can be readily detected by marked differences between the two registers recording fissions from either side of the chamber. With trouble-free conditions, any two registers hooked up to one side of the chamber will check each other to better than 0.1%. However, noise will cause a spread of greater than 0.1%. Consequently, any data exhibiting such a large spread were discarded and another count was taken.

As a test of the reproducibility of comparative fission counting data, three pairs of standard U^{235} foils were counted over periods as long as 18 months. The standard deviations calculated for these tests were 0.25%, 0.28%, and 0.37% respectively. The standard deviation expected on the basis of the number of counts recorded is $\sim 0.2\%$. The highest of the three values calculated represents the standard deviation

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in which one of the two standard foils involved exhibited a poorer discriminator plateau than any of the foils used in the U^{237} measurements. Consequently, it may be said with assurance that the increase in standard deviation of the measured ratio of two foils due to inherent variance in the counting equipment itself is not likely to be more than 0.1% over the 5 week period in which these measurements were made.

1.2.3 Topsy Spectrum Fission Counting

Design of Counting Equipment

The maximum diameter of the comparison fission counter used in Topsy measurements is limited by the diameter of the Topsy "glory hole", which is $7/8$ in. Since only microgram quantities of uranium were to be compared, the coated areas of the foils needed to be at most only a few square millimeters. The foils were in the form of $1/2$ in. squares of 3 mil platinum, with the uranium coated on a central circular patch 0.3 in. in diameter.

The most important design features of the counter had to do with precautions taken to avoid uranium contamination in the event that it had to be opened before the counting was completed. There was some concern over the possibility of contamination because of the presence of bare U^{235} assemblies in the room where the counter was to be irradiated.

Figure 1.3 shows the main assembly features of the counter. Figure 1.4 shows the counter in relation to its position in the Topsy assembly. The joint for opening the counter is well outside the Topsy

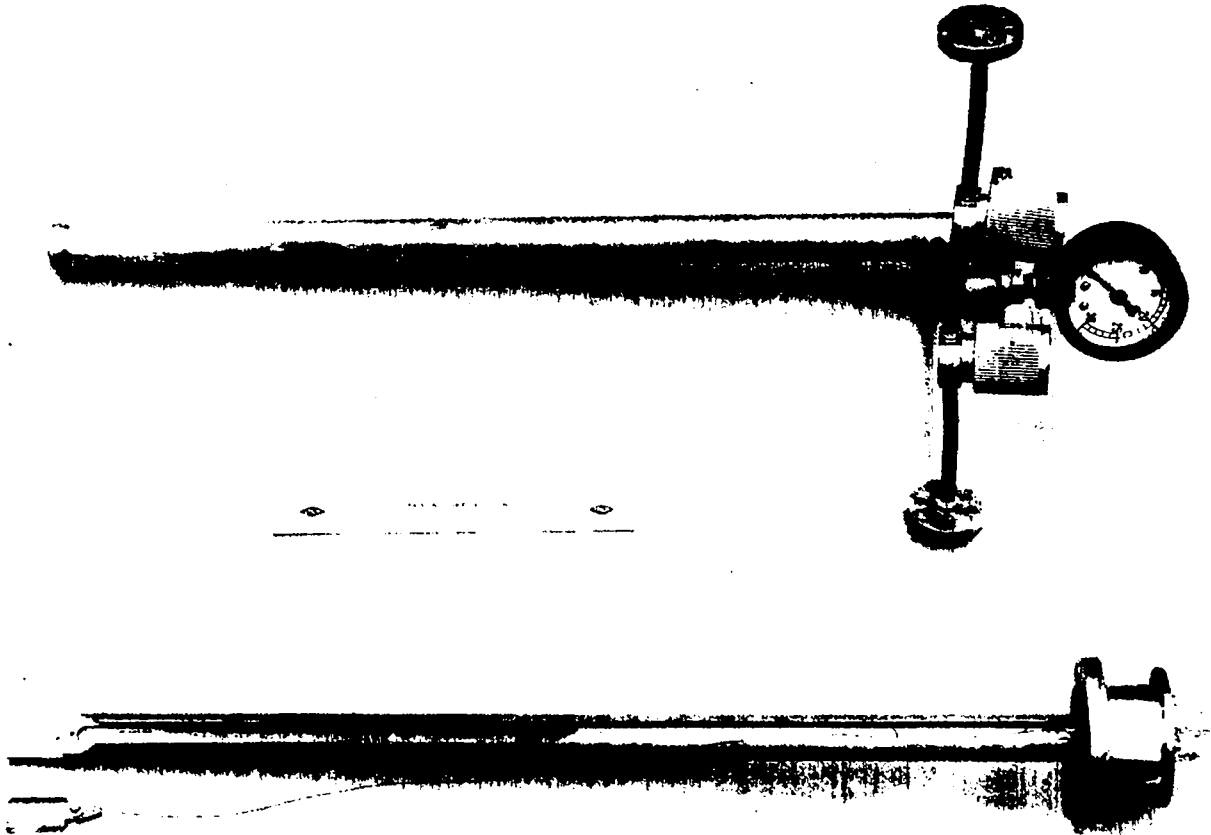


Fig. 1.3 - Topsy small fission counter.

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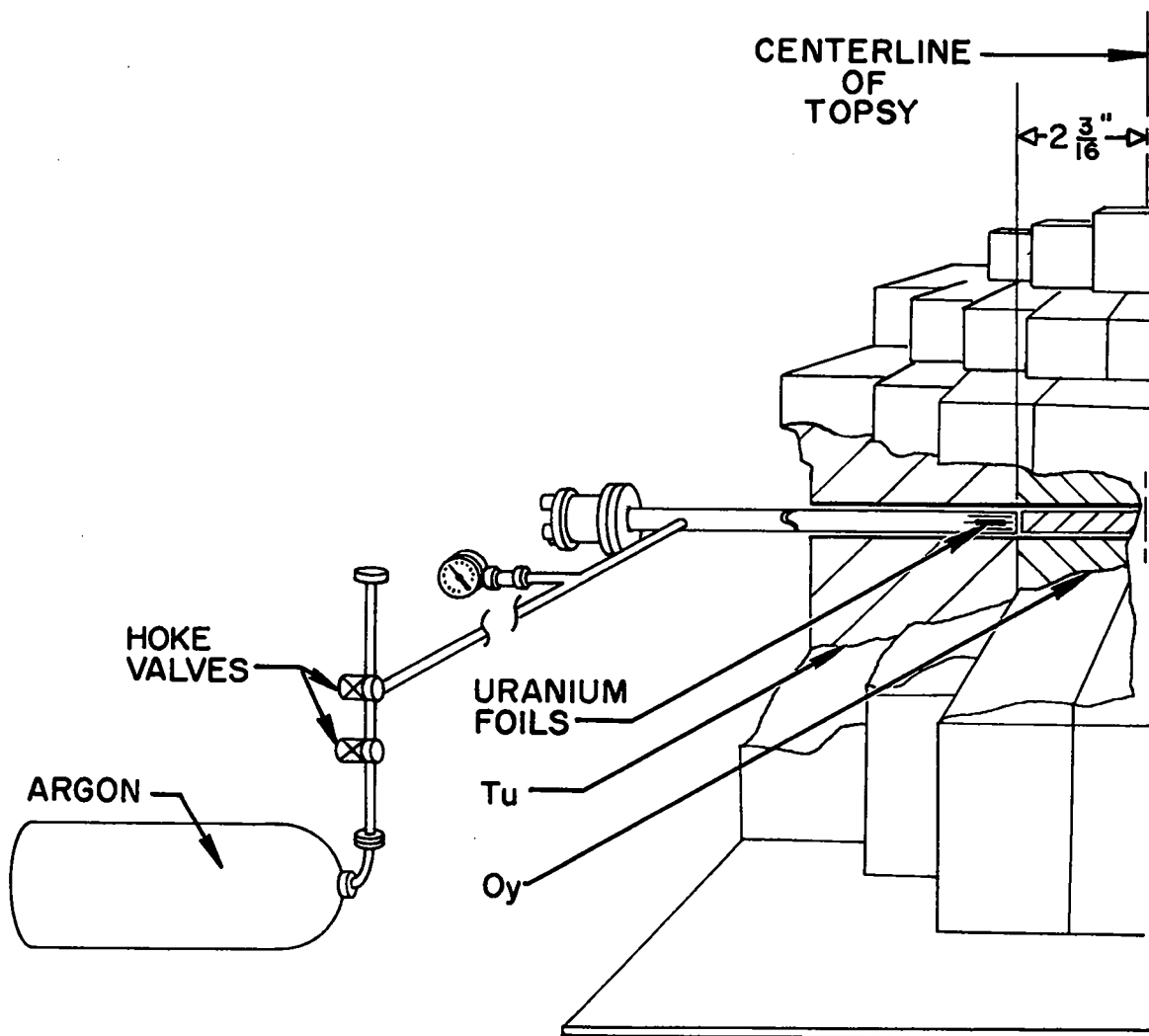


Fig. 1.4 - Counter position in Topsy assembly.

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glory hole. An argon tank with pressure regulator was attached to the counter, with valves arranged to permit flushing with argon and re-filling if operating conditions required. Noise, which developed in some of the early runs, was corrected by flushing out several times with argon. It was presumed that the noisy condition developed as a result of outgassing and insulator bombardment due, in part, to the intense beta activity of the U^{237} and, in part, to the intense reactor radiations resulting from operating Topsy at approximately 200 w. power for each of these runs. No counter noise difficulties occurred after the procedure of flushing out the counter just before each run was adopted.

Nearly 1200 ft. separated the room which housed the Topsy critical assembly from the control room where the data were recorded. Model 130 preamplifiers having output circuits designed to drive long lines were used to send the signals from the fission counters over suitably terminated RG 63-U cables to Model 101A amplifiers in the control room. The amplified signals were presented on scopes for visual monitoring and fed to separate Model 101 ten-channel pulse height analyzers for recording.

Figure 1.5 shows a typical pulse height distribution from one of the fission counters. It also indicates the interval covered by the ten-channel analyzers for the actual recording of data.

