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FURTHER INVESTIGATION OF SPONTANEOUS FISSION IN PLUTONIUM

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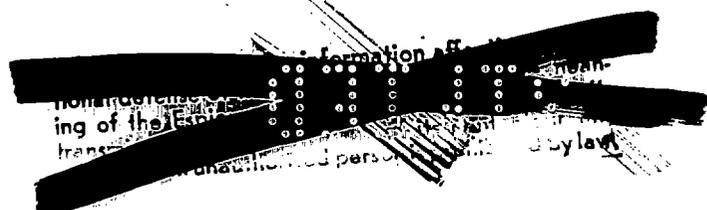
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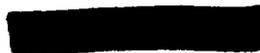
ABSTRACT

49 - 60  $\frac{1}{9}$  hr -

The spontaneous fission decay rates of Pu<sup>238</sup> and Pu<sup>240</sup> are  $7.7 \times 10^6$  and  $1.64 \times 10^6$  fissions per gram hour, respectively. Comparison of spontaneous fission analyses of a number of samples of pile-produced plutonium with Hanford data on the plutonium concentration in the uranium from which they were extracted shows that, in pile material,

$$\frac{\text{Pu}^{240}}{\text{Pu}^{239}} = \frac{70.9}{71} \times \frac{\text{Pu}^{239}}{\text{U}^{238}}$$

The isotope Pu<sup>240</sup> is almost entirely responsible for the spontaneous fission activity observed in pile-produced plutonium; other isotopes of plutonium and isotopes of elements 95 and 96 make no appreciable contribution.



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## FURTHER INVESTIGATION OF SPONTANEOUS FISSION IN PLUTONIUM

I. Introductory; Results of Early Work

One of the requirements of a material which is to be used in an atomic bomb assembly is that it emit few neutrons or none, since stray neutrons could initiate a premature chain reaction which would cause partial or complete failure of the assembly as a bomb. For this reason a study of the spontaneous fission rate of plutonium was undertaken at Los Alamos in order to estimate the spontaneous neutron emission which would almost certainly accompany spontaneous fission in the plutonium to be used.

Results obtained in the early investigations at Los Alamos (1943 and 1944) were reported in September, 1944.<sup>1</sup> Briefly, they were as follows:

Several samples of plutonium which had received successively greater neutron irradiations showed successively higher spontaneous fission activity. It was demonstrated with practical certainty that the activity was due almost entirely to the isotope  $\text{Pu}^{240}$ , formed by an  $(n, \gamma)$  reaction on  $\text{Pu}^{239}$ . Approximate spontaneous fission decay rates of 4.0 and  $1.6 \times 10^6$  fissions per gram hour were assigned to  $\text{Pu}^{239}$  and  $\text{Pu}^{240}$ , respectively.

Several points were considered with further investigation:

(1)  $\text{Pu}^{240}$  might be identifiable by means of mass-spectrographic analysis of plutonium samples.

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<sup>1</sup> Chamberlain, G. Farwell, E. Segre, LAMS-131, 1944.

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(2) If  $\text{Pu}^{240}$  were the high-fission material, further study of samples of pile-produced plutonium should verify the earlier observation that the specific spontaneous fission of plutonium material is proportional to the total neutron irradiation it has received, and thus proportional to the ratio  $\text{Pu}^{239}/\text{U}$  in the material "pushed" from the pile; i.e., that the highly fissioning substance is formed by a second-order neutron reaction starting from  $\text{U}^{238}$ .

(3) A determination of the abundance of  $\text{Pu}^{238}$  in pile plutonium should be made and the spontaneous fission rate of  $\text{Pu}^{238}$  determined if possible in order to estimate the maximum contribution of  $\text{Pu}^{238}$  to the observed spontaneous fission.

(4) Spontaneous fission observations should be made on any higher isotopes of plutonium or isotopes of elements 95 and 96 which might be identified in pile products. Although of considerable interest, such observations would be unnecessary in the positive identification of  $\text{Pu}^{240}$  as the high-spontaneous-fission material if investigation of points (1), (2), and (3) above gave the expected results.

## II. Discussion of Experimental Techniques

### 1. Description of General Method

Spontaneous fission in plutonium was studied by observing fission pulses from samples deposited as thin films on platinum discs and placed in nitrogen or argon-filled parallel-plate ionization chambers. Electron collection was used in either gas.

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\*Most of the foils were prepared by M. Miller and D. Hufford of Group CM-4.

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Battery-operated linear amplifiers were used to amplify the ionization pulses for recording by Brower impulse meters. Esterline-Angus recording milliammeters, fed through pulse-lengthening circuits, afforded periodic checks on all chamber-amplifier units. Complete battery operation was necessary in order to eliminate electrical disturbances. All units were shielded during actual operation by completely closed sheet metal boxes; and an isolated location was chosen for the laboratory site. Boron shielding was used to eliminate fissions induced by cosmic-ray slow neutrons.<sup>2</sup>

Fig. 1 shows one of the shielded booths containing two chamber-amplifier units. The sheet metal cover has been raised. Identifiable in the photograph are: (1) chamber, (2) amplifier chassis, (3) B batteries and bias supply, (4) outlet for 6 volt filament supply from bus along wall, (5) battery high-tension supply, (6) high-tension filter and switch, (7) Esterline-Angus recording milliammeter, (8) pulse-lengthening unit for Esterline-Angus, (9)  $B_2O_3$  shields, (10) Brower impulse counter. Covers of the adjoining booths are seen in closed position. The filling system and the auxiliary counting equipment used for slow-neutron-fission weighing of plutonium samples were mounted on rolling tables and could be moved from booth to booth.

## 2. Use of Fission and Alpha Activity Bias Curves

One of the most difficult problems in the study of spontaneous fission in a strong alpha emitter such as plutonium is that of recording with good efficiency

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See Segre, Chapter on Spontaneous Fission, Los Alamos Technical Series

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a sample of reasonable size while making certain that no spurious counts due to superposition of a number of alpha pulses are recorded.

Fission pulses are from ten to twenty times as large as single alpha pulses. If, however, the resolving time of the chamber-amplifier system is long compared to the average time interval between alpha activity background may simulate fission pulses and be recorded as such. It is this consideration which makes necessary electron collection with its high velocity and amplifiers with rise times and decay times short enough to take advantage of the fast collection. (The rise time of the amplifiers used with the argon chambers was about 0.2 microseconds; the electron collection time was of the order of a microsecond.)

A "fission bias curve" is a curve of fission counting rate, from a sample of fissionable material placed in a constant neutron flux, plotted against the bias of the amplifier discriminator. "30 millivolts bias" indicates a discriminator setting such that a 30-millivolt pulse from a standard pulse generator, impressed on the input of the first amplifier stage, is just large enough to be registered by the counting circuit and register. (In the argon chambers, the largest fission pulses observed corresponded to about 60 millivolts.) Fig. 2 shows a fission bias curve taken with a plutonium sample of 0.434 mg and a Ra+Be neutron source.

An "alpha activity bias curve" is a curve of counting rate due to superposition of alpha pulses plotted against the bias of the amplifier discriminator. Fig. 3 shows such a curve, taken with the same sample used for the fission bias curve. Plotted on semi-log paper, such curves were found to be straightlines over a range in counting rate of four or five powers of ten. Straight-line extrapolation of the curves to biases corresponding to  $10^{-4}$  or  $10^{-5}$  counts per minute was found not to be over-optimistic in determining "safe" operating biases for fission counting, i.e., biases at which no spurious counts due to alpha activity would be registered in weeks of operation.

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This was established by running polonium samples (of alpha activity equal to that of the strongest plutonium samples studied) at biases corresponding to  $10^{-5}$  or  $10^{-4}$  counts per minute; no counts were observed over periods of many weeks.

Extrapolation of the curve of Fig. 3 to bias of 30 millivolts indicates a counting rate of about  $10^{-6}$  counts per minute, so that one might expect at 30 millivolts to have a spurious count no more often than once in two years. (Such low spurious counting rates were necessary for some samples which fissioned extremely rarely.) Reference to the fission curve of Fig. 2 (taken for the same sample) shows that 30 millivolts is a very satisfactory operating bias for fission counting, from the standpoint of counting efficiency. These methods were used to determine the maximum alpha activity which could be tolerated in a given chamber at a bias corresponding to good fission-counting efficiency.

Some attempts were made to construct a theory which would explain quantitatively the characteristics of the alpha activity curves, but none was satisfactorily completed.

### 3. Nitrogen Chambers; use of Collimating Screens

Nitrogen chambers, similar to the argon chamber shown in Fig. 8, were used in the early spontaneous fission work on plutonium at Los Alamos.<sup>1</sup> The associated amplifiers had a rise time of about one microsecond. The resolution of these units was sufficient to permit observation of 20-microgram samples (corresponding to  $2.7 \times 10^6$  alpha disintegrations per minute) with no spurious counts from the alpha activity expected in some months.

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As more material became available, the effective "safe" sample size was increased to about 500 micrograms ( $6.8 \times 10^7$  alpha disintegrations per minute) by the use of collimating metal screens which partially covered the samples. Use of the screens was suggested by O. R. Frisch.

The action of the screens may be understood by reference to Fig. 4, which shows schematically a parallel-plate ionization chamber such as was used in the experiments. Alpha particles A and B (represented by their tracks in the chamber) will cause much larger ionization pulses to appear on the amplifier output than will particles C and D, for two reasons. First, the dimensions of the chamber are such that A and B expend a larger fraction of their total energy within the sensitive volume of the chamber. Second, the "centers of gravity" of ionization tracks A and B are at greater distances from the collecting electrode than are those of C and D. With electron collection, the pulse height is proportional to the average displacement of the electrons from the ionization track as they are drawn to the collecting electrode. (The positive ions do not contribute to the pulse, as their collection time is much longer than that of the electrons and the decay time of the amplifier is too short for any response to the ion pulse.) Thus tracks A and B are again favored over C and D, and contribute more heavily to the undesired alpha activity background.

A flat metal sheet in which circular holes are drilled as close together as possible makes a good collimator, eliminating all particles emitted at small angles to the plane of the sample and the high-tension electrode but allowing reasonable transparency. Fig. 5, shows the effect of such a screen; a sample of plutonium of 600 micrograms effective weight (over 4 milligrams actual weight) becomes safer (one spurious count per 100 years) at the fission operating bias than a 20-microgram sample used with no screen (one spurious count per year).

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The transparency to fission fragments of the screen used for this test was 13% as calibrated with an enriched-uranium sample using neutron-induced fissions. In most of the experimental work, screens of about 30% transparency, nearly as effective as the 13% screen in reducing alpha activity effects, were used. In these screens the ratio of hole diameter to screen thickness was about 2.5.

4. Argon Chambers And Fast Amplifiers

In making more accurate determinations of spontaneous fission rates in various plutonium samples to be studied, it was desirable to eliminate the screen technique. Since with no screen the chamber efficiency would be close to 100%, much thinner samples could be used with no loss in effective weight; this would improve the flatness of the plateau of the fission bias curve, so that slight changes in amplifier gain during operation would have less effect on the chamber efficiency. The uncertainty introduced by screen-transparency calibrations would be eliminated. Much more rapid accumulation of data would be possible where only a limited amount of material was available. In order to make possible the use of larger samples without screens it was decided to use argon chambers, in which the electron - collection time would be considerably shorter, and to built correspondingly faster amplifiers. The shorter effective resolving time of the new units made possible the observation of plutonium samples as large as 1 milligram (corresponding to  $1.36 \times 10^8$  alpha-disintegrations per minute) without the use of any collimating device, and with no spurious counts expected in some months.

Fig. 6 shows the schematic diagram of the amplifier circuit. The first units were designed and built by C. Wiegand and tested for use with the argon chambers by G. Farwell. A number of units were subsequently built by J. Aeby.

