

11-491  
0004

DECLASSIFIED

LOS ALAMOS NATIONAL LABORATORY

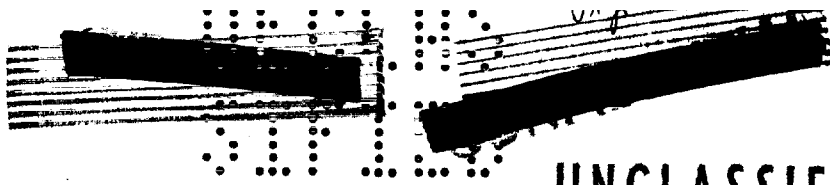


3 9338 00350 2340

DECLASSIFIED

01100  
01100

01100  
01100



UNCLASSIFIED

LA - 491



November 14, 1946

This document contains 20 pages

AVERAGE NUMBER OF NEUTRONS EMITTED

PER SPONTANEOUS FISSION BY Pu<sup>240</sup>

PUBLICLY RELEASABLE

Per Plutonium FSS-16 Date: 3-22-96  
By Kolar . CIC-14 Date: 4-17-96

Work Done By:

Emilio Segré  
Clyde E. Wiegand

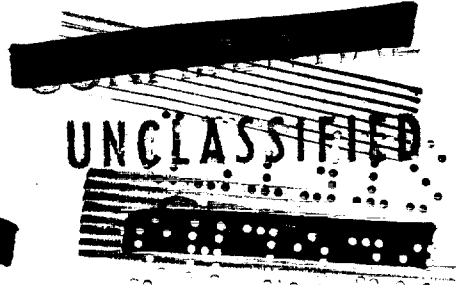
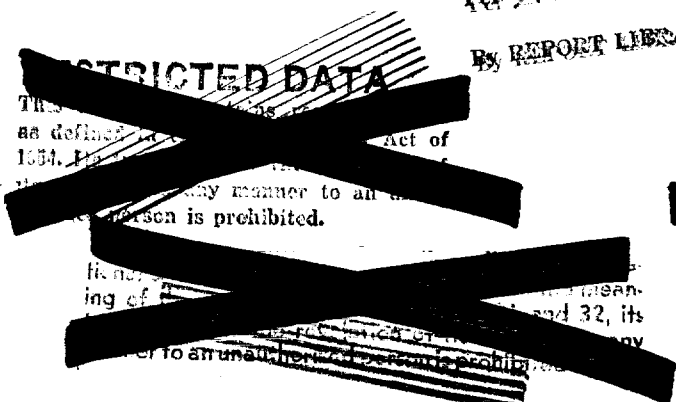
Report Written By:

Emilio Segré  
Clyde E. Wiegand

Classification changed to UNCLASSIFIED  
by authority of the U. S. Atomic Energy Commission  
Per Jack W. Kalin List # 9  
By R. M. Martin 4-26-57  
REPORT LIBRARY 5-2-57

Classification changed to CONFIDENTIAL  
by authority of the U. S. Atomic Commission  
Per ACCT 1108  
By R. M. Martin 11-21-56  
REPORT LIBRARY

NOV 17 1996



  
UNCLASSIFIED

## SUMMARY

The average number of neutrons emitted per spontaneous fission by  $\text{Pu}^{240}$  was measured in two ways. The first was by a coincidence system in which the neutrons emitted in coincidence with individually counted fissions were detected. The second method was to determine the rate at which neutrons were being emitted from a mass of plutonium, the rate of spontaneous fission in the material having been previously determined. The result is  $2.37 \pm 0.3$  neutrons/spontaneous fission.

UNCLASSIFIED  
  


UNCLASSIFIED

## 1. Introduction

( The value of  $\nu$ , the average number of neutrons emitted per fission, is well known in the case of slow-neutron fission of  $\text{Pu}^{239}$ .<sup>1</sup> It is also of considerable interest to know whether  $\nu$  depends on the energy of the neutrons producing fission.) This problem can be attacked by varying the energy of the neutrons used in the bombardment. In such a case the excitation of the compound nucleus undergoing fission is

$$E_n + T = E$$

where  $E_n$  is the binding energy of the neutron and  $T$  its kinetic energy.  $E_n$  is of the order of 5 Mev and  $T$  has been varied by a few hundred KeV. No change in  $\nu$  has been observed.

The effect of a more drastic change in the energy of excitation can be observed by comparing  $\nu$  for spontaneous fission of  $\text{Pu}^{240}$  and  $\nu$  for slow-neutron fission of  $\text{Pu}^{239}$ . The fissioning nucleus is obviously the same, but in the case of spontaneous fission the energy of excitation is 0; in the case of slow-neutron fission  $E_n$  or about 5 Mev.

