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THE FISSION CROSS SECTION OF U^{235} FROM 0.4 to 1.6 MEV

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ABSTRACT

The U^{235} fission cross section was measured with about 80 kev resolution from 400 to 1600 kev by comparison with the n,p scattering cross section.

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1. Introduction

The fission cross section of U^{235} has been measured in the region 400 to 1600 kev by several workers.^{1,2} The present experiment is similar to that of Hall, Koontz, and Rossi,² whose results have been widely used, and who quote an accuracy of about 5% for their most certain points. Those authors made their measurement in 1943-1944 using neutrons from the $Li(p,n)$ reaction. At the time the measurement was made it was not realized that two groups of neutrons were present in this source. This fact should introduce an error into their measurements in some energy regions so that their results may be uncertain by as much as 10%. Since the war there have been many improvements in techniques and in available equipment so that, in view of the importance of the fission cross sections to the project and the desirability of using these cross sections to determine neutron fluxes, a remeasurement of the U^{235} fission cross section was undertaken.

The method used in this measurement was to determine the fission cross section relative to the scattering cross section of hydrogen and to use the known hydrogen cross section to obtain the fission cross section. The n,p total cross section has been measured by several groups³ and is expected to be reliable to about 1/2% near 1-1/4 Mev.

The experiment consists of counting proton recoils ejected from a hydrogenous radiator in a neutron flux while simultaneously counting fissions occurring in a thin film of U^{235} placed near the radiator.

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2. Experimental Equipment

The fission counter was essentially a parallel plate ionization chamber (actually the electrodes were sections of cylinders) and the proton recoil counter was a cylindrical proportional counter. Both counters were housed in a cylinder which contained the argon-carbon dioxide mixture in which the counters operated. Figure 1 shows the essential parts of the proportional counter.

2.1 Proton Recoil Counter

The proton recoil counter was 2" in diameter and had an active volume 2-1/2" long. The cylinder (A) was made of 0.009" stainless steel lined with 0.002" platinum. The wire (B) was 0.002" diameter stainless steel. The active volume was defined by means of 1/4" diameter field tubes (C) held at the proper potential to maintain radial fields up to the ends of the counting volume.⁴ Inside the field tubes and insulated from the wire was a 1/16" diameter guard tube (D) which was maintained at the potential of the wire. Inside the guard tube was a 0.016" outer diameter tube through which the wire was threaded. This tube was in contact with the wire and served as a convenience in assembly. The counter is operated at +1000 to +2500 volts on the wire, with the cylinder grounded.

2.2 Fission Counter

The negative electrode of the fission counter is the grounded cylinder of the proportional counter. The collecting electrode of the

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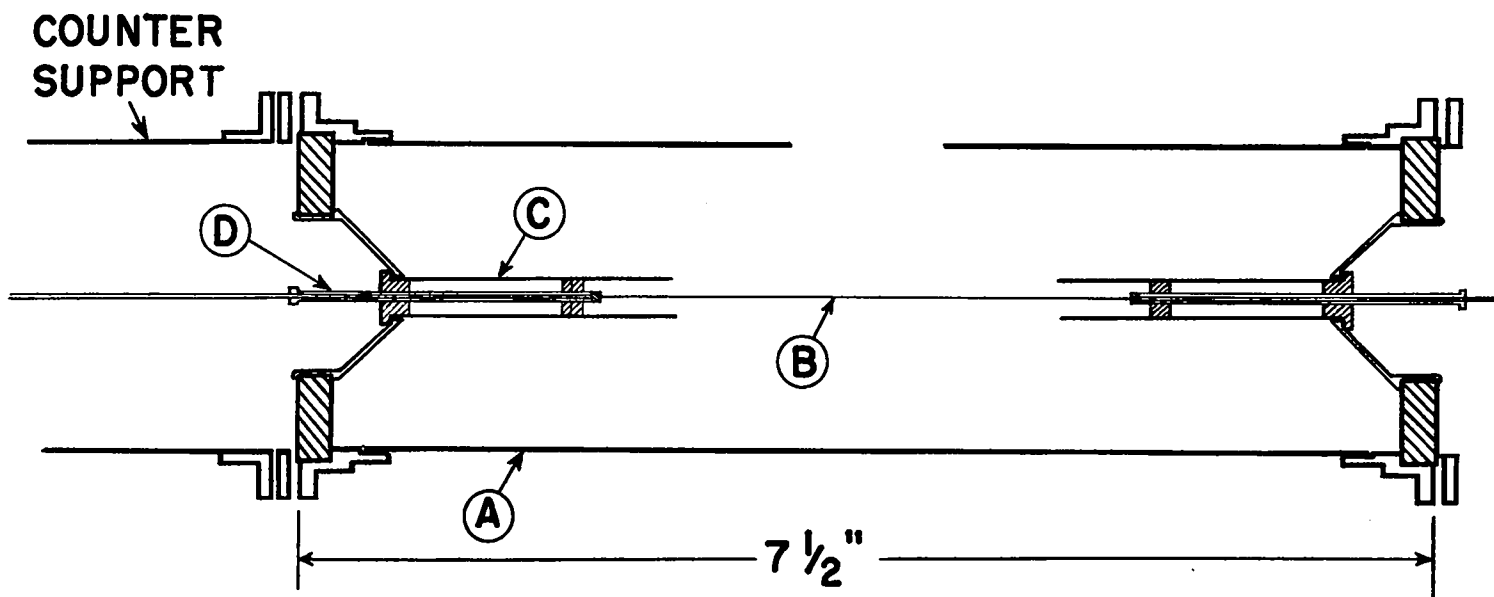