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The amplifiers were designed to use 6-volt automobile storage batteries for the filament supply and 45-volt dry batteries (doubled to 90 volts) for the B supply.

The rise <sup>time</sup> of the amplifiers was about 0.2 microseconds. Tests showed that a very short clipping time favored the fission pulses over the alpha "pile-ups", i.e. for a given sample, at a given efficiency as determined by fission bias curves, the extrapolated alpha activity counting rates became smaller as the clipping time was shortened from several times the rise time to about half the rise time. This was to be expected; the resolution was improved by decreasing the length of time required for single alpha pulses to be forgotten. The fission bias curve did not suffer as a result of the shortened pulse width; the corresponding loss in pulse height was of no consequence, as the amplifier gain was still adequate.

The weighing of samples by neutron-induced fission counts (see sec. II, 5) and the running of alpha activity curves required a faster counting circuit than that used in registering spontaneous fission pulses, if excessive time were not to be lost. For this reason an outlet to an auxiliary counting circuit was provided; the plate and screen of the 6G6 of the counting circuit multivibrator could be opened, permitting use of the 6AK5 output (see Fig. 6) to activate a scale of 64 which could be used with any unit. It was found to be important in this connection to set the amplifier discriminator to correspond to the desired number of millivolts input using the regular counting circuit; then, with the auxiliary circuit in use and the amplifier discriminator bias fixed, to adjust the discriminator on the scaler input so that the same minimum pulse would be registered. Failure to do this resulted in a distortion of the fission and alpha curves when determined with the auxiliary counting arrangement, since the two counting arrangements were no longer equivalent beyond the amplifier discriminator.

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Appreciable shifts in gain over a period of nearly 24 hours could not be tolerated; a large drop in gain would mean a drop in efficiency, while a rise in gain would change the efficiency very little but would mean increased possibility of alpha activity effects. During operation gains were checked daily and biases reset on all units by means of the fast rise time pulse generator drawn schematically in Fig. 7. Its output is a nearly rectangular negative pulse which reaches its full value in a fraction of microsecond. It was found that eight Burgess or Eveready heavy-duty B batteries connected in series-parallel to give 90 volts would provide B voltage for continuous and sufficiently stable operation for one amplifier unit for from four to six weeks. Two large banks of 6-volt automobile storage batteries were used to supply all filaments; one bank could be charged while the other bank (completely shielded) was discharged. The amplifier gains shifted negligibly with the 0.2-to 0.3-volt drop filament voltage which occurred over a 24-hour period. As many as eight fast units were in operation at the same time.

Fig. 8 shows a sectional view of one of the parallel-plate ionization chambers used with argon. The sample (usually electro-deposited on a platinum disc) was mounted on the high-tension electrode. The diameter of the active deposit was 3 to 4 cm; the diameter of the collecting electrode was nearly 7 cm. The chamber depth was 0.8 to 0.9 cm and the operating pressure a few pounds above atmospheric (110 cm Hg abs). Under these conditions, a fission fragment originating anywhere in the sample and travelling either perpendicular to or partially parallel to the plane of the sample would expend most of its energy in a favorable region of

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chamber volume, since its range would be on the average from 1.5 cm to 2.0 cm and it would ionize most heavily in the first few mm of travel. An alpha particle, on the other hand, would on the average expend only a fraction of its total energy before striking the collecting electrode or leaving the sensitive region of the chamber. The geometry and pressure chosen thus discriminated against the alpha activity background.

Because of the large number of units in operation and the frequency of the sample changes, it was highly desirable to use tank argon to eliminate the necessity of purification during the filling procedure. Calcium purifiers mounted on the individual units would be impractical as they would require battery-operated heaters for operation during spontaneous fission observation. "Incandescent lamp grade" argon was obtained. (The purity of this argon is 99.5% to 99.8%, with nitrogen the only impurity present in any measurable quantity. Special effort is made to eliminate oxygen.)

Satisfactory fission and alpha curves were obtained with the first tank argon tried. However, it was found that over a period of some hours the fission and alpha pulse heights dropped noticeably so that day-to-day operation at a constant efficiency was impossible. In attempts to find the source of the gas "poisoning", rubber gaskets and hoses, Scotch tape for sample mounting, and possible traces of Zapon used in making some of the samples were eliminated one at a time; the lowest-vapor-pressure stopcock grease available was used on the filling system; and a liquid-air trap was used in filling to freeze out any condensable vapors. The poisoning persisted. It was finally found that no poisoning occurred in argon from Linde Company tanks, while poisoning did occur in the argon from the Airco tanks tried, although purity specifications were ostensibly the same. A possible source of trouble was some organic vapor from the packing in the Airco tank valves; Linde tank valves made use of metal diaphragms, with no packing present.

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No special precautions were necessary with the Linde Company argon; alpha and fission bias curves could be reproduced after many days of operation on the same filling. Rubber gaskets and small bits of Scotch tape had no bad effect.

Fig. 9 shows two curves of fission counting rate plotted against collection voltage (Ra + Be neutron source, plutonium sample, fixed bias.) Curve (1) was taken with a filling of bad argon, curve (2) with good. With the amplifier bias fixed, the counting rate rises as the average pulse height rises, so that the curves show qualitatively the variation of pulse height with collection voltage. Curve (1), taken with a filling of bad argon, shows a faster rise at low voltages and a drop at higher voltages. A possible explanation is as follows:

The impurity of impurities present in the bad gas may at low voltages increase the electron drift velocity over that in the clean argon, as would addition of a few per cent of CO<sub>2</sub> to purified argon.<sup>3</sup> This would, effectively, increase the pulse height as seen by the amplifier. The drop in pulse height at higher voltages might be explained by increased probability of electron capture in the impurity, due to higher electron velocities in the stronger field. No corresponding drop is observed in curve (2), taken with clean argon, until much higher collection voltages are reached. The operating voltage used was 810 volts, corresponding to a field of about 1000 volts per cm, a value at which pulse height changes negligibly with small changes in collection voltage.

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<sup>3</sup> See, for example, Allen and Rossi, LA 115

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A bad filling could be spotted immediately by watching the alpha background on an oscilloscope screen as the collection voltage was turned on; the pulses would reach a maximum height and then drop as the operating voltage was approached.

Addition of 5% of CO<sub>2</sub> to the clean tank argon made no observable difference in pulse height or resolution under operating conditions; purification of the argon and elimination of rubber would probably be necessary to achieve the fastest collection time afforded by such a mixture. Spectroscopic argon was tried but was not noticeably different from the Linde argon in performance.

#### 5. Calibration of Samples

For calibration of samples a slow-neutron fission weighing procedure was developed.

The neutron source used was a 200-millicurie Ra+Be source contained in a small brass cylinder. The standard source geometry used is shown in Fig. 10. The source is contained in a lead cylinder from which it can be removed through a brass tube. The tube and cylinder are embedded in a paraffin block which serves as a neutron-slowing medium. The paraffin is contained in a metal pail. A cylindrical hole cut in the paraffin exactly fits the chamber lids. The source can thus be placed at an exactly reproducible position with respect to the sample holder in a given chamber, and serves as a source of slow neutrons constant in time. (The neutron spectrum is such that shielding a plutonium sample with cadmium cuts the fission counting rate to about 5% of the unshielded rate.)

A sample of 0.434 mg of plutonium, electrodeposited as a thin layer on a 4-cm-diameter circle on a platinum disc, and containing less than 0.05% Pu<sup>240</sup>, was used as a standard for sample calibrations.

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Using the source and geometry described above and an auxiliary scale of 64 for counting, a fission bias curve was run for the standard sample in each chamber. From these curves, all of which showed good fission plateaus (see, e.g., Fig. 2), the fission counts per minute could be estimated for each chamber for the standard sample for 100% counting efficiency. The amount of plutonium on the standard sample was known to  $\pm 2\%$  from alpha counts made on two different demultiplier units (assuming a half-life of 24,500 years for Pu) and also from a slow-neutron fission comparison with a smaller plutonium sample which had been alpha counted.

The effective weight of a sample in a certain chamber at the operating bias used was determined by a comparison of its fission counting rate (determined with the standard source) with that of the standard sample at 100% efficiency. For example, suppose that the operating bias range for a certain sample in a certain chamber was 30 to 33 millivolts, the initial setting being 30 and the final check for each run of about a day showing about 33. Counts taken with the standard source and source geometry showed an average fission counting rate of 80 counts per minute in this bias range, while the counting rate of the standard was 75 counts per minute in the same chamber at 100% efficiency. The effective weight of the sample under operating conditions would then be  $80/75 \times 0.434$  or  $0.463$  mg Pu<sup>239</sup>, assuming no slow-neutron fission in the small amount of Pu<sup>240</sup> present. These effective weights were usually compared with the weights as estimated from alpha counts of the samples; counting efficiencies under operating conditions were usually found to be between 90% and 98%. These data were useful when substances other than plutonium were under observation and no slow neutron fission weighing was possible.<sup>2</sup>

#### 6. Determination of the Spontaneous Fission Rate of Pu<sup>240</sup>

Mass-spectrographic verification of the presence of Pu<sup>240</sup> in pile-produced plutonium was obtained by Bartlett, Swinehart and Thompson.<sup>4</sup> They reported an abundance ratio Pu<sup>240</sup> of about  $3.3 \times 10^{-4}$ , which was reasonably consistent with the abundance ratio <sup>Pu<sup>239</sup></sup> calculated from the irradiation data for their sample and the capture cross sections of Pu<sup>239</sup> and U<sup>238</sup>. Later and more precise mass-spectrographic analyses<sup>5</sup>

<sup>4</sup> Bartlett, Swinehart, and Thompson, LA 168

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Bartlett, and Swinehart, LA 327

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were useful in obtaining a more accurate value for the spontaneous fission rate of Pu<sup>240</sup> than the  $1.6 \times 10^6$  fissions per gram hour estimated originally (see Sec. I), assuming all spontaneous fission in pile plutonium to be due to Pu<sup>240</sup>. Comparison of mass-spectrograph data and spontaneous-fission data on several plutonium samples of different Pu<sup>240</sup> content gave the following results:

<u>Sample</u>	<u>% Pu<sup>240</sup> in Pu<sup>239</sup> by Mass Spectrograph<sup>5</sup></u>	<u>Fissions per gram hour in Material</u>	<u>Fissions per gram hour in pure Pu<sup>240</sup></u>
CX2A (Clinton reirradiated Pu)	0.118 ± 0.008	1950 ± 200	$1.65 \times 10^6$
CW1 (Hanford reirradiated Pu)	0.647 ± 0.020	10,900 ± 500	$1.69 \times 10^6$
41H (Hanford production Pu)	1.10 ± 0.02	18,200 ± 1000	$1.65 \times 10^6$

It is estimated that the average figure of  $1.66 \times 10^6$  fissions per gram hour for Pu<sup>240</sup> is accurate to 5%.

#### 7. Determination of Pu<sup>240</sup> Content Of Pile Plutonium Samples

The Pu<sup>240</sup> content of a pile plutonium sample was calculated as follows:

$$\frac{\text{Pu}^{240}}{\text{Pu}^{239}} = \frac{\text{fissions observed}}{\text{hours observed} \times \text{gm Pu}^{239} \text{ effective}} \times \frac{1}{1.66 \times 10^6}$$

The spontaneous fission rate in fissions per gram hour for a certain material could usually be determined to ± 5 to 7%. Sources of error were: counting statistics, ± 3 to 4% (standard deviation); effective weights, ± 3 to 5%; weight of standard sample ± 2% (no effect on comparisons between unknown samples). In spontaneous fission observation, from 200 to 500 counts were usually observed on each of two or three plates of the material under observation.