The high spontaneous fission rate of  $\text{Pu}^{240}$  makes this experiment possible and the result is that

$$\nu_{sp} = 2.37 \pm 0.3, \quad \text{whereas } \nu_{slow} = 2.95 \pm 0.05.$$

Hence it seems probable that there is a small difference in  $\nu$ , in the sense that  $\nu$  for the excited nucleus is larger than  $\nu$  for the fundamental state. "

---

1. Snyder and Williams LA 102

UNCLASSIFIED

U  
L  
L  
S  
S  
I  
F  
I  
E  
D

UNCLASSIFIED

## 2. Coincidence Method

In this experiment a fission chamber was placed inside a sensitive neutron detector and the coincidences were observed between fissions and neutron counts. Then the number of neutrons per fission,  $\gamma$  is given by

$$\gamma = C/(f \cdot e)$$

where  $C$  is the number of coincidence counts,  $f$  the number of fissions occurring in the fission chamber, and  $e$  the efficiency for detection of a fission neutron. This equation holds if  $\gamma e \ll 1$ . This condition was amply satisfied in these experiments.

The main reason for using a coincidence system is that one can minimize the effect of the background of the neutron counter.

In our experimental conditions we have a background on the neutron counter of about 0.33 c/sec. The number of spontaneous fission, occurring in the samples used in these experiments is about 0.013 per second; the number of neutrons emitted per fission is about 2.4 and the counting rate due these neutrons is about  $8 \cdot 10^{-4} \text{ sec}^{-1}$ . It is clear that without a coincidence system the experiment would be impossible.

The background due to accidental coincidences is very small: if the gate opened by a fission count stays open 650  $\mu\text{sec}$ , as in our experiments the probability that an opening of the gate be accompanied by an accidental coincidence is

$$6.50 \times 10^{-6} \times 0.33 = 2.15 \times 10^{-4}$$

UNCLASSIFIED

SECRET  
CONFIDENTIAL

We shall see that the probability that an opening of the gate is accompanied by coincidence due to the emission of  $\gamma$  neutrons on fission is of the order of  $6 \times 10^{-2}$ . From these numbers it is seen that the background is about  $1/300$  of the effect we want to measure.

The sensitive neutron detector in our apparatus consisted of a cylindrical chamber filled with boron trifluoride, the boron of the compound being enriched with  $B^{10}$ .<sup>1</sup> A schematic diagram of the neutron detector is shown in Fig. 1. The collecting electrode, which is a hollow cylinder coaxial with the chamber body and suitably insulated, forms the sensitive cavity into which the fission chamber was placed.

In order to have a sufficiently large rate of fission it is necessary to devise a chamber into which a considerable amount of the highly alpha-active plutonium may be put without compromising a well-determined detection of fission pulses. Accordingly a small cylindrical fission chamber was designed which would fit inside the collecting electrode of the neutron-detecting chamber described above. The construction of the fission chamber is shown schematically in Fig. 2. It was necessary to make the chamber as small as conveniently possible in order that the capacity between this chamber and the collecting electrode of the boron trifluoride chamber be kept at a minimum since any capacity at this point is in parallel with the output of the neutron-detecting chamber and reduces its pulse size accordingly. The chamber was filled with continuously purified argon.

The plutonium in which the fissions occurred was deposited on four 0.005-inch-thick platinum sheets of the same size as the brass strips

---

1. LA 108

SECRET  
CONFIDENTIAL

6.

which were in contact with the chamber body and acted as the high-voltage electrodes. 20 mg of Pu containing about 0.8 mg of  $\text{Pu}^{240}$  were used. The surface covered in each foil was about  $35 \text{ cm}^2$ . The platinum sheets were attached to the brass strips and in turn were covered with collimating screens (a detail of which is shown in Fig. 2).

It is advantageous to cover the plutonium deposit with a collimating screen in order to minimize the effect of the fluctuating background of alpha particle ionization. The action of the collimating screen is a rather complex one, but it is easy to see qualitatively that it will lower the fluctuations in a shallow chamber that operates on electron collection, as in our case. Namely, the smallest pulses obtainable from fission fragments are unaffected by the collimating grid; on the other hand the largest alpha pulses, that would be produced by alphas going almost parallel to the source, are suppressed.

Without entering in a detailed discussion of this and other effects, direct experiment shows that the allowable effective amount of plutonium may be increased considerably by the use of collimating screens.