Taking of Data

The amplifier gains of the two counting systems were set

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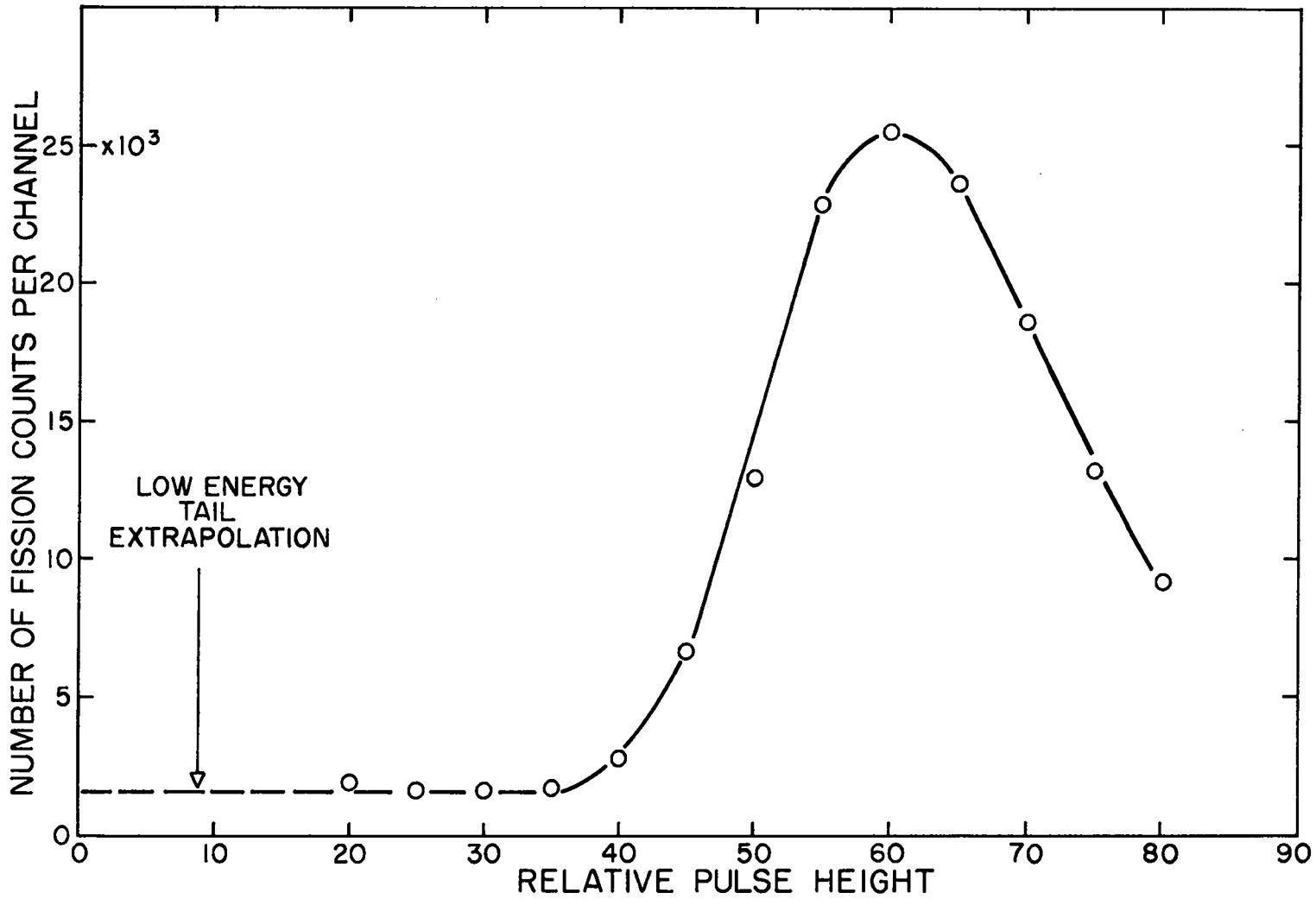


Fig. 1.5 - Typical pulse height distribution from one of the fission counters.

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approximately equal and the analyzer bias settings were adjusted to a value a little below the minimum in the pulse height distribution. The gains were such that the minimum was distributed through several of the analyzer channels. Analysis of the data in this region permitted the extrapolation of the fission pulse height distributions to zero bias for each counting system. This extrapolation was made on the assumption that the low energy area of the fission pulse height distribution was rectangular in shape. The tail correction amounted to about 4% for each counter.

The exact shape one should use for the tail correction is not known. However, since the fission foils had approximately the same weights of material coated over equal areas and were counted in similar geometries, it was assumed that the shape factor was the same for both foils and would cancel out in taking the ratio of the counts. Small differences in the tail shapes would result from abnormal thicknesses of uranium at some spots on the foils and also from different degrees of surface roughness in the two foils. Precautions are routinely taken against either of these conditions existing.

Total fission counts were obtained by extrapolating the fission pulse height distributions to zero bias. Ratios of fission counts in the two foils being compared were always taken on the basis of the observed count corrected to total counts. When this method is used, there is no need to use exactly the same gain and bias settings for the two counting systems. Small gain changes always occurred during a run, but

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no significant error was introduced because the effect of the gain was reflected in the magnitude of the tail correction. Thus, if a reduction in gain occurred, fewer counts would have been recorded on an ordinary scaler whose bias was set at a point in the minimum of the fission pulse height distribution. The number of fission pulses recorded would be in error by an amount depending on the magnitude of the gain change. With the ten-channel analyzer method of recording, a similar reduction in gain results in the recording of a higher number of pulses in the region of the minimum, which increases the tail correction enough to compensate for the gain change.

The above method of data recording and analysis, which is different from the method used with the Water Boiler, was adopted because the high power level required of Topsy to get a reasonable counting rate resulted in a temperature rise of about 15°C per hour. Over half the internal volume of the counter was out in cooler air, so that differential expansion of the counter gas occurred, resulting in a gradual decrease of gas density in the neighborhood of the foils. Because of the small size of the counter, the range of the fission fragments was large compared to the foil to collector plate separation, so that a decrease in gas density resulted in a corresponding decrease in energy deposited in the chamber by fission fragments. This condition appeared as a gradual reduction in the average signal height during the counting period.

The background correction was determined but was not applied since it was found to correspond to only 3.3×10^{-9} g. of U^{235} . The total

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fission rate of the sample corresponded to 8×10^{-7} g. of U^{235} .

1.3 EXPERIMENTAL RESULTS

1.3.1 Isotopic Content of Sample

U^{237} Content

The two 1 ml. aliquots from Sample C were mounted on platinum according to a standard procedure and counted on a beta proportional counter. The over-all efficiency of the beta counter, which is the factor that converts U^{237} counts to disintegrations, has been determined by Warren, Balagna, et al., of Group J-11 by 4π counting and by Louise Smith of Group J-11 by Np^{237} alpha counting. The reciprocal of the over-all efficiency times the decay constant, called the K factor for U^{237} , was used to calculate the atoms of U^{237} in the total Sample A. The assay indicated the total U^{237} content at time t_0 was 3.50×10^{15} atoms on the basis of $K = 5.1 \times 10^4$. The U^{237} decayed to Np^{237} with a half-life of 6.75 days and was counted over a period of several half-lives to confirm the fact that it was not noticeably contaminated with any beta-emitting species other than 14-hour U^{240} . A sample calculation follows:

$$\frac{(U^{237} \text{ activity at } t_0)(K)}{(\text{chemical recovery})(\text{fraction in aliquot})} = \text{total atoms}$$

$$\frac{(3.27 \times 10^5)(5.1 \times 10^4)}{(0.895)(5.33 \times 10^{-6})} = 3.50 \times 10^{15} \text{ atoms of } U^{237}$$

The aliquot taken for each of the three samples U^{237} -A, U^{237} -B, and

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U^{237} -C corresponded to 0.1667 of the total or to 5.83×10^{14} atoms taken for electrolysis per sample. The yield from electrolysis was evaluated by counting the supernate after electrolysis for unplated uranium. The yields of the various samples were:

U^{237} -A	99.99%
U^{237} -B	99.83%
U^{237} -C	99.30%

Thus, the number of U^{237} atoms on each plate at time t_0 was:

U^{237} -A	5.83×10^{14} atoms of U^{237}
U^{237} -B	5.82×10^{14} atoms of U^{237}
U^{237} -C	5.79×10^{14} atoms of U^{237}
U^{237} -D	1.88×10^{14} atoms of U^{237}

The yield of U^{237} -D was evaluated differently by a method described under U^{235} content.

U^{235} Content

Since it will be demonstrated that the thermal neutron fission counting rate of the samples was not measurably affected by any isotope other than U^{235} , the U^{235} content of the samples can be measured simply by the usual method of comparison fission counting. The U^{235} content of all samples was obtained in this way. The number of atoms of U^{235} on each plate was:

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U ²³⁷ -A	3.92 x 10 ¹⁴ atoms of U ²³⁵
U ²³⁷ -B	3.91 x 10 ¹⁴ atoms of U ²³⁵
U ²³⁷ -C	3.90 x 10 ¹⁴ atoms of U ²³⁵
U ²³⁷ -D	1.26 ₅ x 10 ¹⁴ atoms of U ²³⁵

The ratio of atoms of U²³⁷ to atoms of U²³⁵ at time t₀ was:

U ²³⁷ -A	1.487
U ²³⁷ -B	1.488
U ²³⁷ -C	1.485

Average ratio: 1.487

Thus, the number of atoms of U²³⁷ in sample U²³⁷-D, which was not otherwise known due to large losses during electroplating of this sample, was 1.88 x 10¹⁴ as reported above.

Samples of the same solution sent to Argonne National Laboratory were spectrochemically analyzed. The ratio of U²³⁷ to U²³⁵ at time t₀ calculated from the Argonne report³ was 1.754 which was 18% higher than the ratio calculated above.

A similar disagreement exists between the ratio of U²³⁷ to total U as calculated from the analytical results obtained at LASL and the result calculated from mass spectroscopic analysis. The Los Alamos result for U²³⁷ to total U was 0.94. The Argonne value was 1.09, or 16% higher.

The discrepancy appears to be centered on the value obtained for the number of atoms of U²³⁷ since the two laboratories seem to agree fairly

well on the ratio of U^{235} to U^{238} . Work is continuing on resolving this discrepancy. If the error is located, corrections to the values for atoms of U^{237} and calculated cross sections will be issued. As of this writing, the authors believe that the K factor of 5.1×10^{14} is the most reasonable value to use.

U^{236} Content

The U^{236} to U^{235} ratio, measured mass spectrometrically at Argonne National Laboratory, was 0.623. The number of atoms of U^{236} on each plate was:

U^{237} -A	2.44×10^{14} atoms of U^{236}
U^{237} -B	2.44×10^{14} atoms of U^{236}
U^{237} -C	2.43×10^{14} atoms of U^{236}
U^{237} -D	0.788×10^{14} atoms of U^{236}

U^{238} Content

The most precise measurement of the U^{238} content of these samples was obtained by multiplying the U^{235} content, measured by thermal neutron fission counting, with the U^{238} to U^{235} ratio, measured by mass spectroscopy at the Argonne National Laboratory. The U^{238} to U^{235} ratio was 160.5. The number was probably known to the nearest 1%.

Thus, the number of U^{238} atoms in each sample was:

U ²³⁷ -A	6.29 x 10 ¹⁶ atoms of U ²³⁸
U ²³⁷ -B	6.28 x 10 ¹⁶ atoms of U ²³⁸
U ²³⁷ -C	6.26 x 10 ¹⁶ atoms of U ²³⁸
U ²³⁷ -D	2.03 x 10 ¹⁶ atoms of U ²³⁸

Np²³⁹-Pu²³⁹ Content

Analyses by Louise Smith of Group J-11 for Np²³⁹ in 1 ml. aliquots from Sample C indicated that the Np²³⁹ contamination in the sample amounted to less than 0.01 atom per cent of U²³⁷ on April 30, 1954. This result means that the possible ingrowth of Pu²³⁹ from contaminating Np²³⁹ would cancel out \ll 0.1 barn of U²³⁷ cross section.

Pu²⁴⁰ Content

The early counts on the uranium samples showed enough U²⁴⁰ left at separation time so that, after all the U²⁴⁰-Np²⁴⁰ had decayed, the Pu²⁴⁰ to U²³⁵ ratio was \sim 0.045. Since Pu²⁴⁰ does not have a measurable thermal fission cross section, it did not contribute to the thermal fission counting rate.