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III. Experimental Results

A discussion of the results of the investigations proposed in section I follows:

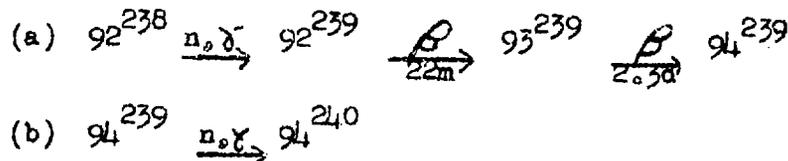
1. Mass-spectrographic verification of the presence of Pu<sup>240</sup> in pile-produced plutonium

This has already been discussed in section II, 6.

2. Specific spontaneous fission of plutonium vs total irradiation

A study of Hanford plutonium samples over a period of some months has shown that the specific spontaneous fission of the plutonium is directly proportional to the ratio Pu<sup>239</sup>/U<sup>238</sup> in the material "pushed" from the pile, and thus directly proportional to the total neutron irradiation suffered by the material.

A more detailed discussion of the production of Pu<sup>240</sup> in a chain-reacting pile is perhaps in order here. The reactions by which it is produced are



Represent the number of atoms of U<sup>238</sup> initially present as N(28); under normal pile operating conditions this number may be considered constant over the period of irradiation of a slug of material. After neutron irradiation for a time t in a flux n v, assumed to be constant throughout the lump, the number of atoms of Pu<sup>239</sup> is

$$(c) \quad N_{49} = \int N_{28} \sigma r(28) n v dt = N_{28} \sigma r(28) n v t$$

where n v t total neutron irradiation on neutrons /cm<sup>2</sup>

and  $\sigma r(28)$  radiative capture cross section of U<sup>238</sup> in cm<sup>2</sup>.

The number of atoms of Pu<sup>240</sup> present is

$$N_{40} = \int [N_{28} \sigma r(28) n v t] \sigma r(49) n v dt$$

or

$$(d) \quad N_{40} = \frac{1}{2} N_{28} \sigma r(28) \sigma r(49) (n v t)^2$$

where  $\sigma r(49)$  is the radiative capture cross-section of Pu<sup>239</sup> in cm<sup>2</sup>.

then

$$\frac{N_{40}}{N_{49}} = \frac{1}{2} \sigma r(49) n v t$$

or

$$(3) \quad \frac{N_{40}}{N_{49}} = \frac{1}{2} \frac{N_{49}}{N_{28}} \frac{\sigma r(49)}{\sigma r(28)}$$

Taking  $\sigma_r(49) = 375 \times 10^{-24} \text{ cm}^2$  (6) and  $\sigma_r(28) = 2.56 \times 10^{-24} \text{ cm}^2$  (7) we have  
for a single lump of material

$$(f) \frac{N_{40}}{N_{49}} = 73.2 \frac{N_{49}}{N_{28}}$$

Corrections must be applied to this formula before it can be considered correct for application to a rod of material irradiated in the Hanford pile. First, a correction must be made for the non-uniformity of the neutron flux over the length of a rod and from outside to center within a rod. Second, a correction must be made  $\sigma_r(28)$  to take into account resonance capture of neutrons in the energy range of a few electron volts. If we call the non-uniformity correction factor  $\beta$  and the resonance correction factor  $\rho$ , equation (f) becomes

$$(g) \frac{N_{40}}{N_{49}} = 73.2 \times \frac{N_{49}}{N_{28}} \times \frac{\beta}{\rho}, \text{ where } \rho = \frac{\sigma_r(28) \text{ effective}}{\sigma_r(28) \text{ thermal}}$$

In this more general expression for  $N_{40}/N_{49}$  the factors  $\beta$  and  $\rho$  are characteristic of the geometry of the particular pile lattice under consideration.

(1) Calculation of the correction factor  $\beta$  :

Equations (c) and (d) for a single small lump of metal in a uniform neutron flux indicate that the amount of  $\text{Pu}^{239}$  present in a large slug of metal is proportional to the average value of  $n v t$  throughout the slug, whereas the amount of  $\text{Pu}^{240}$  present is proportional to the average value of  $(n v t)^2$  throughout the slug. The correction factor for non-uniformity of neutron flux is thus

$$\beta = \frac{\overline{(nvt)^2}}{(\overline{nvt})^2}$$

or

$$\beta = \frac{\overline{n^2}}{(\overline{n})^2}$$

where  $n$  = neutron flux, the neutron velocity distribution assumed to be the same at all points.

(6) Based on  $\sigma_f(49) = 765 \times 10^{-24} \text{ cm}^2$ ,  $\alpha(49)$

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If  $V$  is the volume of a slug of metal, and the flux is a function of position in the slug, then for that slug

$$\beta = \frac{\int_V n^2 dV / \int_V dV}{\left\{ \int_V n dV / \int_V dV \right\}^2}$$

$$\beta = V \frac{\int_V n^2 dV}{\left\{ \int_V n dV \right\}^2}$$

For the cylindrical rod, the correction factor  $\beta$  can be calculated conveniently in two steps:

First, consider non-uniformity of  $n$  along the rod. Call the corresponding correction factor  $\beta_1$ .  $n$  may be represented approximately as a sine distribution having its maximum at the center of the rod's length (about 8 m) and going to zero about 0.4 m beyond the ends of the rod, as represented in Fig. 11.

The flux  $n$  reaches its maximum value  $n_0$  half way along a rod, i.e. at the center of the pile. If  $L$  represents the distance along the rod as measured in meters from the point of zero flux, then

$$n = n_0 \sin x, \text{ where } x = \frac{\pi L}{8.8}$$

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Considering a rod of unit cross-sectional area,

$$\beta_1 = \frac{\int_V n^2 dV}{\left\{ \int_V n dV \right\}^2}$$

$$\beta_1 = \left( \frac{\pi}{2} - \frac{\pi}{22} \right) \frac{\int_{\pi/22}^{\pi/2} n_0^2 \sin^2 x dx}{\left\{ \int_{\pi/22}^{\pi/2} n_0 \sin x dx \right\}^2}$$

$$\beta_1 = 1.13$$

The correction factor  $\beta_1 = 1.13$  is probably accurate to 3 or 4 percent; assumption of a sine distribution going to zero exactly at the ends of the rod (certainly less close to the truth) gives a correction factor of 1.23, and a distribution varying linearly from zero at the ends of the rod to a maximum at the center gives a factor 1.33.

Second, consider differences in  $n$  between the outside of the rod and various points inside. Call the corresponding correction factor  $\beta_2$ .

The equation for diffusion of neutrons in the rod is

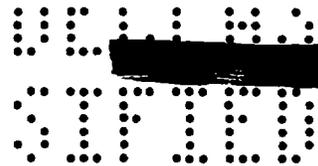
$$\nabla^2 n = \left( 1/l^2 \right) n$$

where  $l$  = diffusion length of thermal neutrons in uranium.

Taking the  $z$  axis along the rod, and assuming cylindrical symmetry, this equation may

be written  $\frac{\partial^2 n}{\partial r^2} + \frac{1}{r} \frac{\partial n}{\partial r} + \frac{\partial^2 n}{\partial z^2} = \frac{1}{l^2} n$

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where  $r$  = distance from the axis of the rod.

The variation of  $n$  along  $z$  is slow and the  $z$  term can be neglected in the consideration of a thin slice of the rod perpendicular to the  $z$  axis (see Fig. 12).

The diffusion equation becomes

$$\frac{d^2 n}{dr^2} + \frac{1}{r} \frac{dn}{dr} = \frac{1}{l^2} n$$

Expressed as a power series the solution of this equation may be written

$$n = n_0 \left\{ 1 + \frac{1}{4} \left( \frac{r}{l} \right)^2 + \frac{1}{64} \left( \frac{r}{l} \right)^4 + \dots \right\}$$

where  $n_0$  = constant

$r$  cannot be greater than 1.73 cm and  $l$  is about 1.55 cm for thermal neutrons in uranium<sup>(8)</sup> for the first several cm of travel. For our purposes, then, the approximate solution

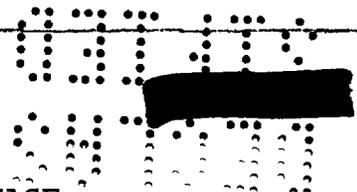
$$n = n_0 \left\{ 1 + \frac{r^2}{4l^2} \right\}$$

can be used to represent the flux distribution in a given cross-sectional slice of the rod.

$n$  has a maximum value of  $n = n_0 (1 + R^2/4l^2) = 1.31 n_0$  at the surface of the cylinder and reaches a minimum value  $n = n_0$  on the axis. The radial distribution correction factor for the rod is

$$\beta_2 = \frac{\pi R^2 \int_0^R n^2(r) \times 2\pi r dr}{\left[ \int_0^R n(r) \times 2\pi r dr \right]^2} = \frac{R^2}{2} \frac{\int_0^R \left( 1 + \frac{r^2}{4l^2} \right)^2 r dr}{\left[ \int_0^R \left( 1 + \frac{r^2}{4l^2} \right) r dr \right]^2}$$

(8) Hughes and Bragdon, CP-1732



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Integrating, one finds for the indicated quantity

$$\beta_2 = \frac{\left[ 1 + \frac{R^2}{4l^2} + \frac{R^4}{48l^4} \right]}{\left[ 1 + \frac{R^2}{8l^2} \right]^2} = 1 + \frac{\left[ \frac{1}{192} \frac{R^4}{l^4} \right]}{\left[ 1 + \frac{R^2}{8l^2} \right]^2}$$

$$\beta_2 = 1.0061 \text{ or } \sim 1.01,$$

a very small correction factor.

The overall correction factor for non-uniformity of neutron flux is thus

$$\beta = \beta_1 \beta_2 = 1.13 \times 1.01 = 1.14$$

and equation (g) may be written

$$(g) \quad \frac{N_{40}}{N_{49}} \approx \frac{73.2 \times 1.14}{\rho} \quad \frac{N_{49}}{N_{28}} \approx \frac{83.4}{\rho} \quad \frac{N_{49}}{N_{28}}$$

(2) Estimate of the resonance correction factor  $\rho$ :

Transmission experiments on uranium have shown the existence of strong absorption resonances at energies of about 6.6, 21, and 38 electron volts.<sup>9</sup> The effect of these resonances is to increase the effective radiative-capture cross section of the surface layers of uranium in the pile.

The best data available on the ratio of capture to fission in pile uranium are reported by Engelkemeir and Freedman<sup>10</sup> and by I. Perlman.<sup>11</sup> Estimates from

9

Anderson, McDaniel, and Sutton, LA 158

10

Engelkemeir and Freedman, CN-1917

11

I. Perlman, CN-2044

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fission product and plutonium assays of slugs of metal irradiated in the Clinton pile give values of the ratio capture to fission of  $0.93 \pm 0.10$ <sup>(10)</sup> and  $0.883$ .<sup>(11)</sup> A ratio, capture/fission =  $0.90 \pm 0.06$  seems reasonable.

The slow-neutron fission cross section of normal uranium is  $542 \times 10^{-24} \text{cm}^2$ <sup>(12)</sup>  $\times 1$  or  $3.87 \times 10^{-24} \text{cm}^2$ . Thus the effective capture cross section of  $\text{U}^{238}$  is about  $3.87 \times (0.90 \pm 0.06) \times 10^{-24}$  or  $(3.5 \pm 0.25) \times 10^{-24} \text{cm}^2$ . By comparing this value with the thermal-neutron cross-section  $\sigma_r(28) = 2.53 \times 10^{-24} \text{cm}^2$ , one finds that the resonance correction factor is

$$\rho = \frac{\sigma_r(28) \text{ effective}}{\sigma_r(28) \text{ thermal}} = \frac{3.5 \pm 0.25}{2.53}$$

i.e. probably lies between 1.28 and 1.48. Thus the determination of  $\rho$  introduces the largest uncertainty present in the calculation of the ratio  $\frac{\text{Pu}^{240}/\text{Pu}^{239}}{\text{Pu}^{239}/\text{U}^{238}}$ .