Fig. 3 shows the operation of our fission chamber. The data are obtained by putting the fission chamber in a constant neutron flux. The number of fissions per minute is plotted versus effective amplifier sensitivity with the argon gas pressure and collection voltages noted on the graph. The operating conditions during our experiment were at bias 7. At this bias it is clear that one will not register any spurious fission, simulated by alpha fluctuations. The efficiency for counting fission is perhaps 0.7, but this fact is irrelevant to our experiment.

An over-all block diagram of the experimental set-up is shown in Fig. 4. The collecting electrode of the fission chamber was connected



7.

to the input of a linear amplifier with a time of rise of about 0.1 microsecond and an effective resistance-capacitance decay time of about 0.1 microsecond. Although pulse height was sacrificed with this value of the decay time constant, it was proved experimentally that it offered greater differentiation between fission pulses and alpha particle ionization and gave a suitable plateau curve as shown in Fig. 3. The output of the fission pulse amplifier was connected to a scaling and register circuit. The discriminator of the scaling circuit was arranged to feed a pulse to a coincidence circuit each time a fission count was registered. This pulse held the coincidence circuit open by means of a multivibrator for a measured time of 650 microseconds.

The neutron-detecting boron trifluoride chamber, in which electrons were also collected, was connected to a linear amplifier with a time of rise of 0.2 microseconds and a decay time of 20 microseconds. The output of this amplifier was connected to a scaling circuit the discriminator of which was also arranged to feed a pulse to the coincidence circuit each time a neutron was detected. If the pulse from the neutron detector arrived at the coincidence circuit within 650 microseconds from the time a fission was detected, the coincidence circuit fed a pulse to still another scaling and register unit. The reasons for holding the coincidence circuit open for a considerable time after the occurrence of a fission will be discussed later.

In addition to the three ordinary mechanical registers, a recording millimeter with an electronic driving device was arranged to record on a clock-driven chart the time of each coincidence count. Using this arrangement the random distribution of coincidences with respect to time could be verified.

8.

A tabulation of the experimental data taken is given in Table I.

TABLE I

fission counts, f	coincidence counts, C	measured efficiency, e	$\frac{C}{e}$
270	24	0.0268	895
504	25	0.0264	947
607	31	0.0275	1127
461	36	0.0285	1263
640	45	0.0249	1807
502	28	0.0242	1157
498	25	0.0246	935
670	34	0.0255	1333
672	36	0.0259	1389
842	56	0.0244	2295
128	10	0.0252	397
684	39	0.0250	1696
<u>6478</u>			<u>15241</u>

Column 1 gives the number of fissions registered in a certain time interval; column 2 gives the number of coincidences registered in the same time interval; column 3 gives the efficiency of the neutron detector measured in the manner described in Section 3; column 4 is the ratio of the numbers in column 2 and column 3.

The data were taken over a period of seven days with an operating time of 134 hours.

The number of neutrons emitted on the average in connection with spontaneous fission is given by

$$\bar{\nu} = \frac{\sum C/e}{\sum f} = \frac{15241}{6478} = 2.35$$

This value is subject to the corrections and probable errors discussed in the Section 3.

### 3. Discussion of the Experiment

The main difficulty of this experiment is the determination of the actual efficiency of the boron trifluoride chamber for the detection of a neutron emitted in connection with spontaneous fission. This difficulty arises primarily from the difference in energy spectra of the spontaneous neutrons and the standardizing sources. Experiments were made to determine the efficiency using as primary standard a cylindrical mass of natural metallic uranium and a radium plus beryllium source as auxiliary standard.

The auxiliary standard was used to verify the constancy of the efficiency of our apparatus during the runs. The maximum change of the efficiency observed was 10% of its normal value and these drifts were noted and taken into account in Table I.

The primary standard was used to measure the absolute efficiency of our neutron counter. A uranium cylinder 8.7 cm long, 3.7 cm in diameter and weighing 1789 gr was used. It was assumed to emit 26.8 neutrons/sec corresponding to 15  $\nu$ /kgr sec. We also assume that the efficiency of our counter for spontaneous fission neutrons emitted by the two substances  $U^{238}$  and  $Pu^{240}$  is the same.

The determination of the efficiency was made by determining  
a. the counting rate  $C_S$  (13804 c/m) of the auxiliary Ra + Be source in a dummy chamber identical to our fission chamber; b. the counting rate  $C_2$  (18020 c/m) of the same source with the dummy chamber replaced by our uranium standard. The Ra + Be source was on the center of the uranium cylinder; c. The counting rate  $C_U$  (56.7 c/m) of the uranium cylinder above.

10.