1.3.2 Measurement of the Thermal Neutron Fission Counting Rate as a Function of Time

The data obtained on samples U²³⁷-B and U²³⁷-C in two comparison fission counters are plotted in Figure 1.6 as the ratio, R, of the fission counting rate of unknown to that of standard as a function of

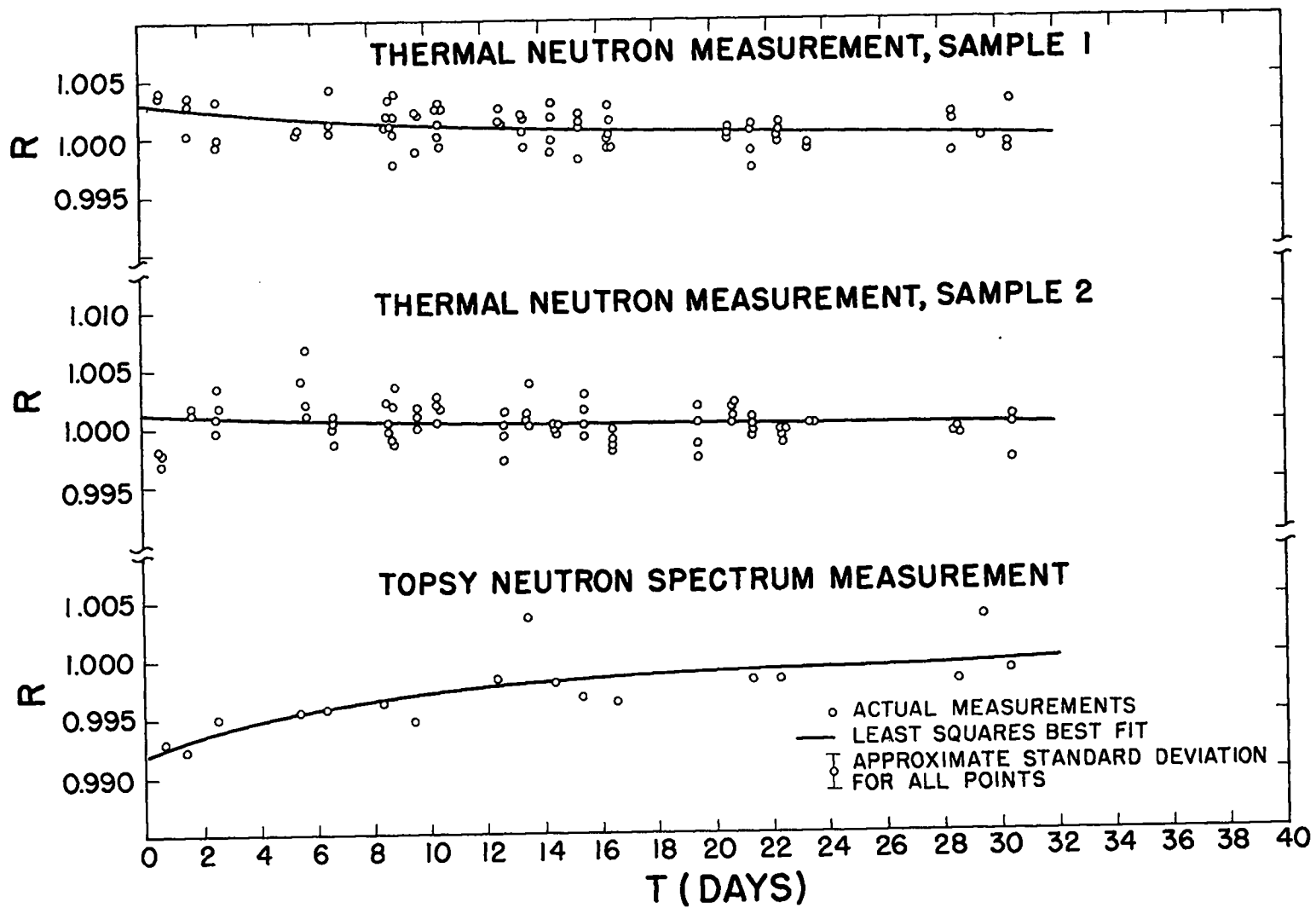


Fig. 1.6 - Measurement of thermal and Topsy neutron spectrum fission counting rates as a function of time.

time. The actual counting rate ratio between unknown and standard is in each case normalized to 1.000 at t equal to infinity. The line drawn through the points is a best fit as determined by the method of least squares.

1.3.3 Measurement of the Topsy Neutron Spectrum Fission Rate as a Function of Time

The data obtained from sample U²³⁷-D with Topsy are plotted in Figure 1.6. The ratio between unknown and standard is normalized to 1.000 at infinite time. The line drawn through the points is the best fit as determined by the method of least squares.

1.3.4 Measurement of the Topsy Neutron Spectrum

The fission counter was located in the Topsy assembly as shown in Figure 1.4. The neutron spectrum in Topsy at this point was characterized in the course of these measurements by evaluating the ratios

$\sigma_{f,Np^{237}}$ to $\sigma_{f,U^{235}}$ and $\sigma_{f,U^{238}}$ to $\sigma_{f,U^{235}}$. It has also been calculated by LASL Groups W-2 and T-1 in six flux groups from extensive detector data.⁴ The Carlson description is plotted as a smoothed spectrum in Fig. 1.7.

The six-group description at 5.27 cm. and 6.02 cm. from the center of Topsy has been interpolated to 5.56 cm., the actual position of the fission counter samples. The interpolated values are listed in Table 1.1.

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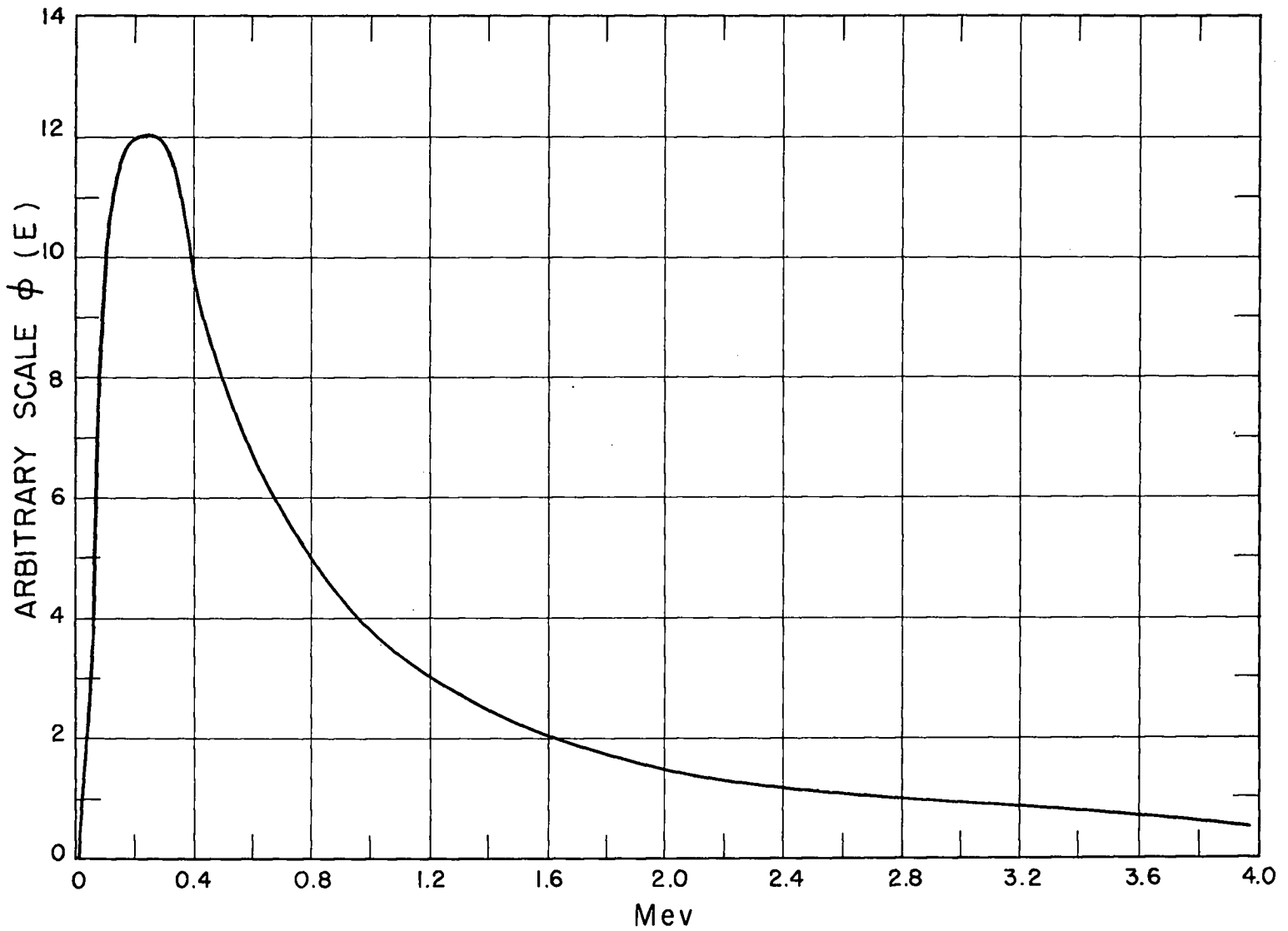


Fig. 1.7 - Carlson description of Topsy spectrum plotted as a smoothed spectrum.

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TABLE 1.1

CARLSON DESCRIPTION OF NEUTRON SPECTRUM INTERPOLATED TO
ACTUAL POSITION OF FISSION COUNTER IN TOPSY

Group	Energy (Mev)	Neutron flux fraction (6 groups)	Neutron flux fraction (3 groups)
1	0-0.1	0.0346	0.3203
2	0.1-0.4	0.2857	
3	0.4-0.9	0.2609	0.3949
4	0.9-1.4	0.1340	
5	1.4-3.0	0.1888	0.2848
6	3.0- ∞	0.0960	

The ratios of fission cross sections measured in the comparison
fission counter at 5.56 cm. were:

$$\frac{\bar{\sigma}_{f, \text{Np}^{237}}}{\bar{\sigma}_{f, \text{U}^{235}}} = 0.585$$

$$\frac{\bar{\sigma}_{f, \text{U}^{238}}}{\bar{\sigma}_{f, \text{U}^{235}}} = 0.1001$$

From these ratios, a three-group description of the flux that checks
fairly well with Carlson's description has been calculated. The check
is not expected to be exact since the presence of the fission counter
perturbs the spectrum significantly. It is necessary to assume

average U^{235} , Np^{237} , and U^{238} fission cross sections in each of the three groups. The value for the average fission cross section of U^{235} in the 0.0 to 0.4 Mev group is sensitive to the assumption made on the number of neutrons in this group below 0.1 Mev. The Carlson ratio for the fraction of flux below 0.1 Mev to the fraction of flux between 0.1 and 0.4 Mev is used in calculating an average fission cross section of 1.66 barns in the 0.0 to 0.4 Mev region. The calculated neutron flux distribution is presented in Table 1.2. The bottom horizontal column presents the calculated average cross section for each of these three isotopes at the 5.56 cm. position in Topsy when the fission counter is in place.

TABLE 1.2

CALCULATED NEUTRON FLUX DISTRIBUTION FROM FISSION MEASUREMENTS

Group	Energy, Mev	Assumed av. fission σ			Calculated neutron flux fraction
		for U^{235} , barns	for Np^{237} , barns	for U^{238} , barns	
1	0.0-0.4	1.66	0	0	0.33
2	0.4-1.4	1.21	0.982	0.013	0.42
3	1.4- ∞	1.29	1.53	0.523	0.25
Av. $\bar{\sigma}_f$		1.38	0.795	0.136	

1.4 ANALYSIS OF DATA

1.4.1 Thermal Neutron Measurement

The ratio of counting rate of unknown to counting rate of standard as a function of time in a thermal neutron spectrum is expressed by the equation

$$R(t) = \left(\frac{N_{O,U^{237}} \sigma_{f,U^{237}} e^{-\lambda t} + N_{U^{235}} \sigma_{f,U^{235}}}{N'_{U^{235}} \sigma_{f,U^{235}}} \right) \left(\frac{\epsilon_1}{\epsilon_2} \right)$$

$$R(t) = K_1 e^{-\lambda t} + K_2$$

ϵ = counting efficiency of chamber; $\epsilon_1 / \epsilon_2 = 1.007$ for both sets of counters

λ = decay constant of U^{237}

N_O = atoms of U^{237} at some time, t_0

$N'_{U^{235}}$ = atoms of U^{235} in standard

The assumption is made that there are no other thermally fissionable species in the sample.

A best fit to the data has been calculated by the method of least squares.