Results of spontaneous-fission analyses made on nineteen plutonium samples ranging in  $\text{Pu}^{240}$  content from 0.17% to 1.6% are tabulated below. The  $\text{Pu}^{240}$  content was calculated from the spontaneous-fission data, assuming a spontaneous fission rate of  $1.66 \times 10^6$  fissions per gram hour for pure  $\text{Pu}^{240}$  (see Sec II, 7). The ratio  $\text{Pu}^{239}/\text{U}$  refers to the  $\text{Pu}^{239}$  content of the uranium as it was used from the pile; these data were received from Hanford and are based on assays of the solutions resulting from the initial U - Pu separation process.

(12)  $\sigma_f(\text{U}^{235})$  as given in LA 140A

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Pu Sample *	ppm Pu <sup>240</sup> / Pu <sup>239</sup> by Spontaneous fission Measurement	ppm Pu <sup>239</sup> / U from Hanford Data	Pu <sup>240</sup> / Pu <sup>239</sup> : Pu <sup>239</sup> / U <sup>235</sup>
1 W	1.75 x 10 <sup>3</sup>	26.5	65.5
2 W	2.83 x 10 <sup>3</sup>	37.7	74.5
3 W	2.91 x 10 <sup>3</sup>	38.9	74.4
4 W	3.34 x 10 <sup>3</sup>	41.9	79.1
5 W	4.15 x 10 <sup>3</sup>	53.0	77.8
5871	4.45 x 10 <sup>3</sup>	62.2	71.0
41 H	1.10 x 10 <sup>4</sup>	145	75.4
50 H	9.45 x 10 <sup>3</sup>	138	68.0
60 H	1.39 x 10 <sup>4</sup>	185	74.5
71 H	1.16 x 10 <sup>4</sup>	180	63.8
72 H	1.17 x 10 <sup>4</sup>	173	67.1
74 H	1.29 x 10 <sup>4</sup>	198	64.7
75 H	1.30 x 10 <sup>4</sup>	187	69.3
76 H	1.39 x 10 <sup>4</sup>	177	78.0
90 H	1.35 x 10 <sup>4</sup>	206	65.3
110 H	1.31 x 10 <sup>4</sup>	193	67.2
130 H	1.55 x 10 <sup>4</sup>	217	71.0
150 H	1.60 x 10 <sup>4</sup>	218	72.8
170 H	1.60 x 10 <sup>4</sup>	233	68.2
		Average	70.9

\* in the control of plutonium material on the project a number of other plutonium samples have been analyzed by this method. Results are not given here because the samples were mixtures for which Pu<sup>239</sup>/U data were not available.

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Fig. 13 shows a plot of the ratio  $\text{Pu}^{240}/\text{Pu}^{239} + \text{Pu}^{239}/\text{U}^{238}$  against the ratio  $\text{Pu}^{240}/\text{Pu}^{239}$  as determined by spontaneous fission measurements. The estimated probable error in most of the  $\text{Pu}^{240}/\text{Pu}^{239}$  determinations is  $\pm 6\%$ ; it has been assumed that the  $\text{Pu}^{239}/\text{U}^{238}$  data are correct, although uncertainties of considerable magnitude exist in these assays.<sup>13</sup> It is readily seen that, within the limits of the experimental errors involved, the ratio  $\text{Pu}^{240}/\text{Pu}^{239}$ , calculated directly from the specific spontaneous fission of the plutonium, is equal to a constant times the ratio  $\text{Pu}^{239}/\text{U}^{238}$ . This establishes the certainty that the substance having the high spontaneous fission rate is formed by two successive neutron processes starting from  $\text{U}^{238}$ . The contribution of  $\text{Pu}^{238}$  has been shown to be negligible (see next section) and the isotope  $\text{Pu}^{240}$  is therefore definitely identified as the high spontaneous fissioner.

From the experimental data one obtains the relation

$$(h) \quad \frac{N_{40}}{N_{49}} = 70.9 \frac{N_{49}}{N_{28}}$$

Comparing equation (h) with equation (g'), one finds that the experimental constant 70.9 agrees with the calculated constant 83.4 if the resonance absorption correction is 18%, i.e. if  $\rho$  is taken as 1.18. This value for  $\rho$  appears somewhat low in view of the estimate of 1.28 to 1.48 on p.25. However, the uncertainty in the capture/fission ratio used to calculate  $\rho$ , as well as the possibility of fairly large errors in the  $\text{Pu}^{239}/\text{U}$  assays made at Hanford, make the agreement reasonable.

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Letter of Col. Matthias, RIDM-PA 410.2, 20 July 1945

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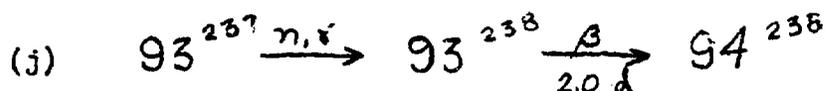
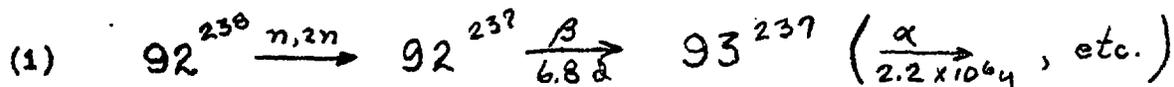


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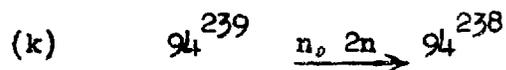
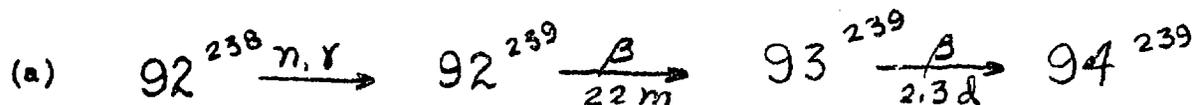
3. Production of  $\text{Pu}^{238}$  in pile plutonium, and measurement of its spontaneous fission:

$\text{Pu}^{238}$  (14) is an  $\alpha$ -emitter having a half-life of about 60 years. (15)

It is produced in the pile from  $\text{U}^{238}$ , principally by the following two successive neutron processes:



Another pair of neutron processes leading to  $\text{Pu}^{238}$  is:



It is of primary interest to estimate the ratio  $\text{Pu}^{238}/\text{Pu}^{240}$  in pile-produced plutonium. Remembering that  $\text{Pu}^{240}$  is produced by reactions (a) and (b)  $94^{239} (n, \gamma) 94^{240}$ , it is easy to show that the  $\text{Pu}^{238}$  produced by reactions (i) and (j) compares with  $\text{Pu}^{240}$  in the ratio

$$(1) \quad \frac{\text{Pu}^{238}}{\text{Pu}^{240}} = \frac{\sigma_{n,2n}(28) \sigma_{\gamma}(37)}{\sigma_{\gamma}(28) \sigma_{\gamma}(49)}$$

where  $\sigma_{\gamma}(37)$  is the radiative-capture cross section of  $\text{Np}^{237}$  in  $\text{cm}^2$ .

For the mass assignment see

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Kennedy, Perlman, Segre, Wahl, A-207

15

Seaborg, CN-2767



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Measurements of the yield of  $\text{Np}^{237}$  in the x pile have shown that it is of the order of 0.3% of that of  $\text{Pu}^{239(16)}$ , indicating that  $\frac{\sigma(n, 2n)(28)}{\sigma_r(28)} \approx 3 \times 10^{-3}$ , and thus that  $\sigma_{n, 2n}(28)$  for pile neutrons  $\approx 0.01 \times 10^{-24} \text{cm}^2$ .  $\sigma_r(37)$  is about  $110 \times 10^{-24} \text{cm}^2$ , (17) and  $\sigma_r(49)$  is about  $375 \times 10^{-24} \text{cm}^2$  (see sec III, 2). Using these cross-sections, one finds

$$(1') \quad \frac{\text{Pu}^{238}}{\text{Pu}^{240}} \approx 0.9 \times 10^{-3}$$

Reaction (k), i.e. the  $n, 2n$  reaction on  $\text{Pu}^{239}$ , contributes very little to the concentration of  $\text{Pu}^{238}$ . Considering this reaction alone, and assuming  $\sigma_{n, 2n}(49)$  to be of the same order of magnitude as  $\sigma_{n, 2n}(28)$ , one finds

$$\frac{\text{Pu}^{238}}{\text{Pu}^{240}} = \frac{\sigma_{n, 2n}(49)}{\sigma_r(49)} \approx \frac{0.01}{375} \approx 3 \times 10^{-5}$$

That the contribution from this reaction is small is confirmed by the fact that range analysis of reirradiated plutonium having a  $\text{Pu}^{240}$  content of as much as several percent has shown that no appreciable increase in  $\text{Pu}^{238}$  content occurs as a result of prolonged neutron bombardment of  $\text{Pu}^{239}$ .

Equation (1') for the ratio  $\text{Pu}^{238}/\text{Pu}^{240}$  is in good quantitative agreement with the statement by Perlman and Seaborg<sup>18</sup> that 13% of the increase in specific activity of pile plutonium over plutonium of negligible  $\text{Pu}^{240}$  content is due to  $\text{Pu}^{238}$ ,

16

Beard, CF-2914

17

Jaffey and Magnusson, CF-2914

18

Perlman and Seaborg, MUC-GTS-1872, 28 July 1945

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the remainder of the increase is due to Pu<sup>240</sup>.

The spontaneous fission measurements on Pu<sup>238</sup> were carried out in the argon-filled ionization chambers. The samples used were thin films deposited by evaporation on platinum discs. The Pu<sup>238</sup> used to make the samples was kindly furnished by G. T. Seaborg. It was prepared by a (d, 2n) reaction on U<sup>238</sup>.

Alpha-range analysis of the samples indicated that a rather heavy Pu<sup>239</sup> contamination had occurred in the laboratory processing. It shows range curves taken with a variable pressure differential range chamber. Resolution of the Pu<sup>239</sup> and Pu<sup>238</sup> peaks (at 3.68 and 4.10 cm, respectively) may be accomplished by giving to either peak the shape of the Po curve as measured under identical conditions and subtracting it from the combined Pu<sup>238</sup> - Pu<sup>239</sup> curve, as shown. An approximate ratio of alpha activities of Pu<sup>238</sup>/Pu<sup>239</sup> = 0.895 was obtained, corresponding to a ratio by weight of

$$\frac{\text{Pu}^{238}}{\text{Pu}^{239}} = 0.895 \times \frac{T_{\frac{1}{2}}(238)}{T_{\frac{1}{2}}(239)} = 0.895 \times \frac{60}{24400} = 0.0022$$

The effective amount of Pu<sup>238</sup> present in each sample under the conditions of observation in an ionization chamber was determined by a slow-neutron fission weighing of the sample against a Pu<sup>239</sup> standard to determine the effective weight of the Pu<sup>239</sup> contaminant. It was assumed here that the contribution of the small amount of Pu<sup>238</sup> present to the slow-neutron fission rate was negligible. The effective amount of Pu<sup>238</sup> present was then

$$\text{Pu}^{238}(\text{eff.}) = \text{Pu}^{239}(\text{eff.}) \times 0.0022$$

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The spontaneous-fission data obtained are summarized in the following table:

<u>Sample</u>	<u>Weight of Pu<sup>239</sup> (eff.)</u>	<u>Weight of Pu<sup>238</sup> (eff.)</u>	<u>Hours Obs'd</u>	<u>Gm x hr Observed (Pu<sup>238</sup>)</u>	<u>Fissions Observed</u>
M-23-1	15.4 x 10 <sup>-6</sup> gm	3.4 x 10 <sup>-8</sup> gm	180	6.1 x 10 <sup>-6</sup>	42
M-23-2	7.8 x 10 <sup>-6</sup>	1.7 x 10 <sup>-8</sup>	384	6.5 x 10 <sup>-6</sup>	51
M-23-3	10.3 x 10 <sup>-6</sup>	2.3 x 10 <sup>-8</sup>	269	6.2 x 10 <sup>-6</sup>	51
Totals.....				18.8 x 10 <sup>-6</sup>	144

From these data one obtains for the spontaneous fission rate of Pu<sup>238</sup> a value of  $144 / (18.8 \times 10^{-6})$  or  $7.7 \times 10^{-6}$  fissions per gram hour. The estimated probable error is  $\pm 15\%$ .