The efficiency  $\epsilon$  is given by

$$\epsilon = \frac{C_1}{N_1} \frac{C_3}{C_2}$$

Under the identical experimental conditions we determined also the counting rate  $C_5$  of our auxiliary Ra + Be standard in a conveniently reproducible position near the real fission chamber. During the runs we checked periodically this last counting rate and we assumed that the efficiency was proportional to it.

$$\epsilon = \frac{C_5}{5.04 \times 10^5}$$

Other uncertainties entering into the experiment are: statistics in the number of coincidences counted,  $C$ ; the absolute number of neutrons emitted per unit time by the uranium block used as a standard; systematic error arising because  $\nu$  is greater than one; and an error brought in by the fact that the coincidence circuit did not stay sensitive after the neutrons are emitted for a time, infinitely long compared with the life time of slow neutrons in paraffin.

The determination of  $C$  is based on 387 counts and is accordingly uncertain by 5 percent. The number of neutrons emitted per unit time by the uranium cylinder is thought to be accurate to 10 percent. A correction can be made for  $\nu$  being greater than one as follows:

Let  $\epsilon$  be the efficiency for counting one neutron, as measured by a standard source.

Let  $P_m$  be the probability of getting one count when  $m$  neutrons are emitted within one resolving time of the detector.

for:

$$m = 1$$

$$P_1 = e$$

$$m = 2$$

$$P_2 = 2e - e^2$$

$$m = 3$$

$$P_3 = 3e - 3e^2 + e^3$$

Neglecting  $e^3$  since  $e$  is a small number, the actual efficiency for counting

$$2 \text{ neutrons is } (2e - e^2)/2$$

$$3 \text{ neutrons is } (3e - 3e^2)/3$$

and since we know  $\gamma$  is between 2 and 3 we average the above efficiency and get for  $e'$ , the efficiency for counting neutrons simultaneously emitted in groups of 2 or 3 such that 2 is 2.5.

$$e' = e - 3/4 e^2$$

The average efficiency  $e$  from Table I is 0.0256. Substituting this value to get  $e'$  we obtain 0.0251.  $\gamma$  must be multiplied by 0.0256/0.0251 or 1.02. This correction is obviously quite small compared with other uncertainties in the experiment.

The correction needed for the coincidence circuit being open a finite time can be estimated from a knowledge of the half life of thermal neutrons in paraffin. This was measured by Manley<sup>1</sup> who found the mean life to be 205 microseconds in an infinite block of paraffin.

The time the coincidence circuit was sensitive, was 650 microseconds or 3.15 mean lives. The correction factor by which one has to multiply the actual number of observed coincidence counts to obtain the number of counts which would be obtained with a gate of infinite length is

$$\frac{1}{1 - e^{-3.15}} \quad \text{or} \quad 1.04.$$

---

1. Physical Review 61, 152 (1942)

12.

This factor is lowered appreciably by the presence of the boron trifluoride chamber which acts as an absorber. We have used 1.02.

From Table I the crude value of  $\nu$  is 2.35. The correction factor due to the effect on the efficiency of being greater than one is 1.02. The correction factor for the coincidence circuit staying open a finite time is also 1.02. This gives  $\nu = 2.44$ .

The error of this value is due to errors in determination of the efficiency, assuming that the standard is known exactly, probably 5%; errors in the neutron standard (uranium) about 10%; error due to counting statistics 5%; All together this amounts to less than 13 percent so it is concluded that this experiment gives a value for  $\nu$  of  $2.44 \pm 0.3$ .

  
 4. Direct Counting Method

In the second series of experiments the number of neutrons emitted per unit time from a mass of plutonium was compared with the number of neutrons emitted per unit time from a standard radium plus beryllium source and a standard mock-fission source. These sources were calibrated by an integral method and by comparison with primary standards. They are known on the hypothesis that source #43 emits  $* \text{ n/sec}^1$ . If this standard value should be changed by future redeterminations, our values of  $\gamma$  would change in proportion, but the ratio  $\gamma_{sp}/\gamma_{slow}$  would not be affected because also  $\gamma_{slow}$  is determined with reference to this standard.

The number of spontaneous fissions occurring per unit time in the plutonium was determined by counting the number of fissions occurring in a small sample of the same material. This was done by counting the fissions from a thin foil in a regular spontaneous fission analysis as described by Farwell<sup>2</sup>.

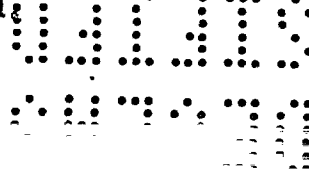
The plutonium used in this experiment was in the form of a cylinder 1.75 cm in diameter averaging 0.65 cm thick and weighing 29.85 grams. It gave 7.44 f/g sec = 5 percent.