The values calculated for the constants are:

$$\text{Sample } U^{237}\text{-B} \quad K_1 = 1.305 \times 10^{-3}$$

$$K_2 = 1.1375$$

$$\sigma = 0.20\%$$

$$\text{Sample U}^{237}\text{-C} \quad K_1 = 4.893 \times 10^{-3}$$

$$K_2 = 1.7088$$

$$\sigma = 0.11\%$$

For purposes of the least squares analysis of the data, an arbitrary zero time was taken at 2400 on April 27, 1954. The decay constant used for U^{237} was 4.278×10^{-3} per hour. At the time t_0 , the ratios of $N_{\text{O},\text{U}^{237}}$ to $N'_{\text{U}^{235}}$ were calculated to be 1.296 for sample $\text{U}^{237}\text{-B}$ and 1.942 for sample $\text{U}^{237}\text{-C}$. Thus, the values calculated for the fission cross section of U^{237} are:

Sample $\text{U}^{237}\text{-B}$

$$\frac{\sigma_{\text{f},\text{U}^{237}}}{\sigma_{\text{f},\text{U}^{235}}} = 1.305 \times 10^{-3} / (1.296)(1.007)$$

$$= 1.0 \times 10^{-3} \pm 2.3 \times 10^{-3}$$

$$\sigma_{\text{f},\text{U}^{237}} = 0.6 \pm 1.3 \text{ barns}$$

Sample $\text{U}^{237}\text{-C}$

$$\frac{\sigma_{\text{f},\text{U}^{237}}}{\sigma_{\text{f},\text{U}^{235}}} = 4.893 \times 10^{-3} / (1.942)(1.007)$$

$$= 2.5 \times 10^{-3} \pm 1.0 \times 10^{-3}$$

$$\sigma_{\text{f},\text{U}^{237}} = 1.4 \pm 0.6 \text{ barns}$$

We can conclude with some assurance that the thermal fission cross section of U^{237} is < 2 barns.

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1.4.2 Topsy Spectrum Measurement

The ratio of counting rate of unknown to that of standard as a function of time is given by the equation:

$$R(t) = \frac{\epsilon_1}{\epsilon_2} \frac{\left[\begin{array}{l} N_{O,U237} \bar{\sigma}_{f,U237} e^{-\lambda t} + N_{O,U237} \bar{\sigma}_{f,Np237}(1 - e^{-\lambda t}) \\ + N_{U235} \bar{\sigma}_{f,U235} + N_{U236} \bar{\sigma}_{f,U236} \\ + N_{U238} \bar{\sigma}_{f,U238} + N_{240} \bar{\sigma}_{f,240} \end{array} \right]}{N'_{U235} \bar{\sigma}_{f,U235}}$$

It will be remembered that the mass 240 nuclide was present at the beginning of this measurement partly as U²⁴⁰ and partly as Pu²⁴⁰. No distinction as to element is made in the above equation since the contribution of the nuclide with this mass number to the total fission rate is almost negligible. The equation can be reduced to a form which can be readily fitted by the method of least squares.

$$R(t) = \frac{\epsilon_1}{\epsilon_2} \left[\frac{N_{O,U237}}{N'_{U235}} \left(\frac{\bar{\sigma}_{f,U237}}{\bar{\sigma}_{f,U235}} - \frac{\bar{\sigma}_{f,Np237}}{\bar{\sigma}_{f,U235}} \right) e^{-\lambda t} \right]$$

$$+ \left[\frac{\begin{array}{l} N_{O,U237} \bar{\sigma}_{f,Np237} + N_{U235} \bar{\sigma}_{f,U235} \\ + N_{U236} \bar{\sigma}_{f,U236} + N_{U238} \bar{\sigma}_{f,U238} \\ + N_{240} \bar{\sigma}_{f,240} \end{array}}{N'_{U235} \bar{\sigma}_{f,U235}} \right]$$

$$R(t) = K_1 e^{-\lambda t} + K_2$$

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The values calculated for the constants are:

$$K_1 = -5.91 \times 10^{-3}$$

$$K_2 = 0.8071$$

$$\frac{\epsilon_1}{\epsilon_2} = \text{ratio of counting efficiencies of two sides of fission counter} = 0.9893$$

$$N'_{U^{235}} = \text{atoms of } U^{235} \text{ in standard} = 2.50 \times 10^{15}$$

$$N_{O,U^{237}} = \text{atoms of } U^{237} \text{ in unknown at 4:00 P.M., MST, April 28, 1954} \\ = 1.37 \times 10^{14}$$

Thus,

$$\frac{N_{O,U^{237}}}{N'_{U^{235}}} = 1.37 \times 10^{14} / 2.50 \times 10^{15} = 5.48 \times 10^{-2}$$

From experimental measurement,

$$\frac{\bar{\sigma}_{f,Np^{237}}}{\bar{\sigma}_{f,U^{235}}} = 0.585$$

Therefore

$$\frac{\bar{\sigma}_{f,U^{237}}}{\bar{\sigma}_{f,U^{235}}} = 0.476$$

From the three-group description of the neutron flux given earlier

$$\bar{\sigma}_{f,U^{235}} = 1.38 \text{ barns and, therefore, } \bar{\sigma}_{f,U^{237}} = 0.66 \text{ barn.}$$

The standard deviation of the experimental points about this least squares fit suggests a probable error of $\pm 10\%$. Two other main sources of error are:

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1. The ratio of $N_{O,U^{237}}$ to $N'_{U^{235}}$ may be in error by as much as 10%. This much error in the ratio will introduce about 3% error in the final result.

2. The ratio $\bar{\sigma}_{f,Np^{237}}$ to $\bar{\sigma}_{f,U^{235}}$ may be in error by as much as 5%, mostly due to the uncertainty in the Np^{237} half-life. A 5% error here will make a 6% error in the final result.

When these additional sources of error are taken into account, a probable error of $\pm 15\%$ is estimated for the ratio $\frac{\bar{\sigma}_{f,U^{237}}}{\bar{\sigma}_{f,U^{235}}} = 0.476$.

References

1. G. L. Pyle, Sherman Fried, W. C. Bentley, and J. L. Lerner, Argonne National Laboratory Report ANL-WMM-1113, May 1, 1953.
2. R. W. Spence and M. G. Bowman, "Radiochemical Efficiency Results of Sandstone", Sandstone Report, Vol. 10, Annex 1, March 1949.
3. Sherman Fried, Argonne National Laboratory Report ANL-WMM-1215, June 30, 1954.
4. Memorandum, Bengt Carlson, LASL, to Gordon Hansen, LASL, Ref. sym. T-1-62, April 5, 1954.

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PART 2

AVERAGE FISSION CROSS SECTION OF U^{237} FOR
INTERMEDIATE ENERGY NEUTRONS

(This report is essentially a verbatim transcript of LAMS-1700, corrected to include best values for the isotopic content of the uranium samples measured. The results reported here supersede those reported in LAMS-1700 because of the change in the accepted value of U^{237} in the sample.)

Work done by:

R. E. Carter
E. T. Journey
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J. L. Yarnell

Written by:

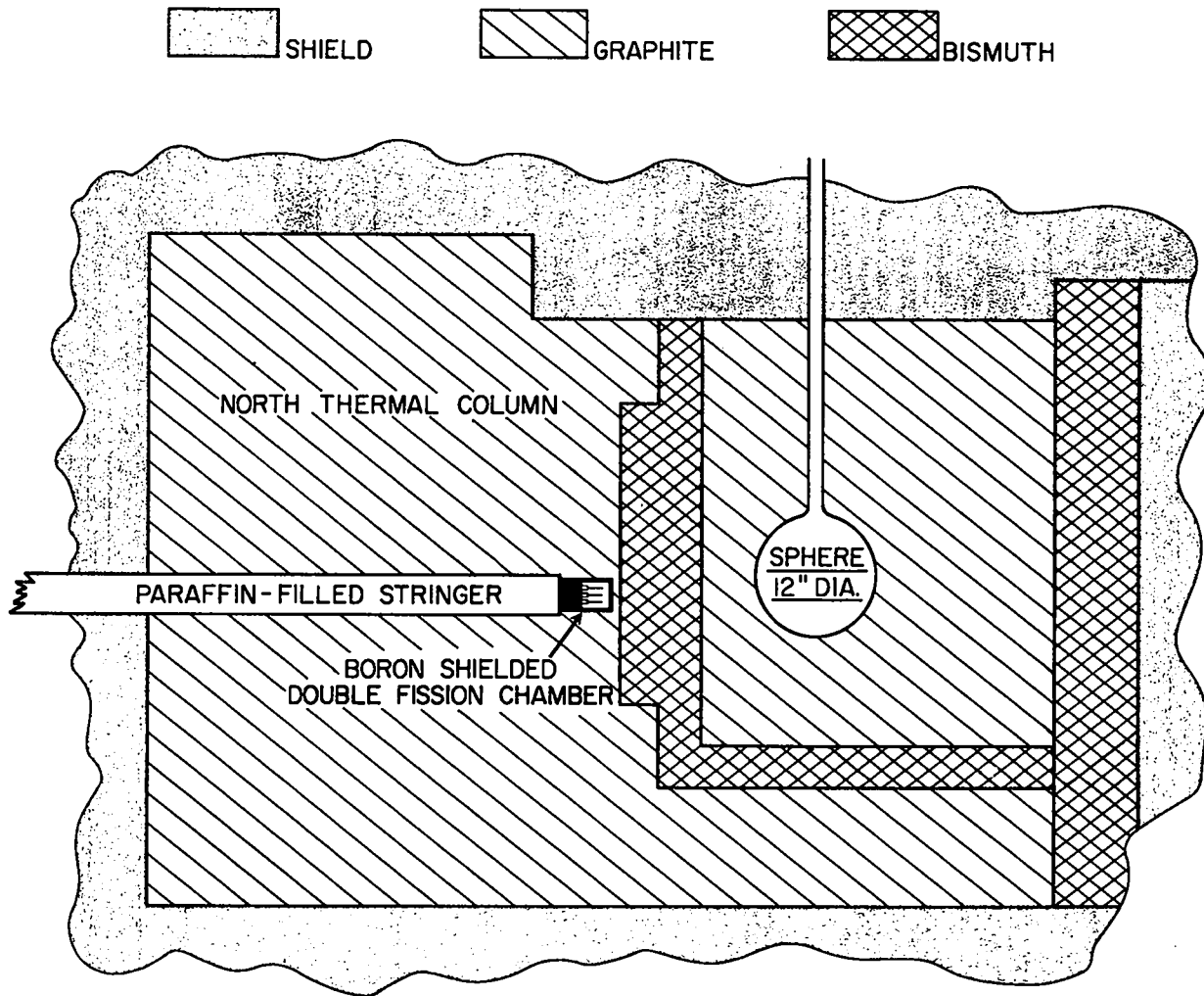
J. L. Yarnell

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A measurement of the average fission cross section of U^{237} for neutrons of intermediate energies was made by members of LASL Group P-2. The experimental arrangement is given in Fig. 2.1 and Figs. 2.2, 2.3, and 2.4 show the details of the fission chamber. The chamber had on one side a foil of Np^{237} , and on the other side a foil made from bombarded uranium containing U^{237} in sufficient concentration to permit measurement of its fission cross section. The ratio of the counting rates of the two sides of the chamber was observed as a function of the time. Since U^{237} was the only isotope present having a short half-life, a time-variation of the ratio could be related to the U^{237} cross section. The neutron spectrum was investigated by replacing the U^{237} foil with foils of U^{238} , U^{236} , and U^{235} , and measuring the ratio of fission counting rates of these isotopes to that of Np^{237} .