Fig. 1--Counter assembly (shaded areas Teflon).

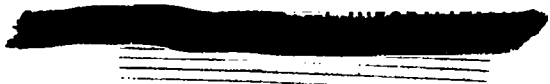
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fission counter is an aluminum plate 3" wide by 3-1/2" long by 0.005" thick, bent into a section of a cylinder concentric to the proportional counter and separated by 3/4" from that electrode. The double counter is mounted inside a gas envelope consisting of an 0.018" wall stainless steel cylinder 3-1/2" diameter and 17" long with a hemispherical copper cap on one end and 1/4" thick steel base plate on the other. The foils are located 12-3/4" from the base plate. The envelope is normally surrounded by 1/32" of cadmium to reduce the effect of room-scattered neutrons. The counter gas is continuously purified by a hot calcium purifier.

2.3 Foils

The proton radiators consisted of thin films (65 to 650 micrograms total weight) of glycerol tristearate deposited in a 1-1/32" diameter circle on a platinum disk 2" diameter by 0.003" thick. The glycerol tristearate was deposited by evaporation in a vacuum of 5×10^{-5} mm Hg or better, and at a temperature of about 250°C. The platinum foil was bent around the cylinder of the proportional counter, the radiator being placed over a 1-1/2" diameter hole in the cylinder. The amount of glycerol tristearate on the foil was determined by weighing it on a microbalance. Examination of foils subjected to severe bending indicated that the glycerol tristearate does not flake off under the handling conditions used.

The U²³⁵ foils were prepared by Buddy Warren of Group J-11 and


consisted of 200 to 500 micrograms of U^{235} electroplated onto a 1-1/32" ^{5.36 cm²} diameter circle on a platinum disk 2" diameter by 0.003" thick. The platinum foil containing the U^{235} was bent around the cylinder of the proportional counter so that the two foils were back to back and the deposits were centered one upon the other.

3. Experimental Method

3.1 Setup

The source of neutrons for this experiment was the T(p,n) reaction using the 2.5 Mev electrostatic accelerator for proton acceleration. A 3 cm long tritium gas target was used. The entrance foil was 0.2 mil aluminum and produced straggling in the proton energy of about 20 kev half width at half maximum. This straggling was due primarily to inhomogeneity of the foils. In addition, the stopping power of the tritium gas produces an energy spread of 20 or 30 kev at the target pressures used in this experiment.

The counter was located at zero degrees to the proton beam and the foils were 10-1/2 cm from the center of the target.

3.2 Fission Counting

Pulses from the fission counter were fed into a Model 101 preamp and amplifier with a decay time of 4 microseconds. From the amplifier the pulses were sent to a pair of Model 710 discriminator-scalers.