Comparisons with the standard sources were made by using the sensitive neutron detector described in the first experiment and by the long boron trifluoride proportional counter designed by Hanson<sup>3</sup>.

In using the sensitive neutron detector for these comparisons the sources were not placed in the center of the cavity as usual but were placed

1. See LA 400
2. See LA 490
3. LAMS 66, A. O. Hanson, Flat-Response Neutron Detector.

\* The source strength is not given in the manuscript. LA 400 gives the emission of source 43 as  $8.95 \times 10^6 \text{ n/sec}$ , with an accuracy of one percent. Ed:



out in the paraffin block at a distance of 12 cm from the axis of the chamber. This was done because measurements in the paraffin block of sources emitting different energy neutrons showed that at this distance into the paraffin from the chamber, the efficiency was almost independent of the neutron energy.

The long boron counter was designed to have an efficiency independent of the energy of the incident neutrons. However, for the high-energy neutrons in radium-plus-beryllium sources, the efficiency of the long counter falls off considerably being as much as 10 percent less efficient for radium-plus-beryllium neutrons as for intermediate energy neutrons.

The standard mock-fission source was designed to have a neutron energy spectrum similar to the energy spectrum of slow-neutron-induced fission neutrons but this is an approximation only and the energy spectrum of spontaneous neutrons may be slightly different from that of induced fission neutrons. In our actual experiments we found that the ratio of the mock-fission source strength to the Ra Be source strength measured by the long boron counter was 1.07 times the same ratio as determined by the integral calibrating experiments. This means that the efficiency of the long B counter for mock-fission neutrons is 1.07 times the efficiency for Ra + Be neutrons.

Table II shows in columns 2 and 3 the ratios of the counting rates of the standard sources to the counting rates of the plutonium button obtained from the two detectors.

TABLE II

	long counter	BF <sub>3</sub> chamber	corrected long counter	corrected BF <sub>3</sub> chamber	long ctr n/sec	BF <sub>3</sub> chamber n/sec
*MF/Pu	60.2 ± 2	61.9 ± 1	63.6 ± 2	72.3 ± 1.0	541 ± 60	472 ± 47
Ra-Be/Pu	13.2 ± 0.5	12.3 ± 0.2	15.0 ± 0.5	14.3 ± 0.2	551 ± 61	578 ± 58

\*MF denotes mock-fission source



: : : :  
: : . . : : : :  
: : : : : : : :  
: : : : : : : :  
15.

These crude ratios must be corrected because the plutonium button multiplies the spontaneous neutrons due to fast-neutron fission within itself. The multiplication was estimated to be 1.06 by the theoretical group. If the plutonium button is embedded in paraffin as it is the case when the sensitive neutron counter was used multiplication is increased due to slow neutron fission. An estimate of the multiplication factor under our experimental conditions used for the BF<sub>3</sub> chamber is 1.16.

The corrections are then made as follows:

MF/Pu	=	13.2	x	1.06	long counter comparison
Ra + Be/Pu	=	13.2	x	1.06	x 1.07 " " "
MF/Pu	=	61.9	x	1.06	x 1.10 BF <sub>3</sub> chamber comparison
Ra + Be/Pu	=	12.3	x	1.06	x 1.10 " " "