Background was found to be negligible; no counts were observed when the voltage was removed from the center electrode with the reactor on, and none were seen with the voltage on and the reactor off. In order to avoid drift, the electronic circuits were operated from voltage regulated supplies, and were kept in constant-temperature cabinets. The circuits were tested, and discriminator biases set by means of a pulser, before each set of six half-hour runs. The chamber was neither moved nor refilled during the U^{237} measurements. Bias curves for each side of the chamber were taken before each set of runs, and no changes were observed throughout the experiment. To investigate possible drift of the apparatus, the counting rates of the two sides of the chamber were



NORTH-SOUTH SECTION OF THE LOS ALAMOS WATER BOILER (SUPO)

Fig. 2.1 - A schematic view of the Los Alamos Water Boiler showing the location of the fission chamber in the reactor.

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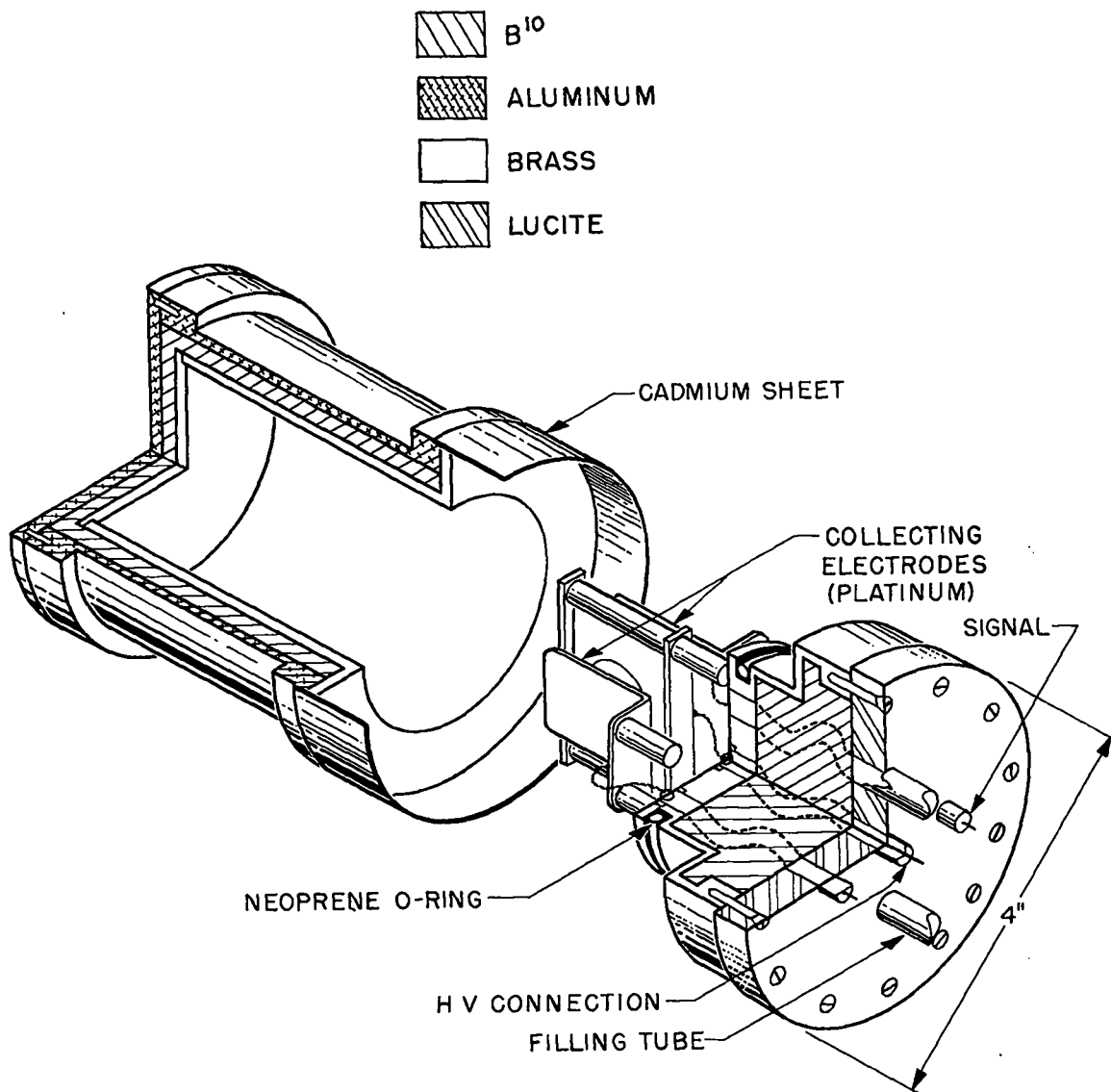


Fig. 2.2 - The boron shielded double fission chamber. The center high-voltage electrode carries a foil on each side. The two outer plates serve as collecting electrodes.

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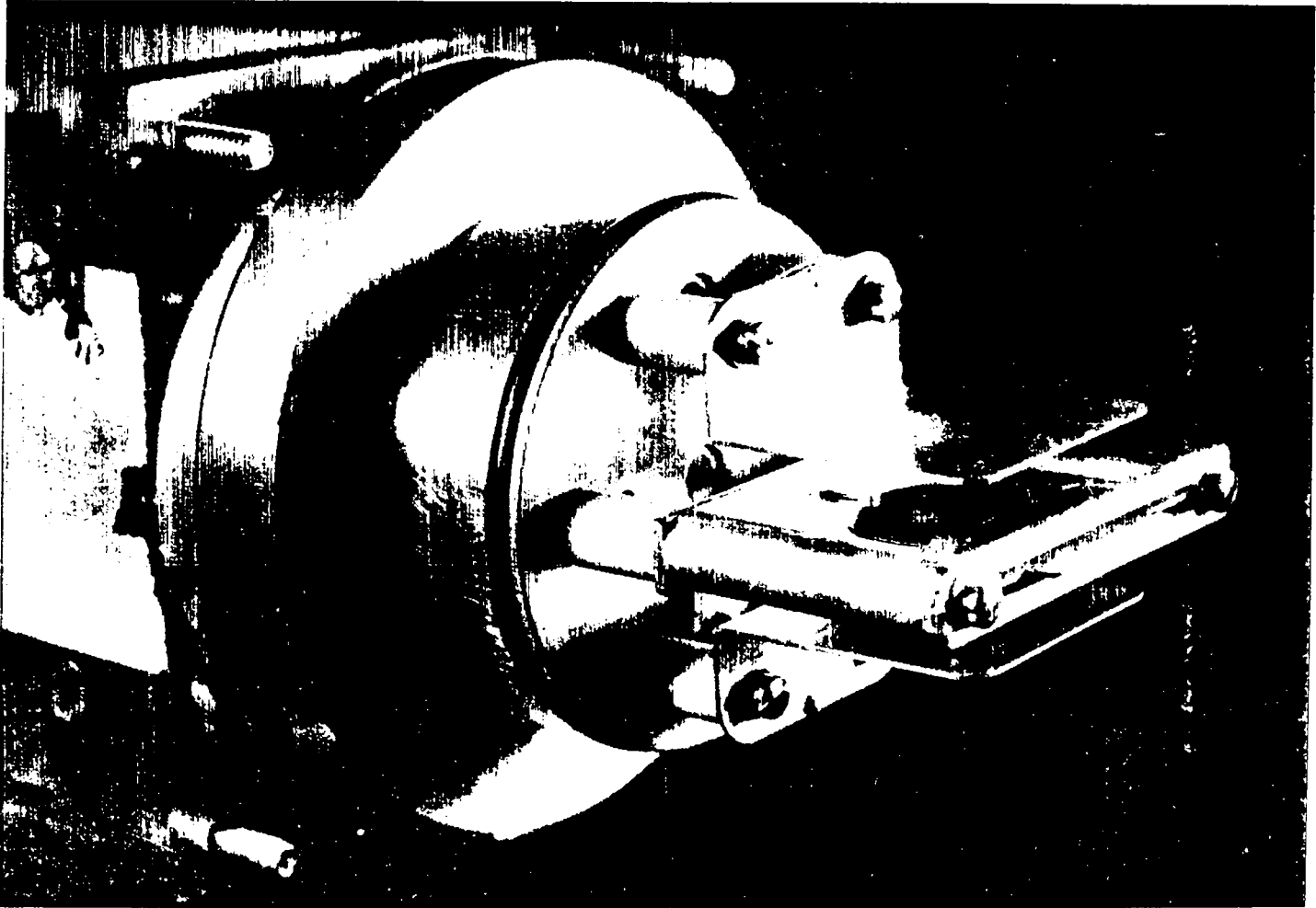


Fig. 2.3 - A photograph of the fission chamber with the cover removed.

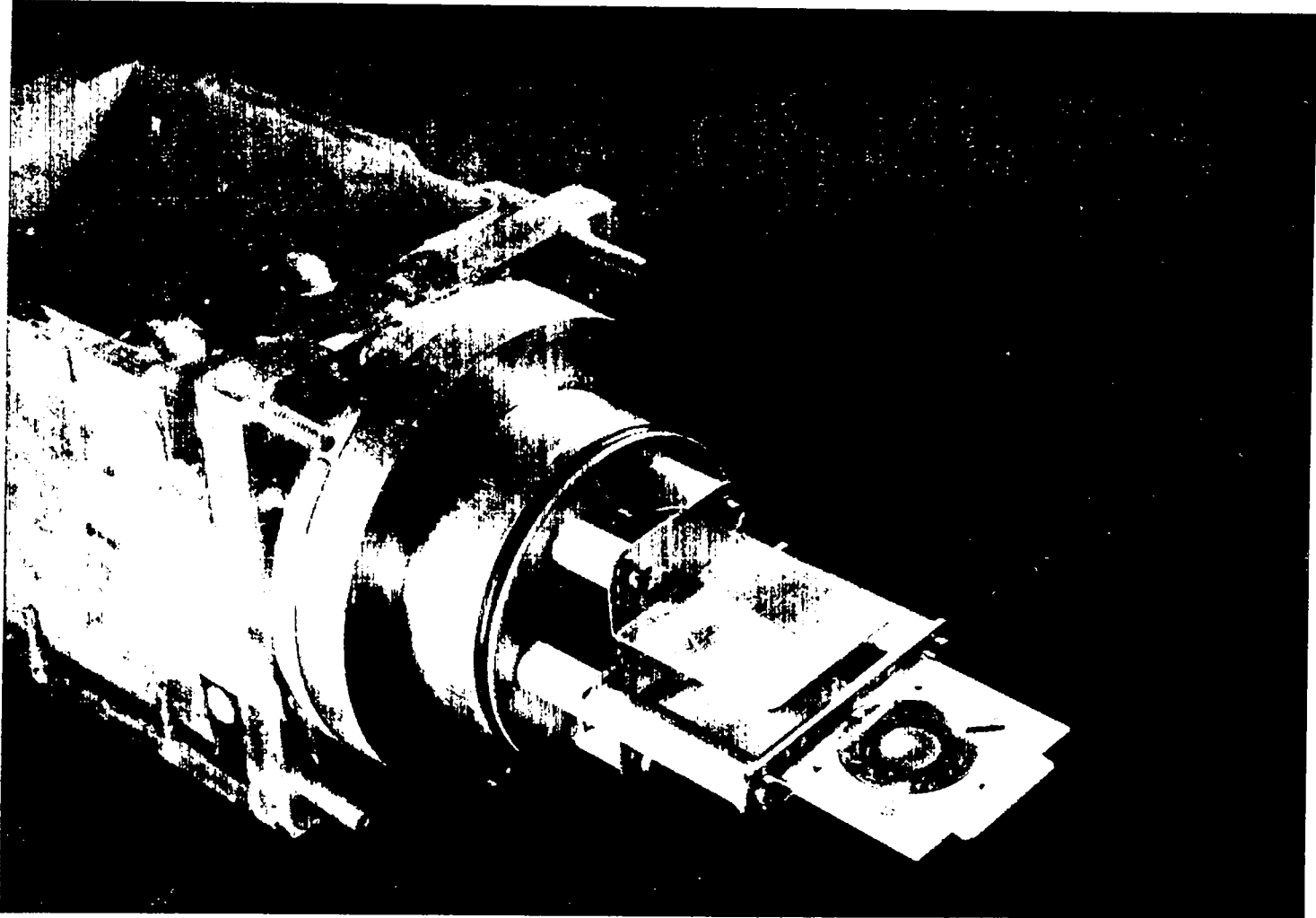


Fig. 2.4 - A photograph of the fission chamber showing the method of removal of the foils.