In order to determine the optimum bias setting for the discriminators the pulse height distribution of fission fragments was observed on a Johnstone 18 channel pulse height analyzer.⁵ A convenient counting

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rate was obtained by surrounding the counter with paraffin to reduce the average neutron energy. From observation of the pulse height spectrum and from the height of alpha pulses from the U foil, a bias voltage was chosen such that no alphas were detected, but as large a fraction of the fissions as possible was detected. Except for fixing the bias for the fission discriminators, the multichannel analyzer was not used to count fission pulses. The two discriminator-scalers were used in parallel as a check that the scalers were operating properly. Figure 2 is an example of a fission pulse height distribution obtained with this counter. The pulse spectrum is distorted from the energy spectrum of fission fragments because of the electron collection effect in the ionization chamber.

3.3 Proton Recoil Counting

The proton recoil (proportional) counter was operated at a pressure of argon-carbon dioxide which would assure that all protons of interest would be stopped in the gas. Since the counter is cylindrical, a proton at a large angle to the neutron beam must not be allowed to strike the wall. A pressure of 32 psi absolute was sufficient to achieve this result for 1 Mev neutron energy. The counter pressure had to be sufficiently low that pulses caused by X rays from the accelerator did not reach sizes large enough to become confused with proton recoil pulses. This condition required pressures of less than about 45 psi absolute for 1 Mev neutrons. Because of these requirements, the counter gas pressure was adjusted as required for different neutron energies. The