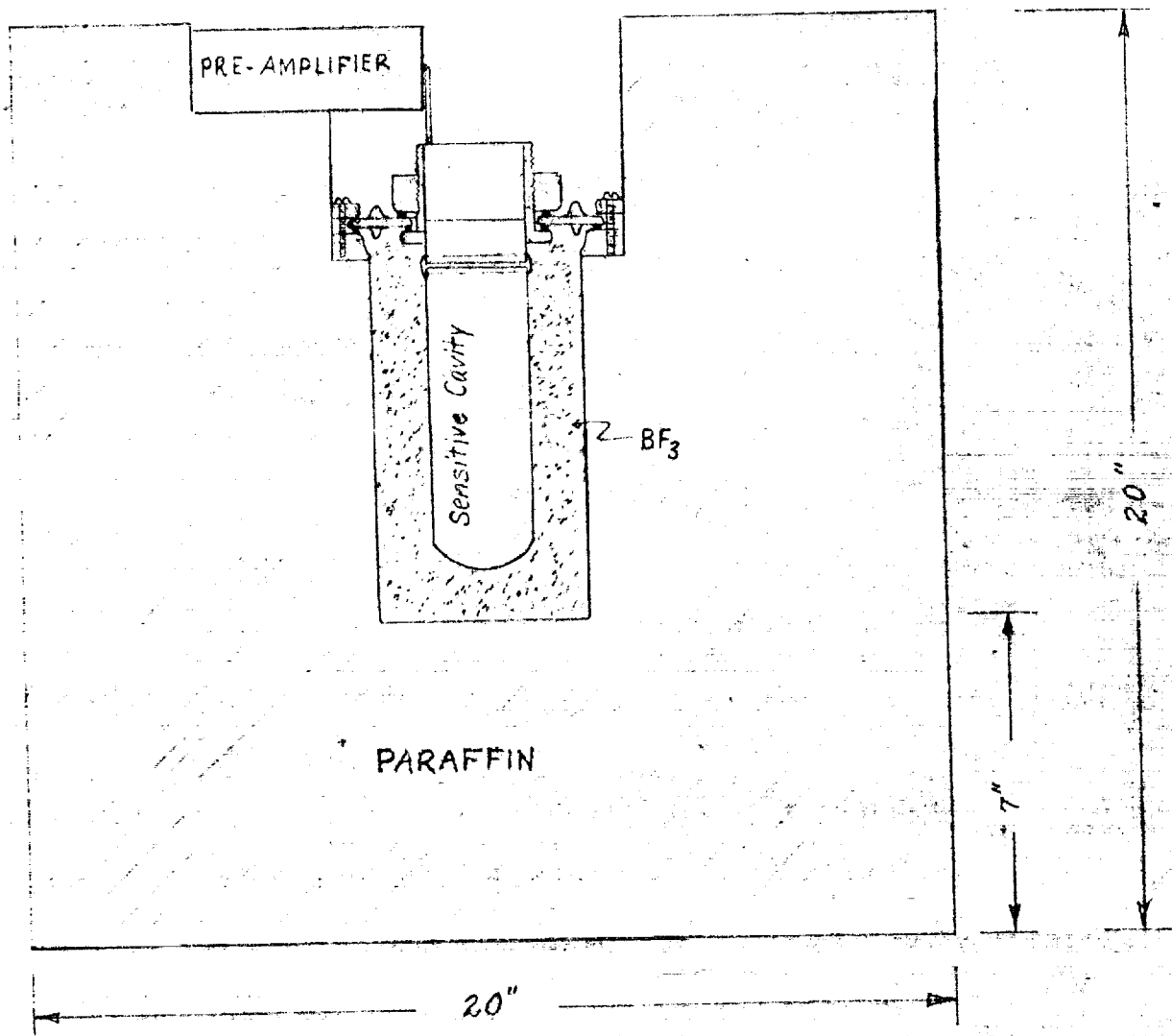
In addition another correction must be made because of the different efficiency of the long boron counter for fission and Ra + Be neutrons discussed above. With these additional corrections we obtain the numbers in columns 4 and 5. Columns 5 and 6 give the number of neutrons emitted per second by the button, according to the various systems of measurement and calibrations.

The numbers reported in the first 5 columns of Table II include the uncertainties due to counting statistics; columns 6 and 7 contain also the uncertainties in the absolute emission of the standard sources. The measured average number of neutrons emitted by the plutonium is 536  $\pm$  30 per second or dividing by the weight of the button 18.0  $\pm$  1.0 n/gr sec. In addition to neutrons emitted by spontaneous fission, some may be generated by alpha particles impinging on light-element impurities in the button. From chemical analysis of several lots of plutonium purified chemically in the same way as the sample used in this experiment, it appears