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separately compared to the pile monitor, a fission counter imbedded in the top of the bismuth wall which separates the north thermal column from the reactor core. Within the accuracy of the monitor, the Np^{237} counting rate remained constant, while that of the U^{237} decreased with time.

Masses of the standard foils were estimated by alpha counting and pulse height analysis. These measurements were made by Groups CMR-4 and J-11. The U^{235} content of each foil was measured by Group J-11, using thermal fission counting techniques.

The foil containing U^{237} was analysed by LASL Group J-11, using radiochemical and thermal fission counting techniques. The composition of the foil at the time counting was started (t_0) was found to be:

U^{235}	3.92×10^{14} atoms
U^{236}	2.43×10^{14} atoms
U^{237}	4.20×10^{14} atoms
U^{238}	$621. \times 10^{14}$ atoms

The Np^{237} foil used as a comparison contained 588×10^{14} atoms of Np^{237} .

The measured ratios of fission counting rate were extrapolated to zero bias. The magnitude of this correction, which arose from differences in slope of the bias curves for the various foils, was in no case more than 2%. The ratios of average cross sections obtained from measurements with the standard foils were found to be:

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$$\frac{\bar{\sigma}_{28}}{\bar{\sigma}_{37}} = 0.091, \quad \frac{\bar{\sigma}_{25}}{\bar{\sigma}_{37}} = 8.8, \quad \frac{\bar{\sigma}_{26}}{\bar{\sigma}_{37}} = 0.27$$

The ratio of the fission counting rate of the U^{237} foil to that of the Np^{237} foil is given by the expression:

$$R = \frac{N_{25} \bar{\sigma}_{25} + N_{26} \bar{\sigma}_{26} + N_{28} \bar{\sigma}_{28} + N_{37} \bar{\sigma}_{37} + N_{27} \bar{\sigma}_{27} e^{-\lambda t} + N_{27} \bar{\sigma}_{37} (1 - e^{-\lambda t})}{N_{37}^* \bar{\sigma}_{37}}$$

where N_i = atoms of isotope "i" in the U^{237} foil at $t = 0$.

N_{37}^* = atoms of Np^{237} in the comparison foil.

$\bar{\sigma}_i$ = average cross section of isotope "i".

λ = decay constant of $U^{237} = 0.1027 \text{ day}^{-1}$.

This may be written

$$R = A + B e^{-\lambda t}$$

where

$$A = \frac{N_{25}}{N_{37}^*} \cdot \frac{\bar{\sigma}_{25}}{\bar{\sigma}_{37}} + \frac{N_{26}}{N_{37}^*} \cdot \frac{\bar{\sigma}_{26}}{\bar{\sigma}_{37}} + \frac{N_{28}}{N_{37}^*} \cdot \frac{\bar{\sigma}_{28}}{\bar{\sigma}_{37}} + \frac{N_{27} + N_{37}}{N_{37}^*}$$

$$B = \frac{N_{27}}{N_{37}^*} \left[\frac{\bar{\sigma}_{27}}{\bar{\sigma}_{37}} - 1 \right]$$

The experimental data are shown in Fig. 2.5. The solid curve was obtained by fitting a curve of the form $R = A + B e^{-\lambda t}$ to the experimental points by the method of least squares. The constant λ was chosen to correspond to a half-life of 6.75 days for U^{237} . The values of the constants, when corrected to zero bias, together with the standard deviations obtained from the dispersion of the experimental points, are:

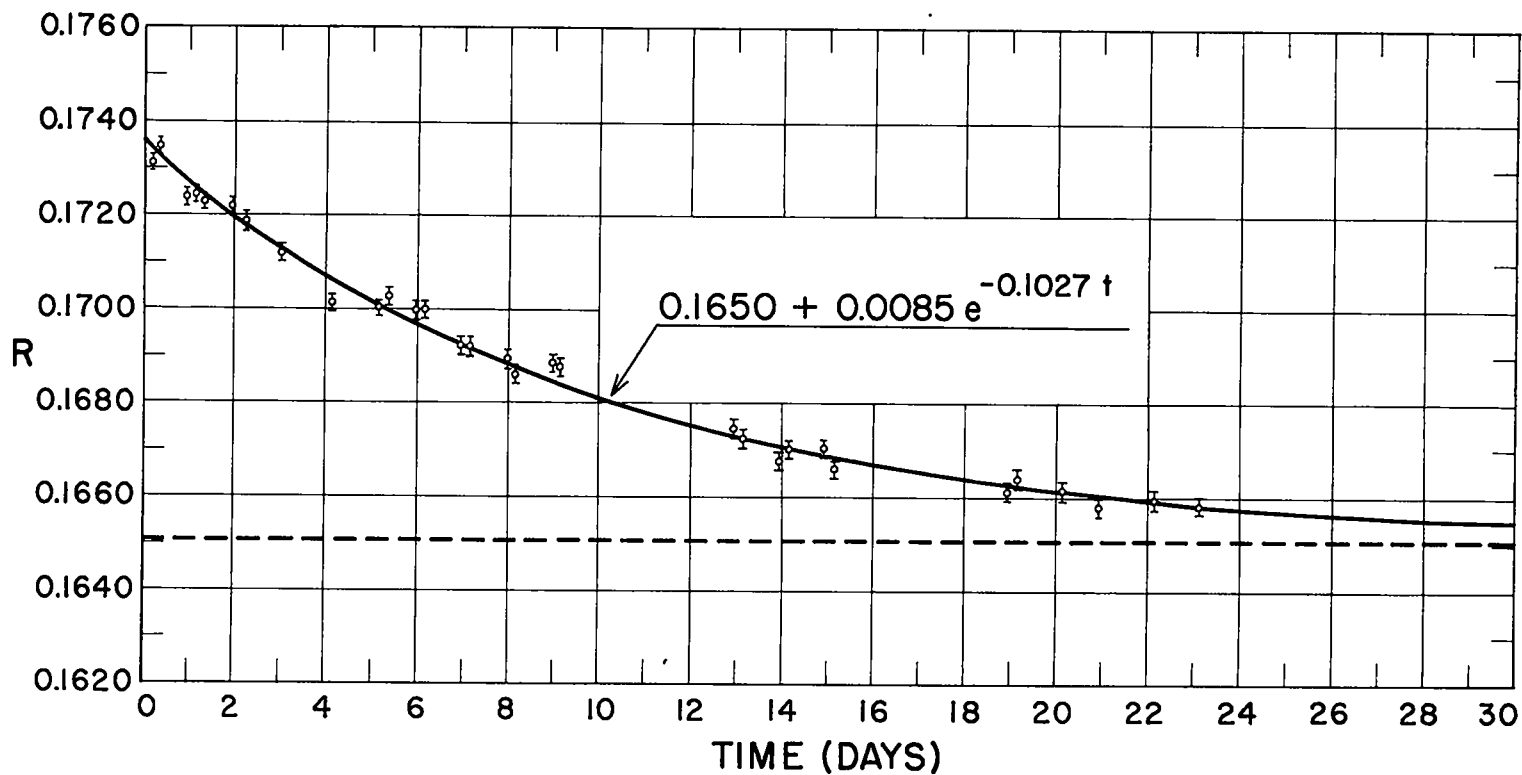


Fig. 2.5 - The ratio of the counting rate of the side of the chamber containing U^{237} to that of the side containing Np^{237} . The solid curve was obtained by fitting a function of the form $R = A + Be^{-0.1027t}$ to the experimental points by the method of least squares. A half-life of 6.75 days for U^{237} was assumed.

$$A = 0.1626 \pm 0.0001$$

$$B = 0.0084 \pm 0.0001$$

N_{27}/N_{37}^* was found to be 7.14×10^{-3} , and from this value and the measured value of B we obtain

$$\frac{\bar{\sigma}_{27}}{\sigma_{37}} = 2.18$$

To interpret these results, it is necessary to construct a neutron spectrum which, when combined with the known fission cross sections of U^{235} , U^{236} , U^{238} , and Np^{237} , will yield the experimental values of the ratios of average cross sections.

The low-energy end of the spectrum is determined by the B^{10} absorber which surrounded the chamber. Boron enriched to 82% B^{10} has a thermal cross section of 3270 barns. The cross section obeys a $1/v$ law up to approximately 10 kev, and then is constant. If the attenuation of the boron absorber is arbitrarily set equal to 1 at 10 kev, then the attenuation remains 1 at higher energies, and is equal to $1.31 e^{-27.2/\sqrt{E}}$ for $E < 10$ kev.

At energies below the lower limits of the fission spectrum and of inelastic scattering, the neutrons should follow the $1/E$ distribution characteristic of slowing down in a moderator. We therefore assume that, for energies below 100 kev, the neutron spectrum follows a $1/E$ law as modified by the boron attenuation.

Measurements of the spectrum above 1 Mev by J. E. Evans¹ indicate a distribution resembling a fission spectrum above 1.5 Mev connected to

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a function which increases rapidly as one goes to lower energies.

The above forms of the two ends of the spectrum were used as starting point of a trial and error construction of a spectrum which would give the measured values of the average cross sections. Values of the fission cross sections used in this process were taken from the compilations by R. L. Henkel² and the Brookhaven compilation.³

The average value $\bar{\sigma}$ of a cross section $\sigma(E)$ over a flux distribution $\phi(E)$ is given by

$$\bar{\sigma} = \frac{\int \phi(E) \sigma(E) dE}{\int \phi(E) dE}$$

This is the same as

$$\bar{\sigma} = \frac{\int E \phi(E) \sigma(E) \frac{dE}{E}}{\int E \phi(E) \frac{dE}{E}}$$

if $\Psi(E) = E \phi(E)$ and $dE/E = C d(\log_{10}E)$,

then
$$\bar{\sigma} = \frac{\int \Psi(E) \sigma(E) d(\log_{10}E)}{\int \Psi(E) d(\log_{10}E)}$$

This expression may be approximated by

$$\bar{\sigma} = \frac{\sum_i \Psi(E_i) \sigma(E_i) \Delta(\log_{10}E)}{\sum_i \Psi(E_i) \Delta(\log_{10}E)}$$

A range of 10 ev to 10 Mev was chosen for E ($\log_{10}E = 1.0$ to 7.0). A uniform interval of 0.1 was taken for $\Delta \log_{10}E$. Table 2.1 gives the values of E, σ_{25} , σ_{26} , σ_{37} , σ_{28} together with the assumed values of Ψ which gave the final fit. Figure 2.6 shows Ψ as a function of $\log_{10}E$. It is to be noted that the neutron flux between any two energies is proportional to the area under the curve between these energies. The