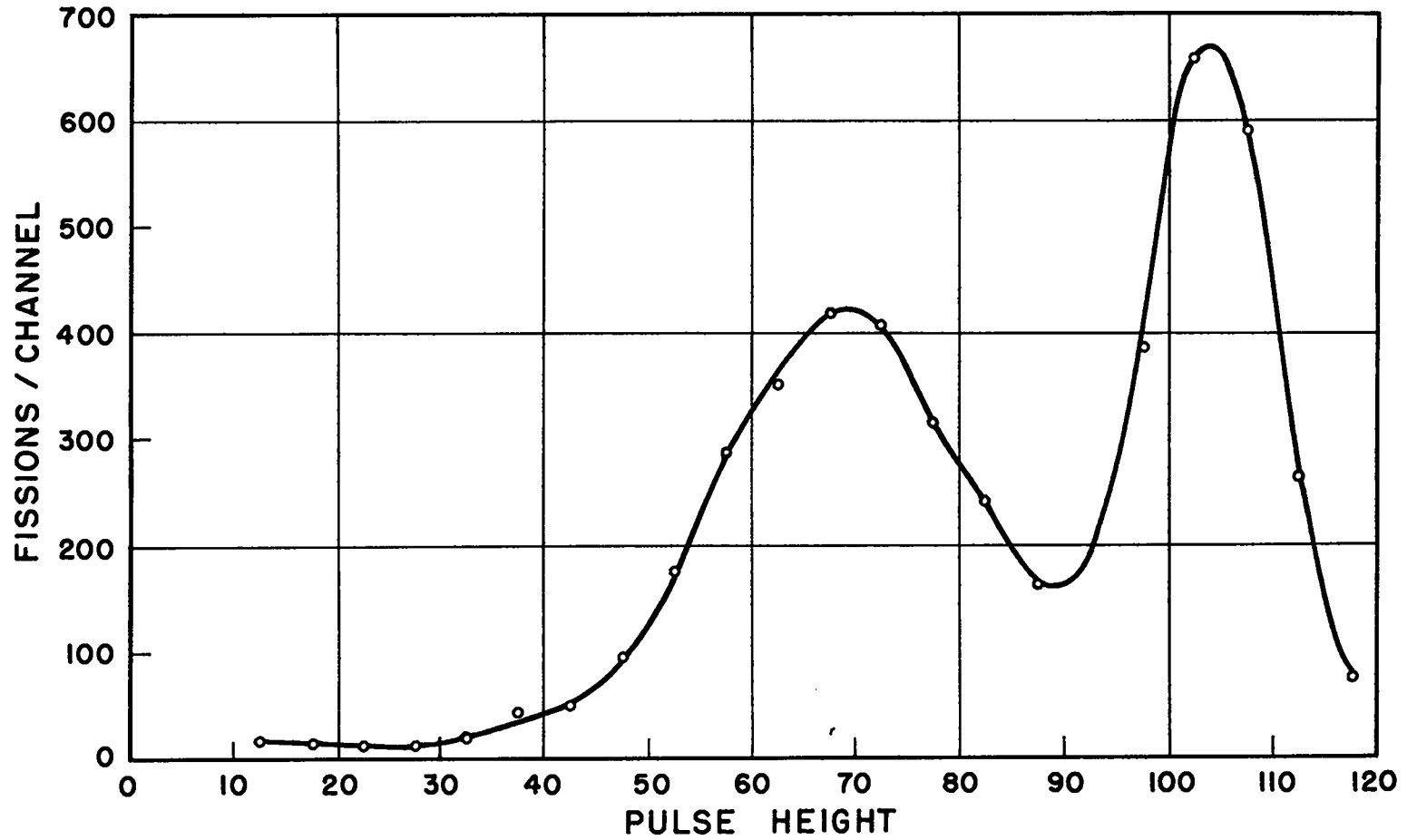


Fig. 2--Fission pulse height distribution.

voltage on the counter was adjusted to give a gas multiplication between 10 and 30.

Pulses from the proton recoil counter were fed into a Model 101 preamp and to a Model 101 amplifier with a delay line clipper which gave 3-microsecond wide pulses. From the amplifier they went to the 18 channel pulse height analyzer. The amplifier gain and counter multiplication were adjusted to give proton recoil pulses with a maximum height of about 100 volts. The analyzer was set to cover the range of pulse heights from about 25 to 115 volts with 5 volt channel widths. Since the proton recoil spectrum is obscured by carbon recoils below 28% of the maximum proton pulse height, this range covers the useful part of the proton recoil energy spectrum.

From the observable portion of the proton recoil spectrum, we must deduce the number of proton recoils which were made during the time a known number of fissions were detected. The pulse height distributions of proton recoils, except for deviations caused by foil thickness and neutron energy spread, are rectangular. That is, there are equal numbers of pulses per pulse height interval up to a maximum pulse height beyond which there are no pulses. To a first approximation, the fraction of the proton recoils which lie above a certain pulse height P is $\frac{P_m - P}{P_m}$ where P_m is the maximum pulse height. In this approximation, if we count only the upper two-thirds of the pulse height spectrum, then the total number of proton recoils which occurred is $3/2$ times the number observed. In practice the theoretical pulse height

distribution is deduced according to the methods given in Rossi and Staub,⁶ as worked out by K. M. Case,⁷ taking into account the thickness of the glycerol tristearate film, and the neutron energy spread. The theoretical curve is fitted to the experimental curve and the total number of proton recoils computed. Figure 3 is an example of the kind of fit which was obtained. The solid line represents the theoretical curve adjusted for the channel width and neutron energy spread. The rise in the solid curve at low energies represents carbon recoils. The dotted rectangle is the energy distribution of protons from an infinitely thin radiator due to neutrons of a single energy. The points are the experimental points.

Knowing the amount of U^{235} on the fission foil and the amount of hydrogen in the proton radiator and the numbers of fissions and proton recoils which occurred during a run, one may compute the ratio of cross sections of U^{235} and H except for numerous corrections and backgrounds.

3.4 Energy Variation of σ_f

The experiment may be thought of as having two parts. One part involves determination of the energy dependence of the fission cross section and the other part the determination of an absolute cross section at some energy. The experiment is broken up in this manner because, in order to find the absolute number of fissions from a foil, the uranium deposit on that foil must be quite thin compared to the range of fission fragments. This limits the counting rates available to about a thousand counts per hour. On the other hand, if the absolute