U. S. A.  
 O. R. I.  
 16.

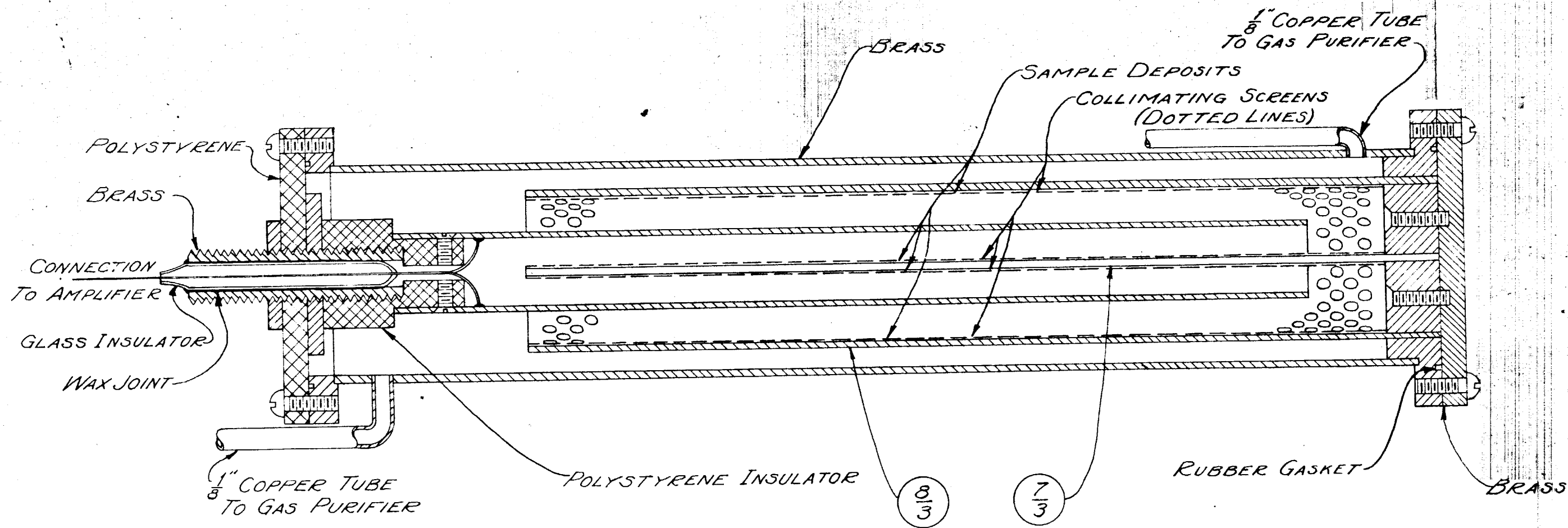
that about one neutron per gram second may be due to alpha, n reactions. This would mean that 17.0 neutrons per gr second were due to spontaneous fission. Dividing this number by the number of fissions per gr second, 7.44, we obtain a value for  $\nu$  of  $2.3 \pm 0.3$ .