TABLE 2.1

FLUX DISTRIBUTION IN BORON-SHIELDED FISSION COUNTER

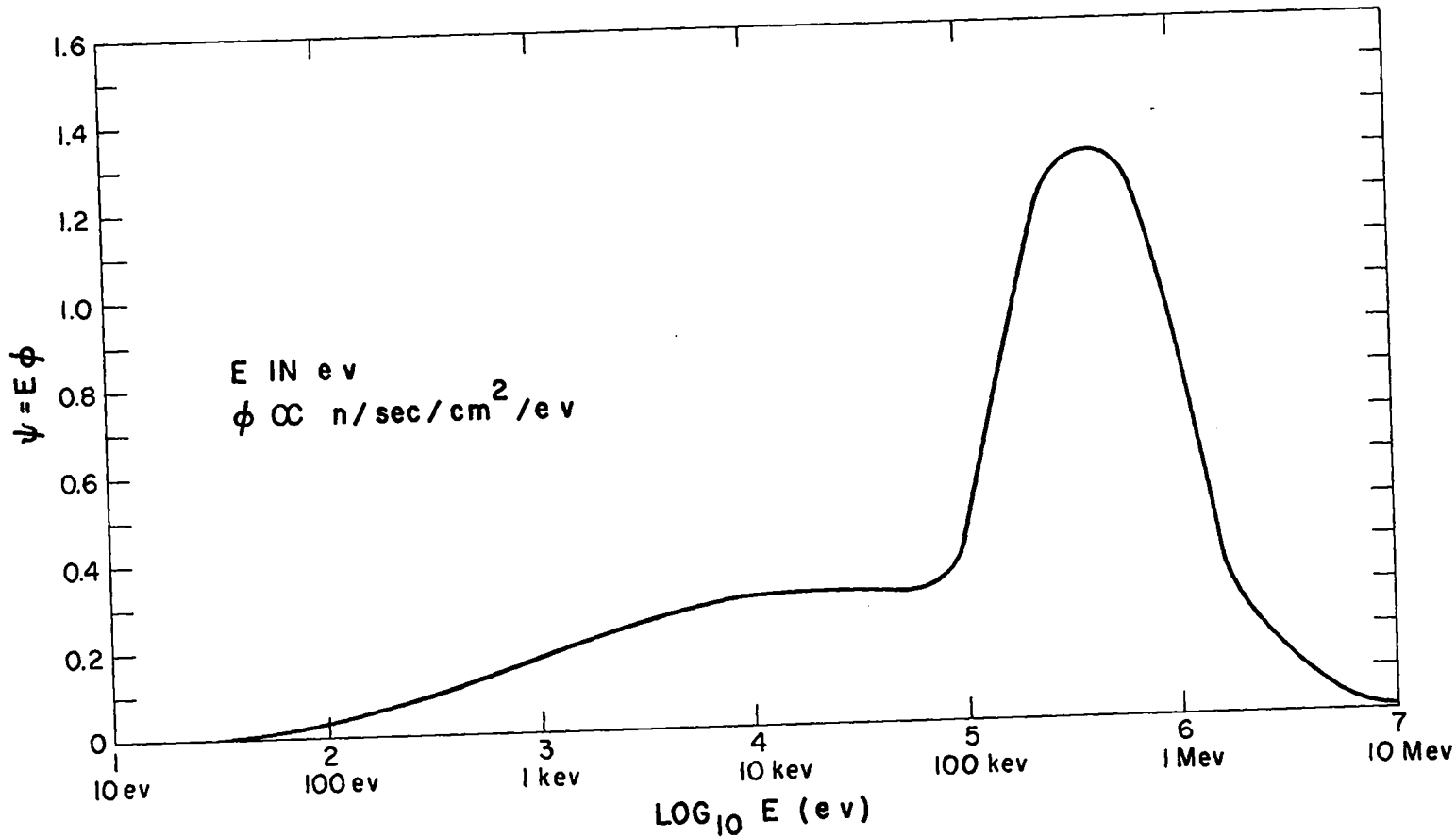
$\log_{10}E$	E, ev	σ_{25}	σ_{37}	σ_{26}	σ_{28}	ψ
1.5	3.16×10^1	29				0.00
1.6	3.98	46				0.01
1.7	5.01	57				0.01
1.8	6.31	36				0.01
1.9	7.94	27				0.02
2.0	1.0×10^2	25				0.03
2.1	1.26	24				0.04
2.2	1.59	22				0.05
2.3	2.00	20				0.06
2.4	2.51	18				0.07
2.5	3.16	16				0.09
2.6	3.98	14.3				0.10
2.7	5.01	12.5				0.13
2.8	6.31	11.4				0.14
2.9	7.94	9.9				0.16
3.0	1.00×10^3	8.8				0.18
3.1	1.26	7.9				0.19
3.2	1.59	7.1				0.21
3.3	2.00	6.4				0.22
3.4	2.51	5.9				0.24
3.5	3.16	5.4				0.25
3.6	3.98	5.0				0.27
3.7	5.01	4.6				0.28
3.8	6.31	4.3				0.29
3.9	7.94	3.8				0.30
4.0	1.00×10^4	3.76				0.31
4.1	1.26	3.53				0.31
4.2	1.59	3.30				0.31
4.3	2.00	3.10				0.31
4.4	2.51	2.88				0.31
4.5	3.16	2.70				0.31
4.6	3.98	2.55				0.31
4.7	5.01	2.40				0.31
4.8	6.31	2.25				0.31
4.9	7.94	2.11				0.31
5.0	1.00×10^5	2.00				0.31
5.1	1.26	1.87				0.63
5.2	1.59	1.75				0.85
5.3	2.00	1.63				1.06
5.4	2.51	1.52				1.22
5.5	3.16	1.43				1.28
5.6	3.98	1.34	0.01			1.29

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TABLE 2.1 (Cont'd.)

$\log_{10} E$	E, ev	σ_{25}	σ_{37}	σ_{26}	σ_{28}	ψ
5.7	5.01	1.24	0.40			1.28
5.8	6.31	1.17	0.76	0.01		1.22
5.9	7.94	1.15	1.12	0.09		1.06
6.0	1.00×10^8	1.24	1.29	0.34		0.85
6.1	1.26	1.25	1.39	0.66	0.02	0.63
6.2	1.59	1.25	1.46	0.63	0.40	0.36
6.3	2.00	1.26	1.49	0.77	0.53	0.24
6.4	2.51	1.27	1.51	0.82	0.54	0.18
6.5	5.16	1.27	1.53	0.83	0.54	0.13
6.6	3.98	1.28	1.54	0.93	0.54	0.09
6.7	5.01	1.24	1.52	0.86	0.55	0.06
6.8	6.31	1.43	1.95	1.34	1.13	0.03
6.9	7.94	2.07	2.43	1.63	1.13	0.01
7.0	1.00×10^7	2.12	2.50	1.63	1.13	0.00

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Fig. 2.6 - Proposed neutron spectrum present in the fission chamber. Total flux between any two energies is proportional to the area under the curve between these energies.

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ratios of average cross sections as computed from the proposed spectrum, and as measured experimentally, are given below:

Ratio	Calculated	Experimental
$\bar{\sigma}_{25}/\bar{\sigma}_{37}$	8.83	8.8
$\bar{\sigma}_{26}/\bar{\sigma}_{37}$	0.269	0.27
$\bar{\sigma}_{28}/\bar{\sigma}_{37}$	0.0921	0.091

The calculated average cross sections for the proposed spectrum are

U ²³⁵	2.88 barns
U ²³⁶	0.088 barn
U ²³⁸	0.030 barn
Np ²³⁷	0.326 barn

The above value of the Np²³⁷ cross section, together with the measured value of $\bar{\sigma}_{27}/\bar{\sigma}_{37}$, results in a value of 0.71 barn for the fission cross section of U²³⁷ averaged over the spectrum of Fig. 2.6.

The peak in the neutron spectrum between 100 kev and 1 Mev may be due to the presence of the bismuth between the chamber and the reactor core (see Fig. 2.1). As an auxiliary experiment, neutron spectra were measured with the chamber partly retracted, and with varying amounts of graphite between the chamber and the bismuth. It was found that the peak at ~ 300 kev decreased with respect to the 1/E portion of the spectrum as the thickness of the graphite was increased.

It is difficult to state precise limits on the accuracy of the results of this experiment, since several of the sources of uncertainty

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can be estimated only subjectively. We feel that the average cross section of U^{237} has been determined to $\pm 10\%$. The proposed neutron spectrum may be divided into four regions by the thresholds of Np^{237} , U^{236} , and U^{238} , and it is believed that the relative flux in each region is known to $\pm 10\%$. The uncertainty associated with individual points on the flux distribution is considerably greater.

References

1. J. E. Evans, Los Alamos Scientific Laboratory Report LA-1395, March 1951.
2. R. L. Henkel, Los Alamos Scientific Laboratory Report LA-1495, November 1952.
3. Brookhaven National Laboratory Reports BNL-170 and BNL-170B.





PART 3

SOME COMMENTS ON THE (n,f) EXCITATION
FUNCTION OF U²³⁷


Work done by:

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The water-boiler average fission cross section for intermediate energy neutrons and the Topsy average fission cross section for degraded fission neutrons are about the same value within experimental error, 0.70 ± 0.07 barns and 0.65 ± 0.10 barns, respectively. The fission cross section at thermal energies was not measurable, but the results of attempted measurement did not exclude a thermal fission cross section of < 2 barns. The water-boiler intermediate energy spectrum and the Topsy spectrum are plotted together for easy comparison in Fig. 3.1.

Although one believes, from these measurements, that U^{237} neutron-induced fission behaves more like fission in U^{234} , U^{236} , and U^{238} , which have effective thresholds for fission, than like U^{235} , Pu^{239} , etc., which have high thermal fission cross sections, it is impossible from the integral type measurements made in the two reactors to reconstruct a unique excitation function. However, certain plausible arguments can be advanced which permit one to guess, within very broad limits, how the excitation function might behave, consistent with the measurements reported in the previous two papers.

The (n,f) excitation functions of all the fissionable heavy elements reach a plateau region at neutron energies below 2 Mev and remain roughly constant ($\pm 10\%$) thereafter to neutron energies in a range that extends to > 5 Mev. The nuclide with the longest plateau is Pu^{239} which varies between 2.2 and 1.75 barns in the energy region from 20 kev to 6 Mev.