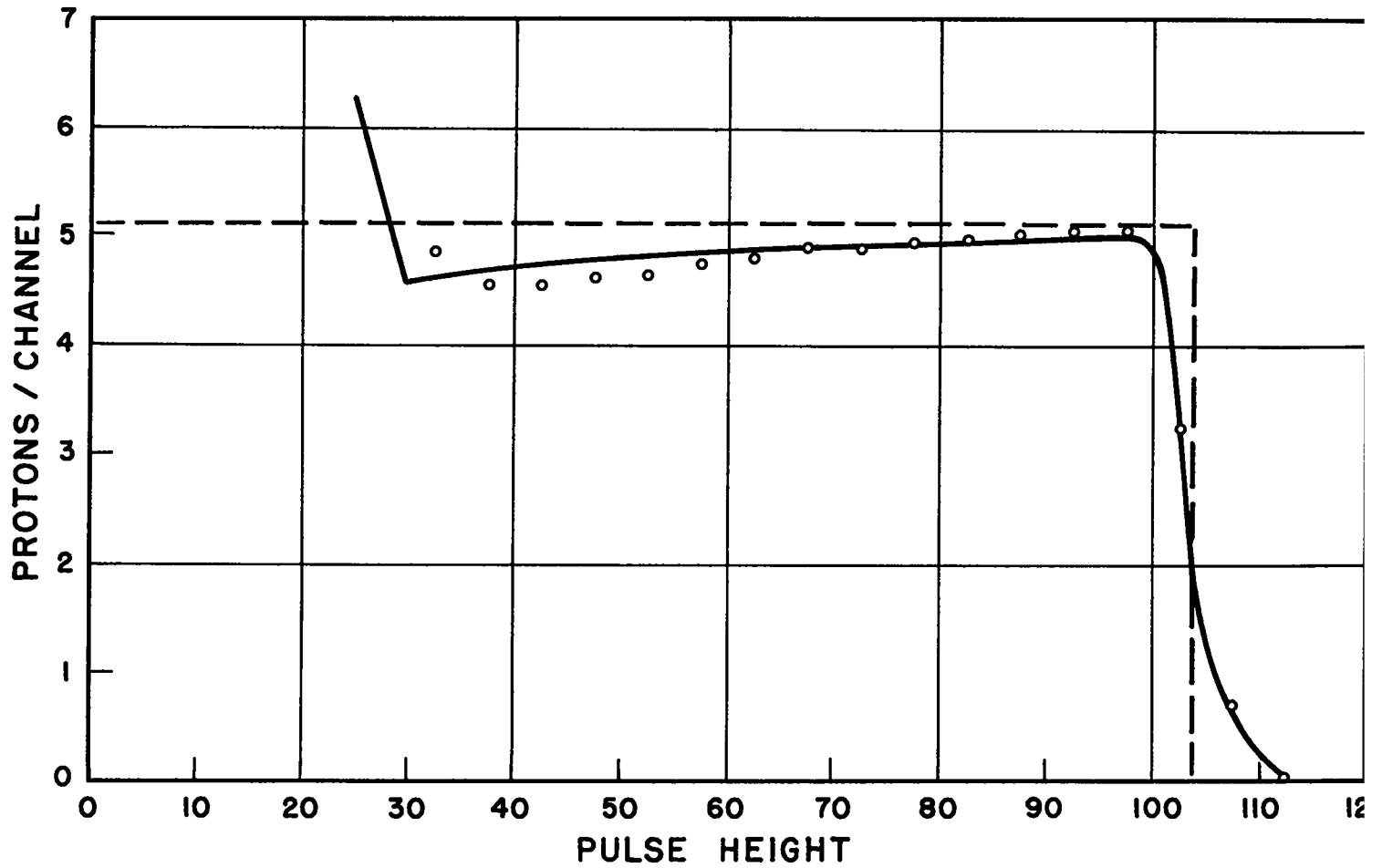


Fig. 3--Proton pulse height distribution, 1.00 Mev neutrons, 70 micrograms/cm² radiator.

[REDACTED]

cross section is known at one energy, then we may use a rather thick uranium layer for counting fissions at other energies and normalize the data to the point taken with the thin layer. In determining the energy variation of the cross section, a foil containing about 5 milligrams of uranium was used, whereas for determining an absolute value, a foil containing about 360 micrograms was used.

.93 mg/cm²

To determine the relative cross section as a function of neutron energy, 14 points were taken at energies between 400 and 1600 kev. Several different thicknesses of proton radiators were used since the variation of maximum proton energy over that range is considerable. In this determination of relative cross section the various backgrounds and corrections listed in the following section have little or no effect. For instance, the hydrogen contamination of the counter would have no effect so long as the counter pressure is undisturbed except for the fact that this background does not produce a flat pulse height distribution and so has a slight effect on the shape of the pulse height distribution with the radiator in place. Likewise, the weight of the proton radiator need not be accurately known since this is involved only in a small thickness correction which varies with neutron energy.

3.5 Corrections, Backgrounds and Errors for Determination of the Absolute Cross Section at 1.25 Mev

The effects listed below have been considered.