03105  
 000000

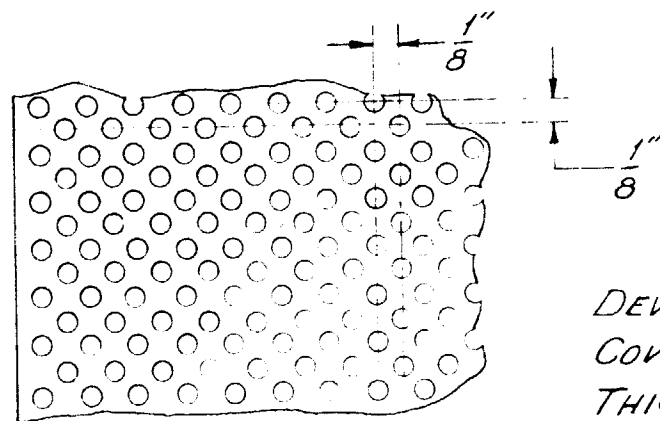


BORON TRIFLUORIDE NEUTRON DETECTOR

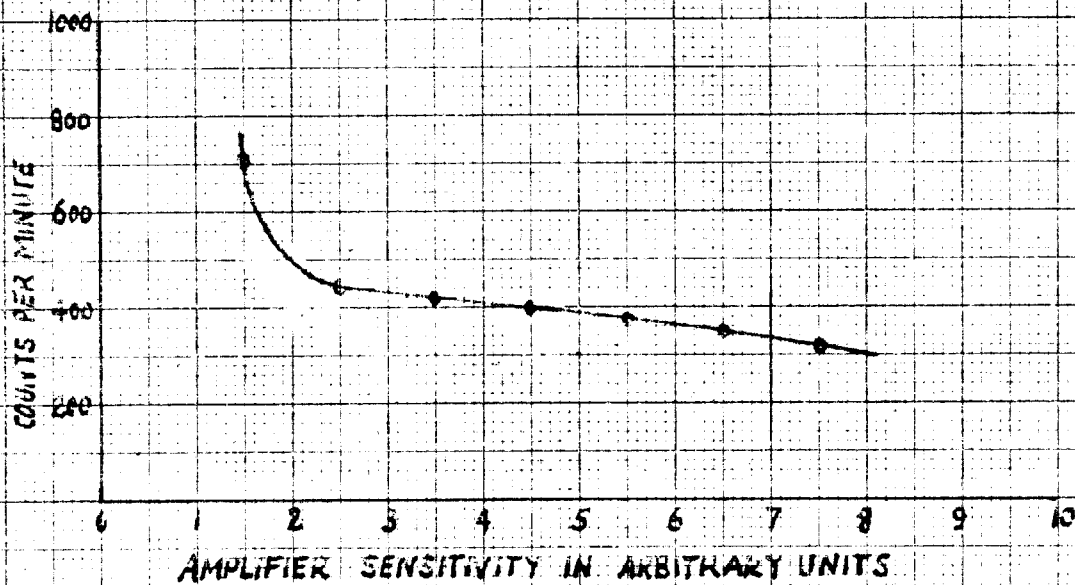
FIG. 1



**FIG. 2**  
**FISSION CHAMBER**  
 SCALE 2"=1"

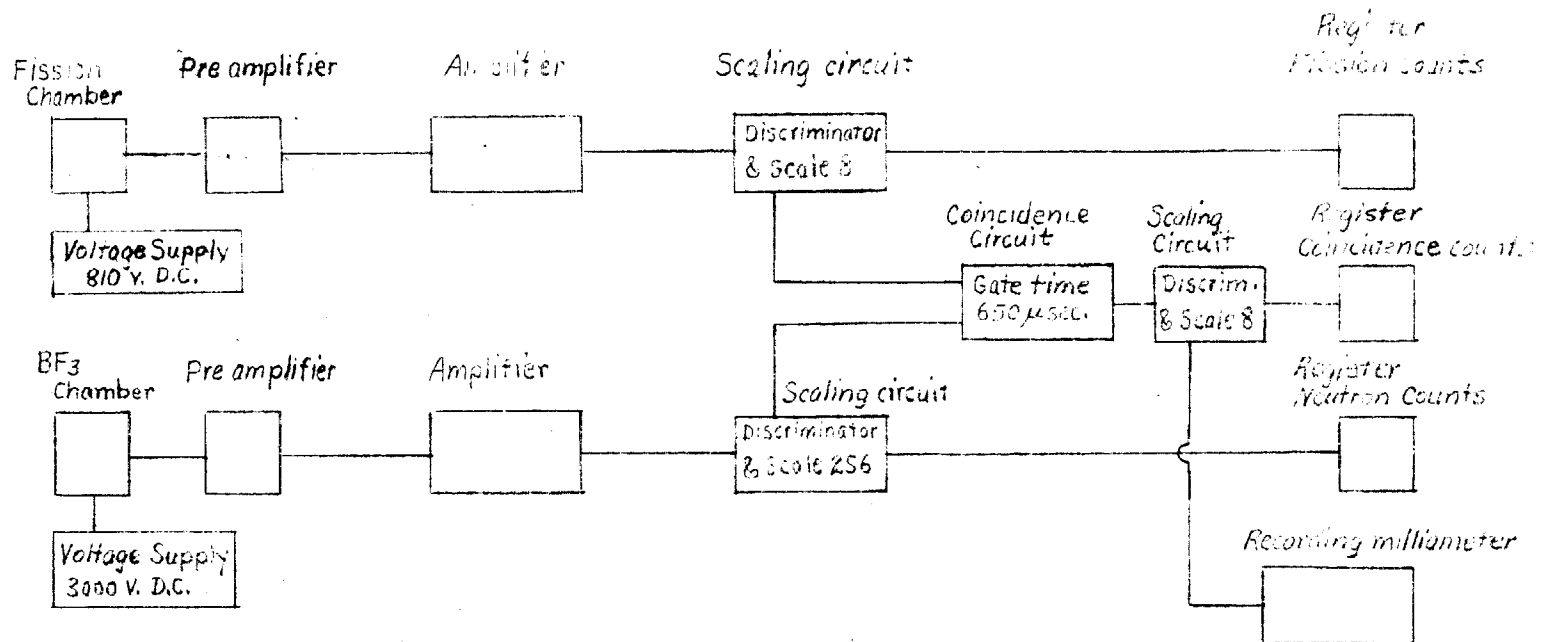


DEVELOPED SECTION OF COLLIMATING SCREENS WHICH COVER PARTS (8/3) AND BOTH SIDES OF (7/3) AS SHOWN ABOVE. THICKNESS OF SCREENS .025 INCHES. DIAMETER OF HOLES .111 INCHES (#34 DRILL) MATERIAL: BRASS



**FIG. 3**  
**FISSION CHAMBER BIAS CURVE**

COLLECTION POTENTIAL 810 VOLTS  
 CHAMBER FILLED WITH 3+ ATMOS. ARGON  
 PURIFIER CALCIUM METAL AT 170° C.



BLOCK DIAGRAM OF ELECTRONIC APPARATUS  
COINCIDENCE METHOD

FIG. 4

DECLAS  
SIFIED

DECLAS  
SIFIED

SECRET  
REF ID: A66342

SECRET  
REF ID: A66342