It was necessary to make certain that the observed counts were not due to Pu<sup>240</sup> present in the Pu<sup>239</sup> contaminant. According to Seaborg (personal communication with Allison), no plutonium which came from the pile at a concentration greater than Pu<sup>239</sup>/U = 2 gm/ton can have gotten into the Pu<sup>238</sup>; from spontaneous fission data one knows that this means about 130 ppm Pu<sup>240</sup>/Pu<sup>239</sup>. The spontaneous fission rate of material of this composition is  $\sim 200$  fissions/gm hr. The total gram hours of observation of the Pu<sup>239-240</sup> contaminant were about  $9 \times 10^{-3}$ , so that 2 fissions might be expected during the time of observation.

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The maximum possible effect due to Pu<sup>240</sup> is thus of the order of 2/144 or 1 to 2% of the observed effect.

Using the above data on Pu<sup>238</sup>, it is possible to calculate the effect of Pu<sup>238</sup> spontaneous fission on the observations made on pile plutonium, in which the observed fission activity was attributed to Pu<sup>240</sup>. As stated above, a reasonable estimate for the Pu<sup>238</sup> content is  $\text{Pu}^{238}/\text{Pu}^{240} \approx 0.9 \times 10^{-3}$ .

The relative spontaneous fission rates are

$$\frac{\text{Pu}^{238}}{\text{Pu}^{240}} = \frac{7.7 \times 10^{+6}}{1.66 \times 10^{+6}}$$

so that the contribution of Pu<sup>238</sup> to the observed spontaneous fission of the plutonium samples studied is of the order of  $7.7 \times 10^{+6} \times 0.9 \times 10^{-3} / (1.66 \times 10^{+6})$ , or less than one percent.

4. Investigation of higher isotopes of plutonium and isotopes of elements 95 and 96.

Spontaneous-fission observations on these isotopes were unnecessary in an identification of the high-fission substance, in view of the experimental results already discussed. However, the few data obtained are perhaps worth mentioning.

According to recent Chicago summary reports,<sup>19</sup> the following identifications

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<sup>19</sup> See, for example, CS 3312, CS 3237.

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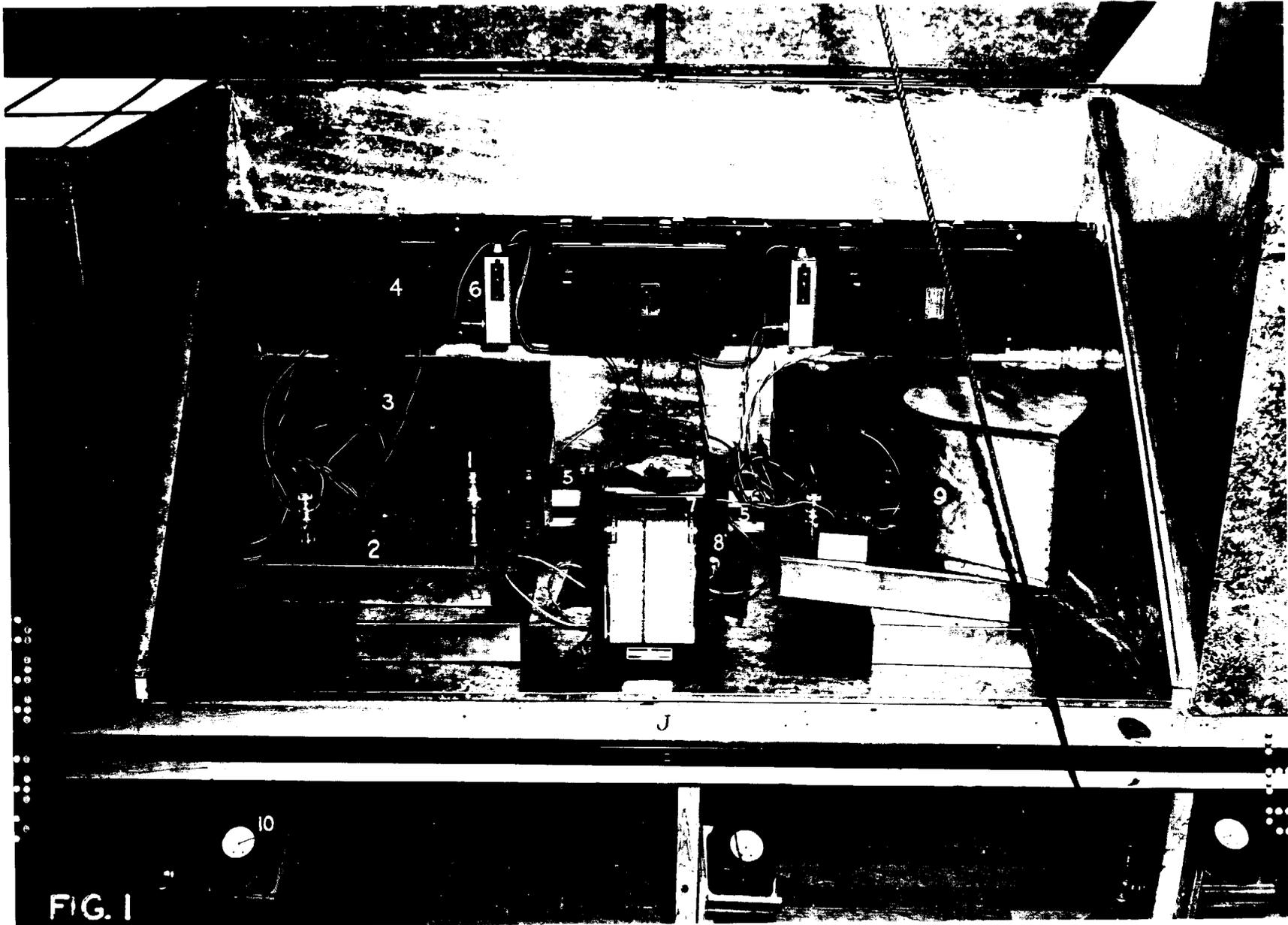
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of pile products have been made:

- (1) A long-life (perhaps some  $10^2$  years)  $\beta$ -activity of about 20 Kev energy is assigned to  $94^{241}$  formed by an  $n, \gamma$  reaction on  $94^{240}$ .
- (2) An  $\alpha$ -activity of range 4.05 cm and half-life of about 40 years, growing from  $94^{241}$ , is assigned to  $95^{241}$ .
- (3) A 17- to 18-hour  $\beta$ -activity is assigned to  $95^{242}$  formed by an  $(n, \gamma)$  reaction on  $95^{241}$ .
- (4) An  $\alpha$ -activity of range 4.75 cm and half-life of about 4 months is assigned to  $96^{242}$  formed by the decay of  $95^{242}$ .

Of these substances, all due to third- or fourth-order neutron reactions, only  $95^{241}$  has been investigated here for spontaneous fission. A sample of this material was kindly furnished by G. R. Seaborg. Three plates were made, each containing about  $7 \times 10^{-9}$  gm of  $95^{241}$  plus several milligrams of lanthanum. (The  $95^{241}$  weights were deduced from the alpha activity assuming a half-life of 40 years.) The samples were observed for a total of 2700 hours, corresponding to  $1.8 \times 10^{-5}$  gm hr. Three fissions were registered. The resulting spontaneous fission rate of  $1.7 \times 10^{+5}$  f/gm hr is considered as an upper limit for  $95^{241}$ .

Assuming for  $94^{240}$  a radiative-capture cross section even as high as  $1000 \times 10^{-24}$  cm<sup>2</sup>, the ratio  $\text{Pu}^{241}/\text{Pu}^{240}$  in pile plutonium would be of the same order of magnitude as the ratio  $\text{Pu}^{240}/\text{Pu}^{239}$  and the ratio  $95^{241}/\text{Pu}^{240}$  would be considerably smaller because of the long period of  $\text{Pu}^{241}$  and to the possibility of capture or fission therein. It is thus only to be expected that no increase in spontaneous fission activity beyond that expected from  $\text{Pu}^{240}$ , attributable to  $95^{241}$  or higher isotopes, has been observed in pile plutonium of increased content of  $\text{Pu}^{240}$  and higher isotopes; nor has any excess activity attributable to the third-order product  $\text{Pu}^{241}$  been observed.