- (a) Proton recoils from hydrogen contamination of the counter gas and walls.
- (b) Fission counts from room-scattered neutrons, most of which


are of too low an energy to be detected by the proton recoil detector.

(c) Neutron scattering by the counter. These neutrons are relatively more likely to be detected by the fission detector than by the proton recoil detector because of the directional properties of the proton recoil detector.

(d) Scattering of neutrons by the platinum backing of the foils. The detectors are affected differently because of their directional properties.

(e) Target scattering. Neutrons scattered from the target surroundings in general have lower energies than the primary beam through the counter and so are relatively more effective in the fission counter than in the proton recoil counter.

(f) Effect of the center of mass motion of fission fragments on the fraction of fragments counted.

(g) Determination of the U^{235} on the fission foils.

(h) Absorption of fission fragments by the uranium.

(i) Measurement of the hydrogen content of glycerol tristearate radiators.

(j) Completely spurious counts in either counter.

Effect (a) is measured by inserting a blank platinum foil in place of the proton radiator and making an exposure to the neutron beam, recording the same information as on a regular run. This background is about 4% of the effect with a 400 microgram radiator.

The effect (b) of room-scattered neutrons was determined by using

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a cadmium covered U^{235} spiral counter shielded from the target by a shadow cone. The background was observed with a 25 cm tungsten cone and 24 and 34 cm paraffin cones backed with boron. For purposes of this experiment the three cones gave the same results. Measurements were made with the spiral counter 38 cm from the target, at zero and 90° to the proton beam, and at neutron energies of 500 kev, 750 kev, 1 Mev, and 1.5 Mev. The magnitude of the background did not change appreciably with neutron energy but was about 30% lower at 90° than at 0° . The background at 0° was taken to be $1.27 \pm .22\%$ of the counting rate for the spiral counter unshielded at 10-1/2 cm from the center of the target.

The effect (c) of counter scattering was observed by surrounding the counter with cylinders of cadmium and of iron in various combinations and observing the effect on the ratio of proton recoils to fissions. The measured effect of $1.35 \pm .36\%$ is in agreement with a crude estimate based on scattering cross sections.

The effect (d) of foil scattering arises because of the increased average path length of neutrons in the uranium or hydrogenous radiator. In computing this effect the angular distribution of elastic scattering for platinum was assumed to be similar to that measured by Journey for gold and mercury with fission neutrons and a U^{238} detector. The net effect on the determination of the fission cross section is taken to be $0.48 \pm 0.25\%$.

The effect (e) of target scattering was estimated by assuming an

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angular distribution for scattering from materials near the target. This effect was taken to be $0.18 \pm 0.08\%$ at 1-1/4 Mev.

In this experiment the effect (f) of the center of mass motion of the fission fragments is to increase slightly the fraction of fissions which are lost because of absorption of the fragments in the uranium. This is computed to be $0.58 \pm 0.10\%$.

One determination of the amount of U^{235} on the fission foils, effect (g), was made by Buddy Warren of Group J-11 at the time the foils were prepared. Measured amounts of a solution containing the uranium (99.85% U^{235}) in known concentration were electroplated onto the platinum foils. This process deposits all but about 1% of the uranium. A fresh foil is then attached to the plating cell and the remainder of the uranium is believed to be deposited on the second foil. This residue is then determined by fission counting in a slow neutron flux, against a known standard foil. It is believed that this procedure should give the amount of uranium to an accuracy of 1%. As a check on the self-consistency of the weights as supplied by J-11 the alpha counting rates of the four foils were measured. When small corrections for counting losses due to foil thickness were made, the ratios of counting rates differed from the ratios of weights by less than 1%. A check on the amount of material on the foils was also made by comparing the foils with thin standard foils of J-11 by fission counting in a slow neutron flux. After making the foil thickness correction (see next paragraph), the weight of uranium checked within $0.2 \pm 0.8\%$. To obtain a third

painted by John Povelites of CMR-4. The amount of uranium on these foils was determined by micro-balance weighing after conversion to the oxide by heat treatment. These foils were compared with the electroplated foils by fission counting and the results agreed within 1%.

Several foils were analysed for uranium content by CMR-1, using a colorimetric method, but the results were in poor agreement with the other determinations and the method was not considered reliable as applied to the electroplated foils. The difficulty may well have been caused by chemical impurities in the plated material. The assumed weight of uranium is believed to be within 1.5% of the correct value.