Attempts have been made to correlate these plateau values with Z

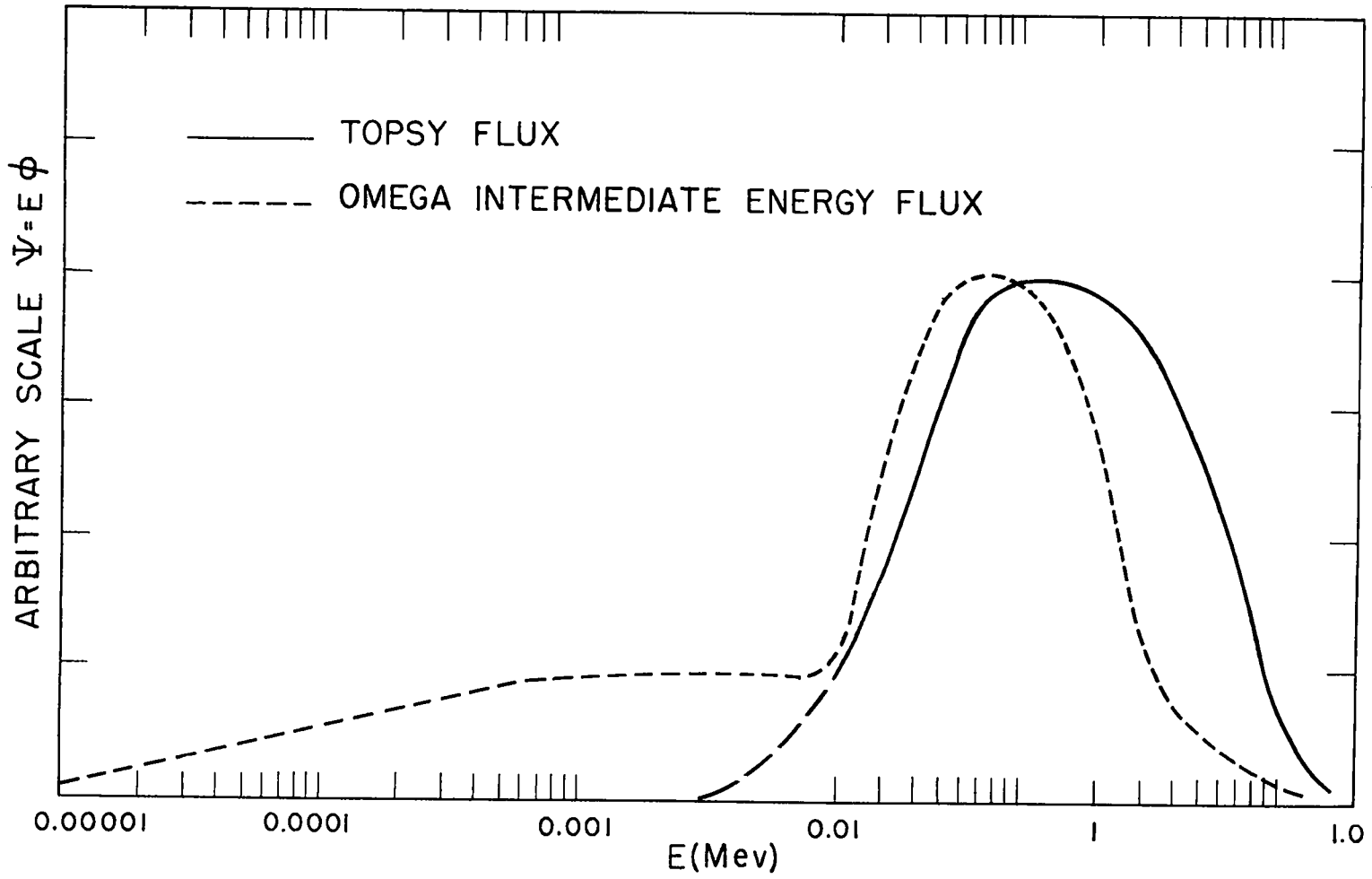


Fig. 3.1 - Plot of Omega water-boiler intermediate energy spectrum and the Topsy spectrum.

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and A. Barschall and Henkel¹ have plotted all the heavy element fission cross sections which have been measured at 3 Mev neutron energy against $Z^{4/3}/A$. Their plot is reproduced in Fig. 3.2. The fission cross section of U^{237} at 3 Mev which one would predict from this curve is ~ 0.7 barns. Since resonances or departures from the smooth curve behavior in the known excitation functions are not so great as to invalidate comparison with a smooth curve, a reasonable hypothesis to explain the similarity in the $\bar{\sigma}_f$ values measured at the water-boiler and in Topsy and the predicted 3 Mev value is that the excitation function is roughly constant at its plateau value over most of the energy distribution of neutron flux in both reactors.

The energy region in which the excitation function first assumes its plateau value may be crudely estimated by assuming a reasonably sloped shape in the region of critical energy for fission. If the excitation function in the critical energy region is described by the equation

$\sigma(E) = A + BE$ one finds that a fit to the limiting assumptions exists when $\sigma(E) = 0.2 + 4 \times 10^{-3}E$ kev up to 150 kev and is constant thereafter. This slope resembles the slopes of all other measured threshold-type excitation functions for neutron-induced fission in Th, Pa, U, and Np isotopes. In the region of the critical energy for fission they show a rise of about a factor of two per 50 to 100 kev of neutron energy.

We infer from these considerations and the measurements that although U^{237} is not noticeably fissionable at neutron energies near thermal, it

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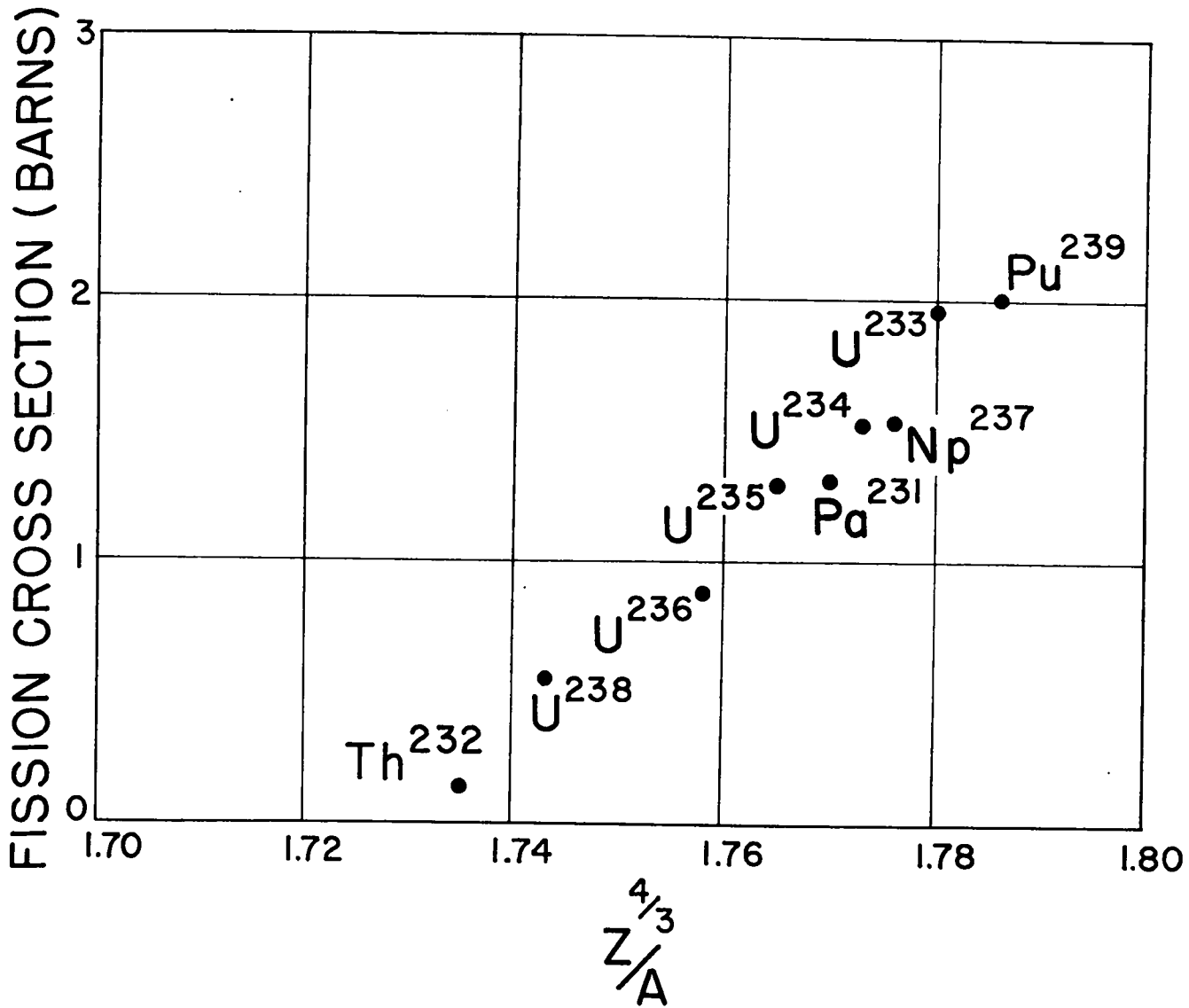


Fig. 3.2 - Plot of fission cross sections of heavy elements measured at 3 Mev neutron energy vs. $Z^{4/3}/A$.

[REDACTED]

is very close to becoming noticeably fissionable. If the maximum value of the cross section is reached at the plateau, then this plateau begins at a neutron energy lower than 200 kev and remains roughly constant at a value of 0.6 to 0.8 barns. If the excitation function passes through a maximum before reaching a plateau region, then the effective threshold for fission will be higher.

The systematic behavior of the critical energy required for fission of heavy elements has been used to guess at the thermal fissionability of U^{237} . The critical energy for fission is nearly constant and falls in the region 5.5 to 6.1 Mev. Evidence for this generalization is that the measured photo-fission effective thresholds in U^{235} , U^{238} , U^{233} , Pu^{239} , and Th^{232} are all within 0.3 Mev of one another² and that the (n,f) excitation functions for the nuclides Th^{232} , U^{233} , U^{235} , U^{236} , U^{238} , and Np^{237} all show sharp increases in the neutron energy region 5.3 to 5.8 Mev. We assume that the process which operates in this region of sharp increase is $M^{Z,A} \xrightarrow{n,n'} M^{Z,A^*}$ and that the excitation energy is approaching the critical energy for fission. We observe that the sharp increase in the $U^{238}(n,f)$ excitation functions begins at 5.4 Mev. Since absorption of a neutron by U^{237} produces 5.98 Mev of excitation energy, we guess that even at thermal neutron energies, neutron absorption by U^{237} should produce a compound nucleus very close to its critical energy for fission and, perhaps, visibly fissionable. The actual size of the thermal fission cross section will be governed by the importance of resonance capture at thermal neutron energies.

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Table 3.1 summarizes the data in the literature from which the above generalizations have been drawn.

References

1. H. H. Barschall and R. L. Henkel, Los Alamos Scientific Laboratory Report LA-1714, August 1954.
2. Koch, McElhinney and Gasteiger, Phys. Rev., 77, 329 (1950).
3. J. R. Huizenga and L. B. Magnusson, Argonne National Laboratory Report ANL-5158, November 1953.
4. M. D. Goldberg, J. A. Harvey, D. J. Hughes, and V. E. Pilcher, Brookhaven National Laboratory Report BNL-250, August 1, 1954.

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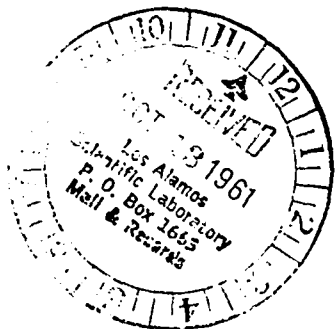
TABLE 3.1

HEAVY ELEMENT FISSION "THRESHOLDS" AND CROSS SECTIONS

Target nuclide	Binding energy of added neutron ³	Measured effective threshold energy for fission from			Thermal (n,f) cross section, barns ⁴
		B.E.+E _t	(γ,f) ²	(n,n'f) ¹	
Th ²²⁷	7.05				1500±1000
Th ²²⁸	5.44				≤ 0.3
Th ²²⁹	6.69				45±11
Th ²³⁰	5.18				≤ 10 ⁻³
Th ²³¹	6.20		5.4	5.8	--
Th ²³²	5.11	6.3			< 0.2 mb
Th ²³³	5.99				≤ 20
Pa ²³⁰	--				1500±250
Pa ²³¹	5.28	5.6			~ 10 ⁻²
Pa ²³²	6.95				700±100
Pa ²³³	5.35				< 0.1
U ²³⁰	5.83				25±10
U ²³¹	7.20				400±300
U ²³²	5.94		5.2	5.6	80±20
U ²³³	6.74				533±10
U ²³⁴	5.37	5.7	5.3	5.8	≤ 0.65
U ²³⁵	6.29			5.3	580±8
U ²³⁶	5.45	6.1			--
U ²³⁷	5.98		5.1	5.4	< 2*
U ²³⁸	4.87	5.8			< 0.5
U ²³⁹	5.77				(12±8)
Np ²³⁴	--				900±300
Np ²³⁶	6.81				< 10 ⁵
Np ²³⁷	5.23	5.5			2x10 ⁻²
Np ²³⁸	6.41				1600±100
Np ²³⁹	4.83				< 3
Pu ²³⁸	5.71				18±2
Pu ²³⁹	6.28				750±15
Pu ²⁴⁰	5.55				< 40
Pu ²⁴¹	6.17				1100±100
Pu ²⁴²	5.15				--
Pu ²⁴³	--				--

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