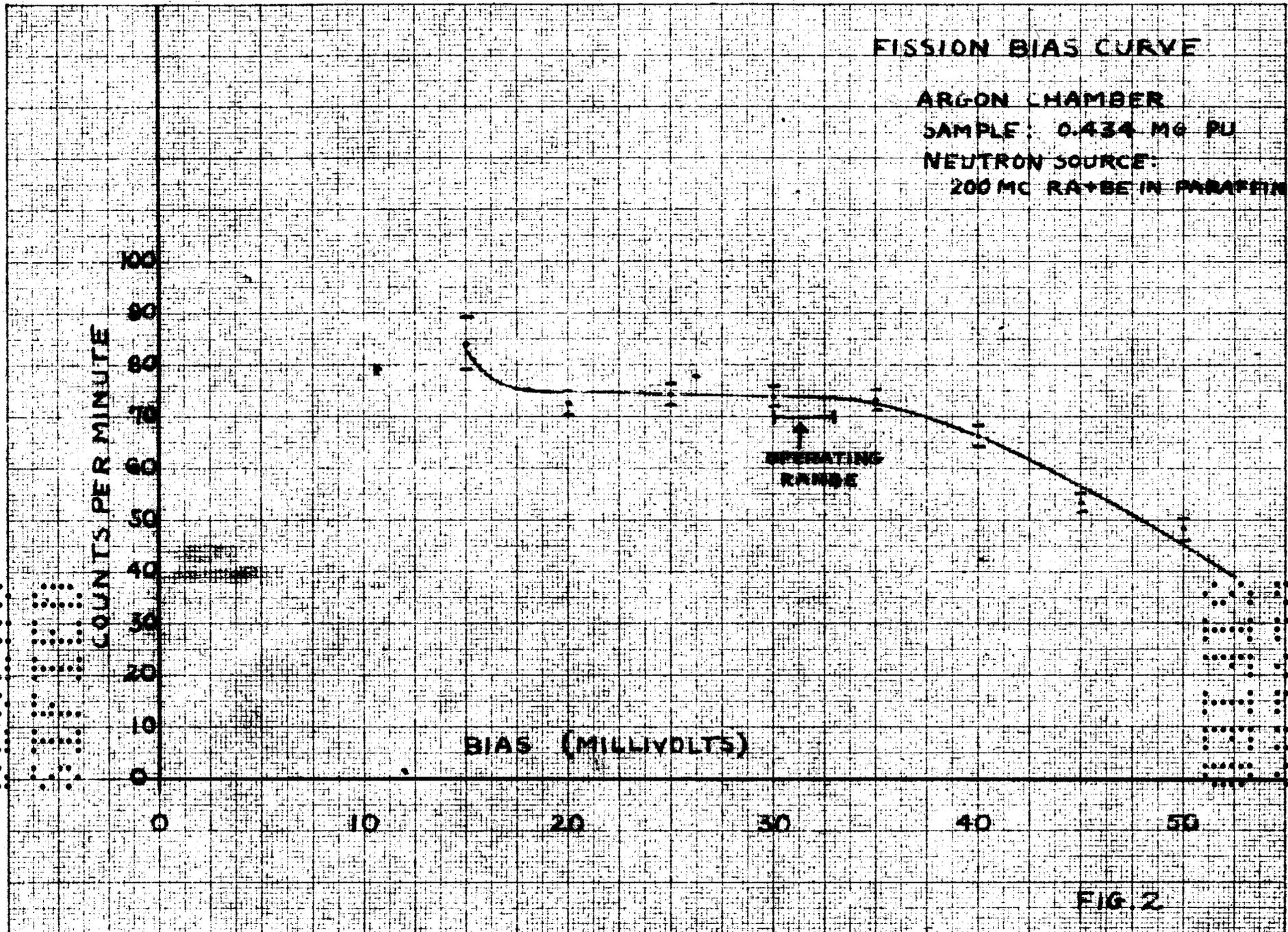
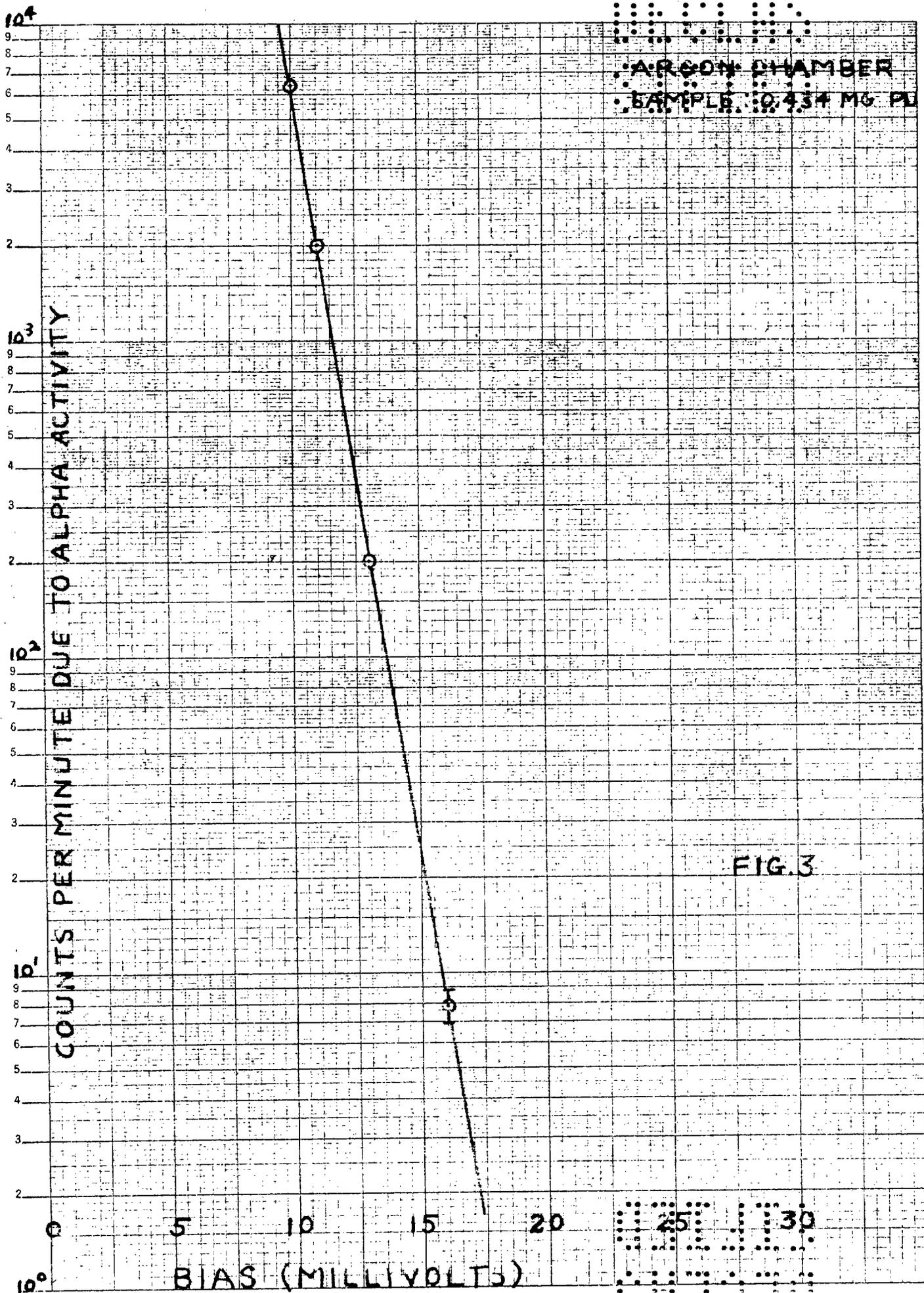


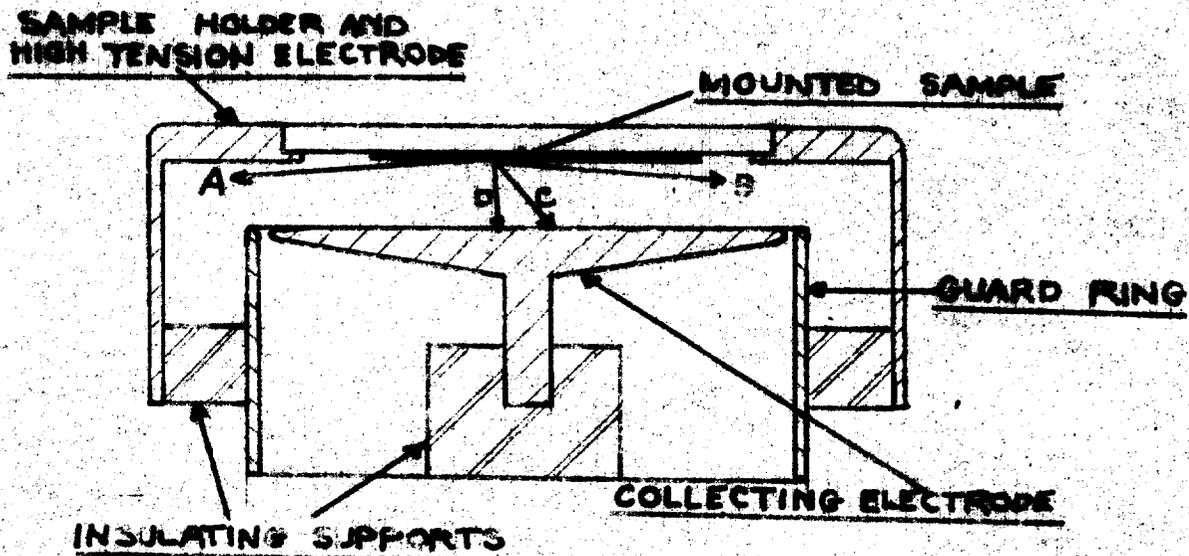
FIG. 2

KEUFFEL & ESSER CO., N. Y. NO. 355-B1  
Semi-Logarithmic, 3 Cycles X 10 to the Inch.  
MADE IN U.S.A.