Effect (h) is absorption of fission fragments in the uranium. If the deposits were uniform and the foils flat, we could make this correction following Rossi and Staub,⁶ in which case the correction for the most used foil (63 micrograms per square centimeter) amounted to 0.74% at the biases used. We may also obtain an estimate of the effect by observing in detail the low pulse height part of the fission pulse height spectrum. When this was done, the low energy "tail" was found to have many more pulses than would be accounted for by the effect of uranium thickness. Also the fraction of the pulses which occurs in this tail seemed to bear little relation to foil thickness and is similar for all four foils (200 to 500 micrograms total weight). Observation of the α -particle pulses from these foils showed the same effect, even considering back scattering, and a weak Pu source on a similar

backing gave a similar pulse height distribution. It is assumed that the effect is caused by a non-uniform surface. Since the platinum must be thin to minimize interference with the neutron beam, it is not possible to obtain a perfectly smooth surface. As a result, some of the fission fragments (and α particles) strike the foil and produce smaller pulses than they should. Since all fragments leaving the uranium layer produce some ionization, we extrapolate the observed pulse height distribution to zero pulse height and then apply the correction for absorption in the uranium. The total correction, including the extrapolation, amounts to less than 2% and the uncertainty introduced into σ_f by this correction should be less than 1%.

The amount of glycerol tristearate on the proton radiators was determined by weighing the platinum foils before and after evaporation. The weights should be correct to about 1%. Since several different radiators were used, we obtained a check on the weights. Of five radiators used, one disagreed (in the fission cross section obtained) with the others by 10%. This result reproduced to better than 1% on a subsequent check and the assumption was made that the particular foil had been incorrectly weighed. The estimate of the standard error from the deviations from the average for the other four foils was 1.6%, whereas the standard error from statistics of fission counting alone is 1.0%. Probably the weight of these radiators can be trusted to 1%. The hydrogen content of the glycerol tristearate after evaporation, effect (i), was measured by A. R. Ronzio. Two samples of about 20



milligrams each were prepared in the same manner as the radiators and a measurement of the hydrogen content was made on these samples as well as on unevaporated glycerol tristearate. The results obtained for the evaporated samples were 12.49 and 12.43% hydrogen by weight compared to the theoretical value of 12.44%. The unevaporated sample contained 12.83%, probably indicating presence of volatile impurities which are lost in the evaporation process.

The effect (j) of completely spurious pulses was troublesome only in the fission counter where very low counting rates were obtained. Such troubles were usually caused by faulty electronic equipment and could be detected by leaving all detection equipment operating when the accelerator was not running. A routine check of this effect plus the effect of background neutrons from various sources along the accelerating tube and target tube was made by operating in the usual manner with the tritium in the target replaced by helium. Under these conditions the counting rates of both protons and fissions were usually reduced to about 1% of their values when the target was filled with tritium. These counts were due mostly to neutrons from tritium left somewhere in the target system, since the ratio of protons to fissions did not change greatly.

4. Results

The results for the absolute cross section at 1.25 Mev are listed in Table 1. If one were to consider the estimate of standard error from

Table 1

<u>Radiator</u>	<u>$\sigma_f(25)$, barns</u>
1	1.260
2	1.273
3	1.267
4	1.309
5	1.236
Average 1.269 barns	

the variations in cross section in Table 1 and the estimates of error on corrections listed in the preceding section as being truly statistical and assume that no systematic errors are involved, then the overall accuracy of the result could be characterized by a standard error of 2%. This is most optimistic and the author would prefer to guess that the value quoted is probably within 3-1/2% of the true cross section.

The energy variation of the cross section is shown in Fig. 4. The n,p scattering cross sections used at the various energies are given in Table 2. The points shown are normalized to 1.27 barns at 1.25 Mev. The solid curve drawn through these points is the cross section determined by Group P-9 using a long counter as a neutron flux monitor. The P-9 data are also normalized to 1.27 barns at 1.25 Mev. It is suggested that the most probable values of the fission cross section be taken from the solid curve, with estimates of error being given by the bars. The cross section is nearly the same as that determined by Hall, Koontz, and Rossi² in the region 1 to 1.6 Mev. Below 1 Mev this work gives considerably lower values than those of Hall, Koontz, and Rossi.²