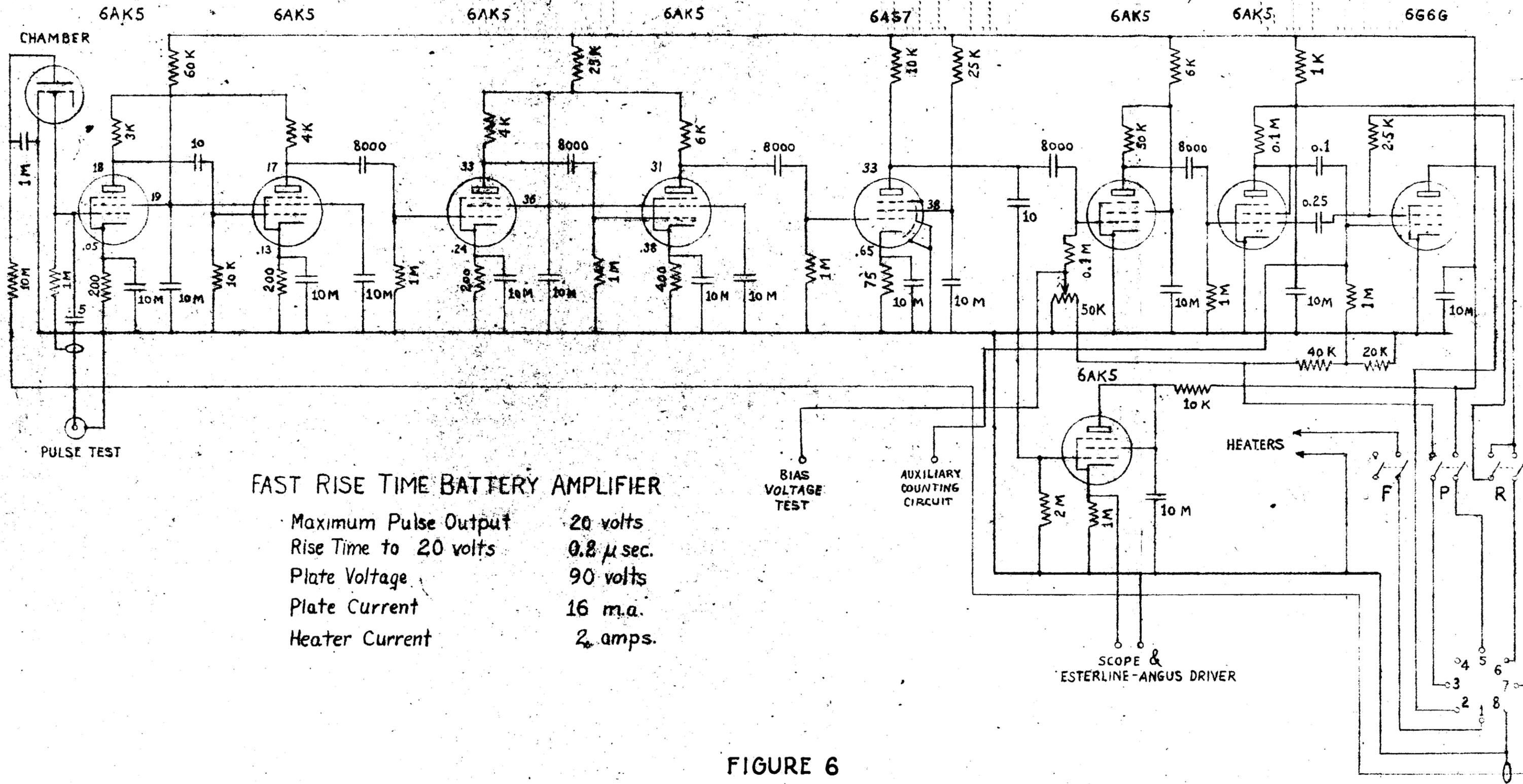


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FIGURE 4



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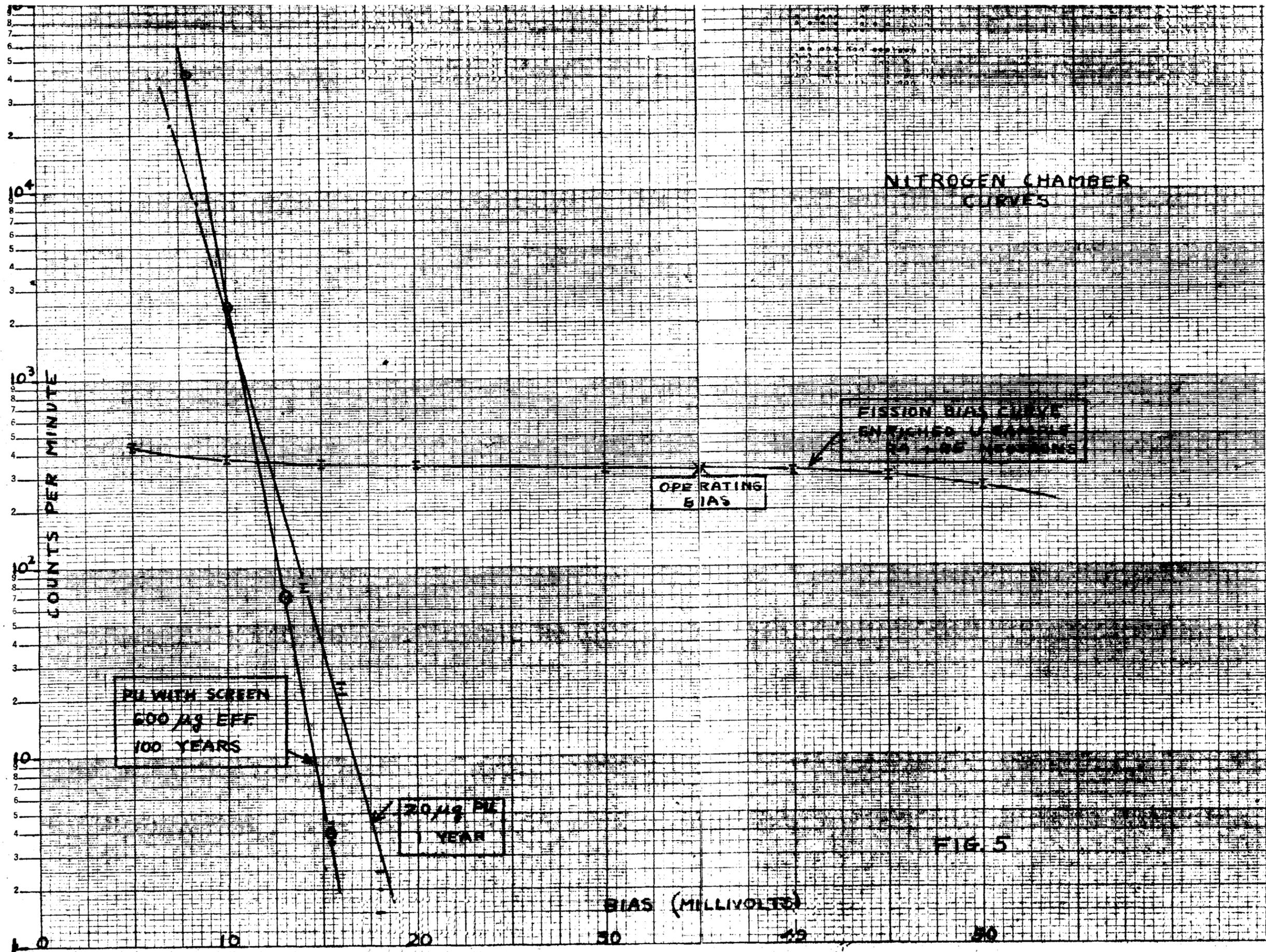
**FAST RISE TIME BATTERY AMPLIFIER**

- Maximum Pulse Output      20 volts
- Rise Time to 20 volts      0.2  $\mu$  sec.
- Plate Voltage                90 volts
- Plate Current                16 m.a.
- Heater Current                2 amps.

**FIGURE 6**

C. WIEGAND 11-7-44  
 FAST R.T. BATTERY AMPL. R-4-21395 A.K.G.W.

KEUFFEL & ESSER CO., N. Y. NO. 160-91  
Semi-Logarithmic, 5 Cycles X 10 to the Inch, 6 1/2" linear vertical  
MADE IN U. S. A.



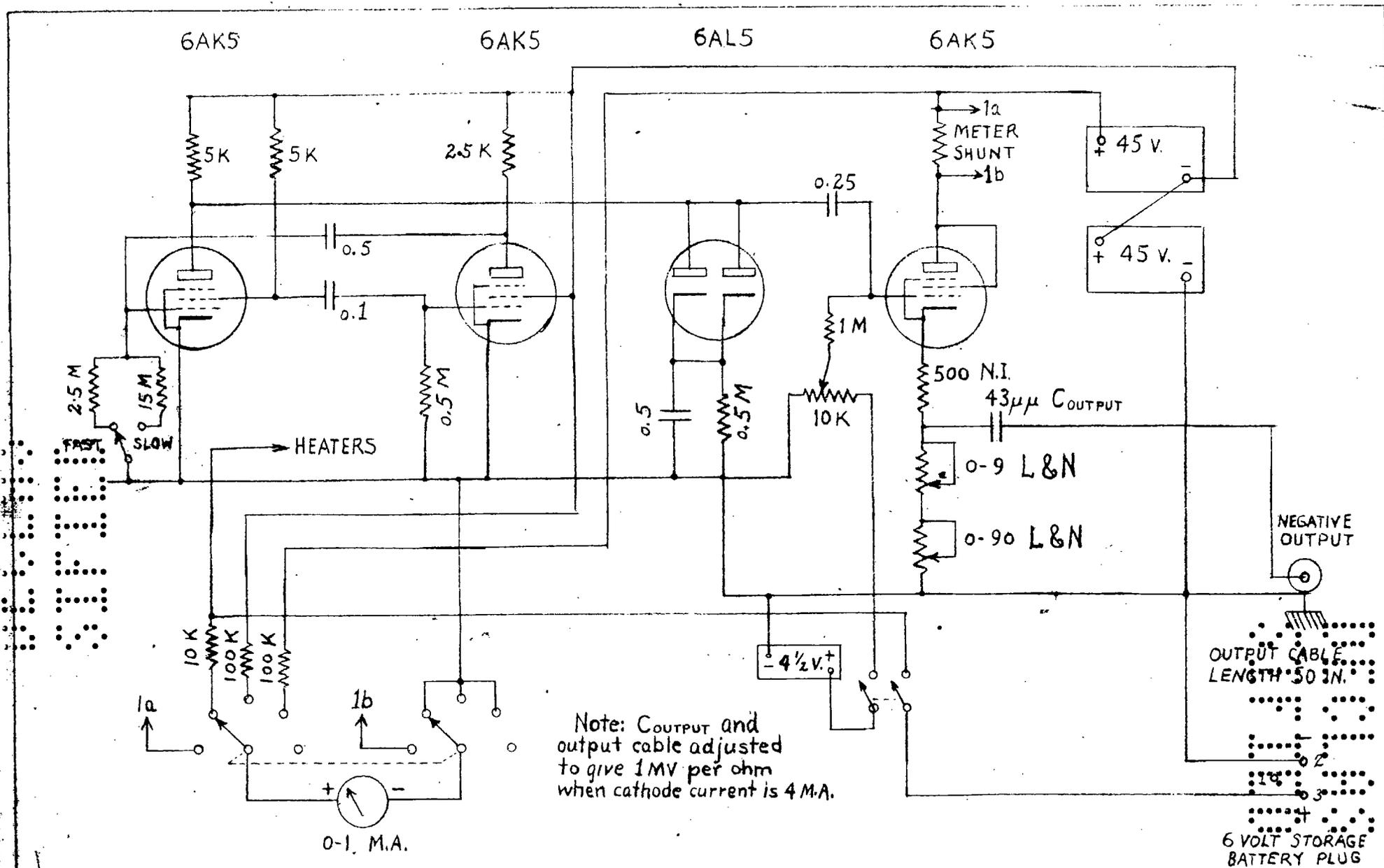


FIGURE 7

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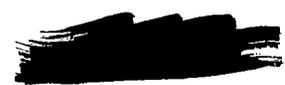
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Fig. 8. Argon Chamber

NUMBERED ITEMS

1. Brass cover
2. Mounted Sample
3. Sample holder and high tension electrode
4. Collecting electrode
5. Guard ring
6. Polystyrene insulating supports
7. Rubber gasket
8. Threaded collar fastens chamber to amplifier chassis
9. High tension lead-(Pt-glass seal waxed in place)
10. Grid lead
11. Gas outlet



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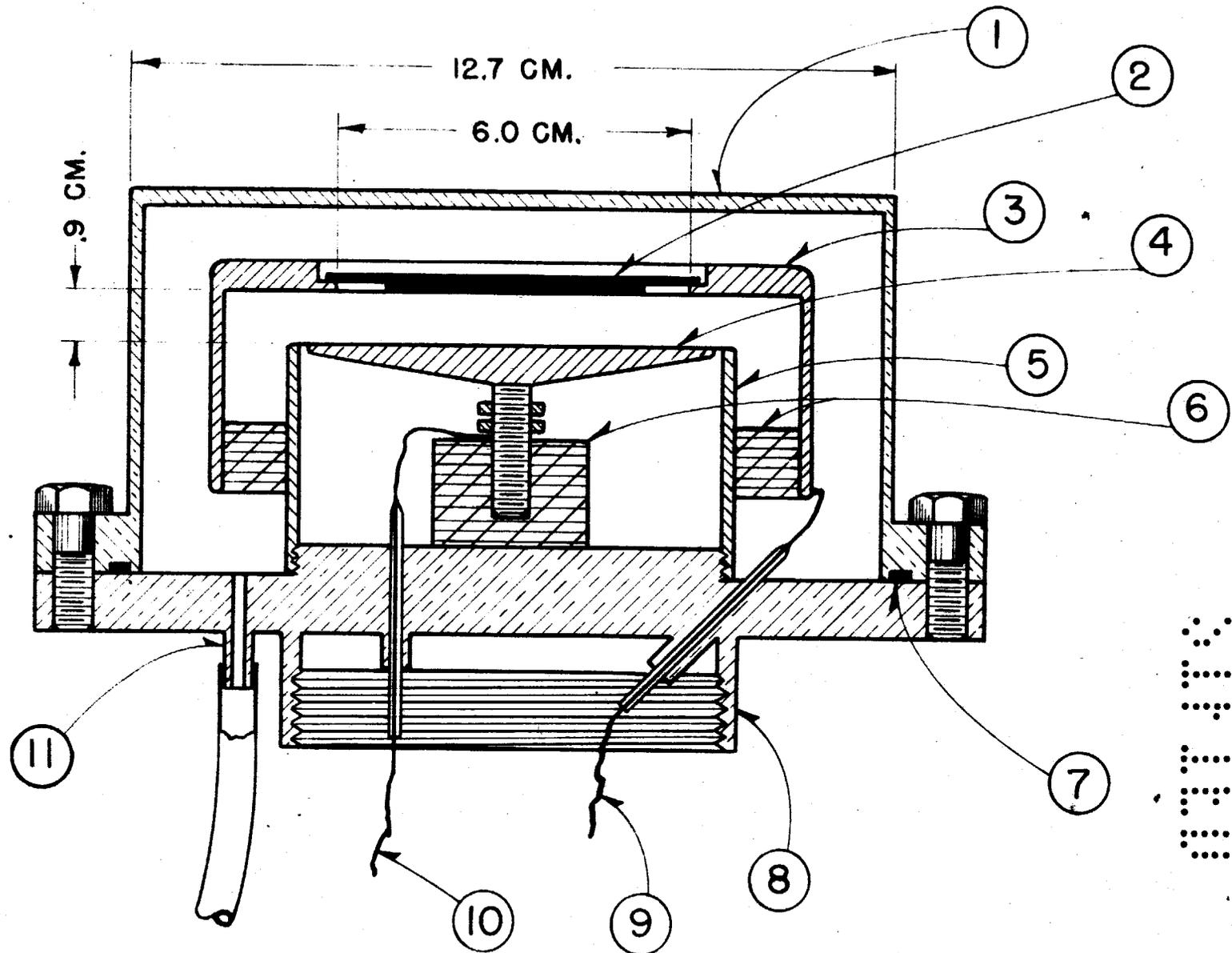
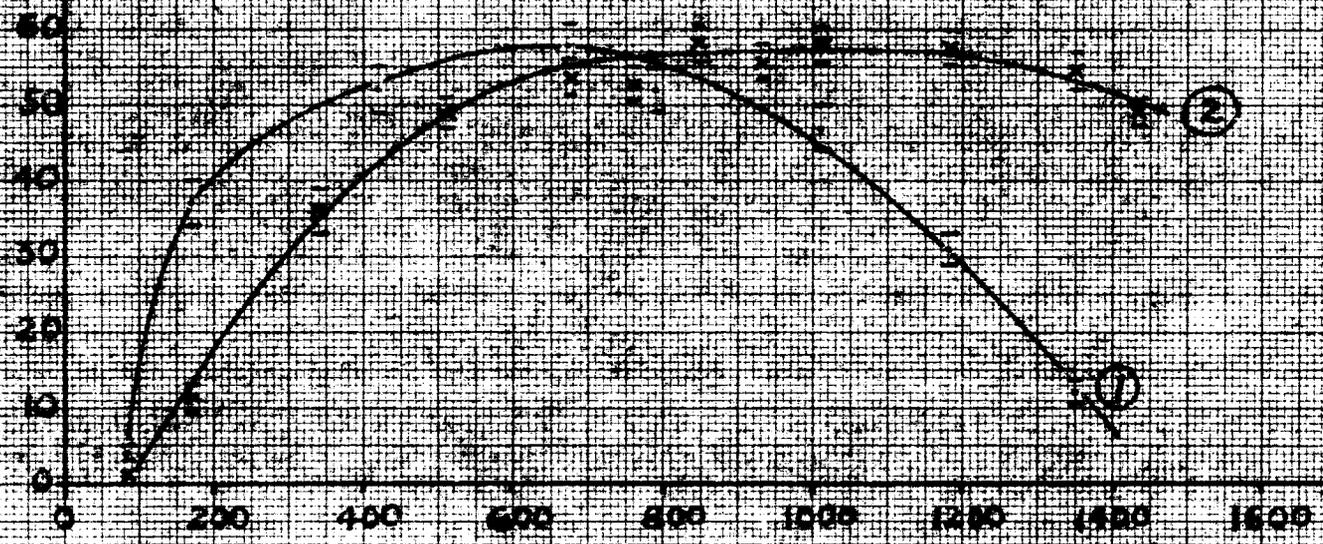