Table 2

<u>E_n,</u> <u>kev</u>	<u>σ_H,</u> <u>barns</u>
1620	3.28
1545	3.37
1424	3.52
1272	3.75
1171	3.93
1095	4.08
1025	4.21
944	4.40
965	4.61
770	4.93
673	5.30
562	5.78
513	6.10
403	6.95

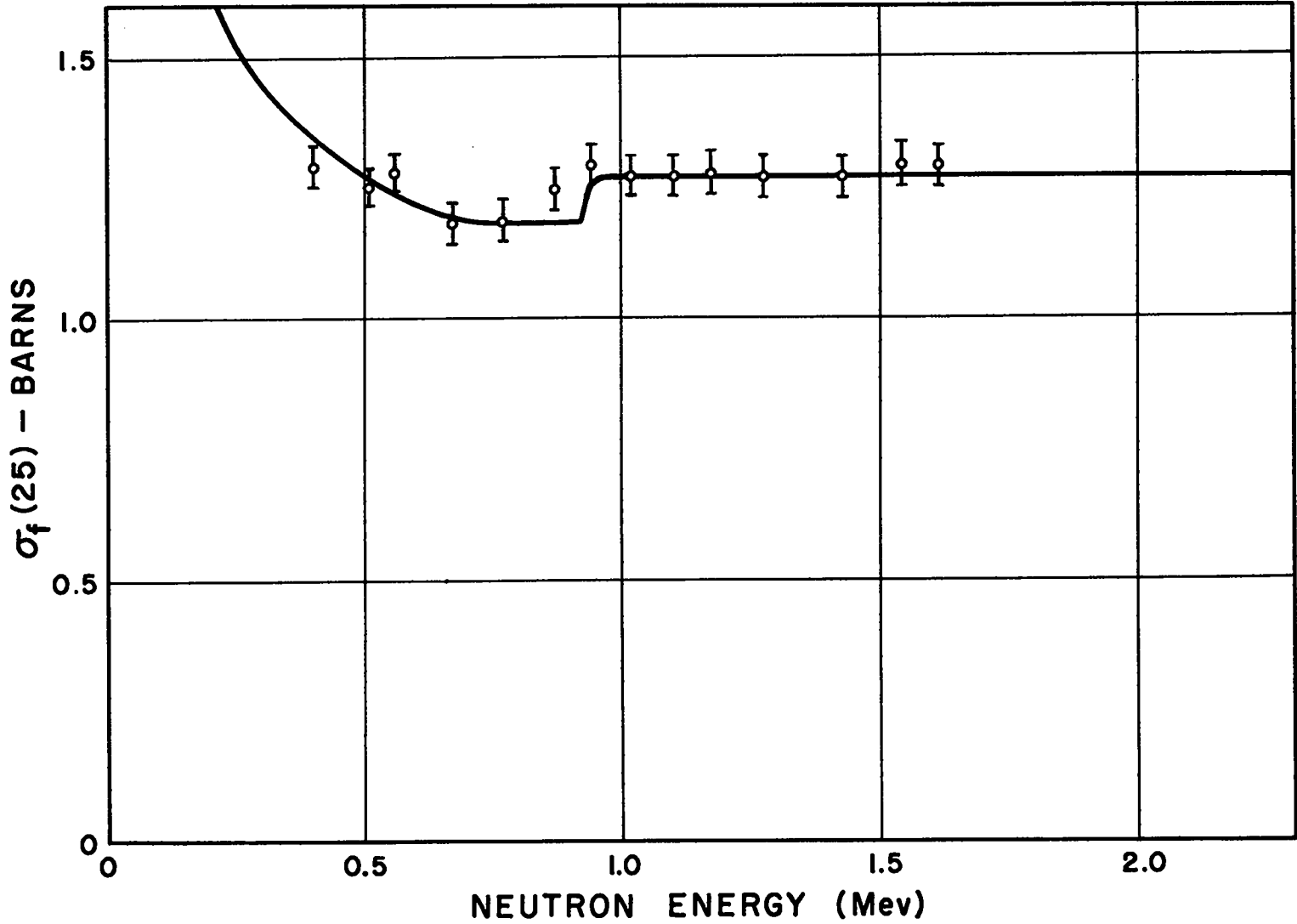


Fig. 4-- U^{235} fission cross section.



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