FIGURE 8

Millimeters, 10th lines heavy.  
MADE IN U. S. A.

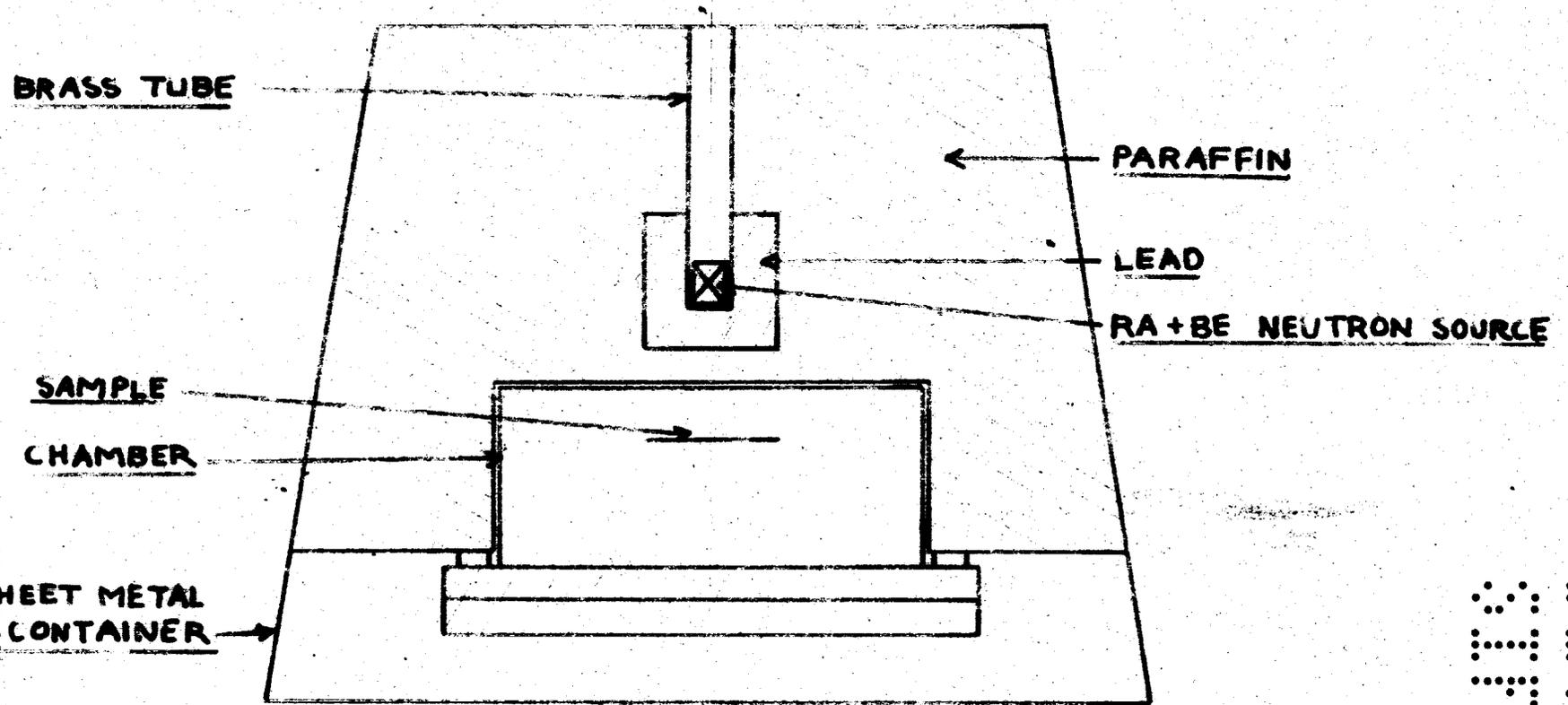
ARGON (PRESSURE 100 CM Hg)  
FISSION PULSES  
BIAS FIXED

COUNTS PER MINUTE



VOLTS PER CM

FIG. 9



WEIGHING GEOMETRY

SCALE: HALF SIZE

FIGURE 10

00113

00700

FIGURE 11

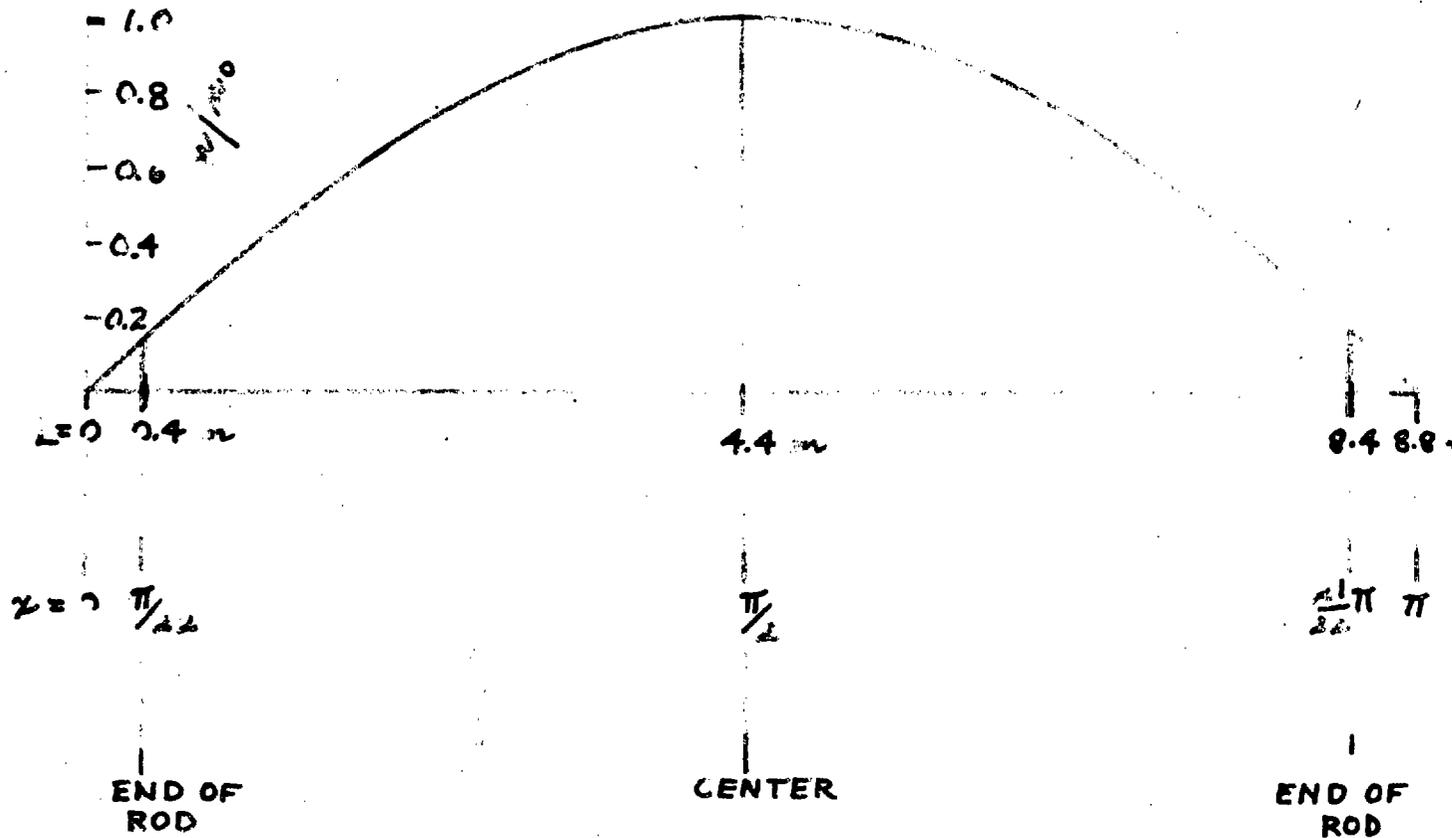
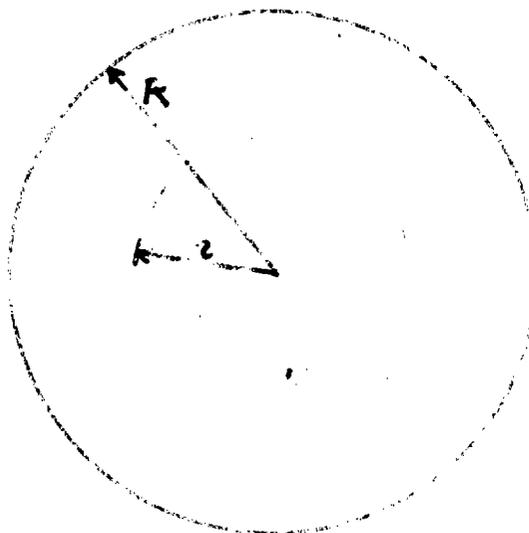


FIGURE 12

$$r = r_0 \left( 1 + \frac{v^2}{4l^2} \right)$$

$$K = 1.75$$

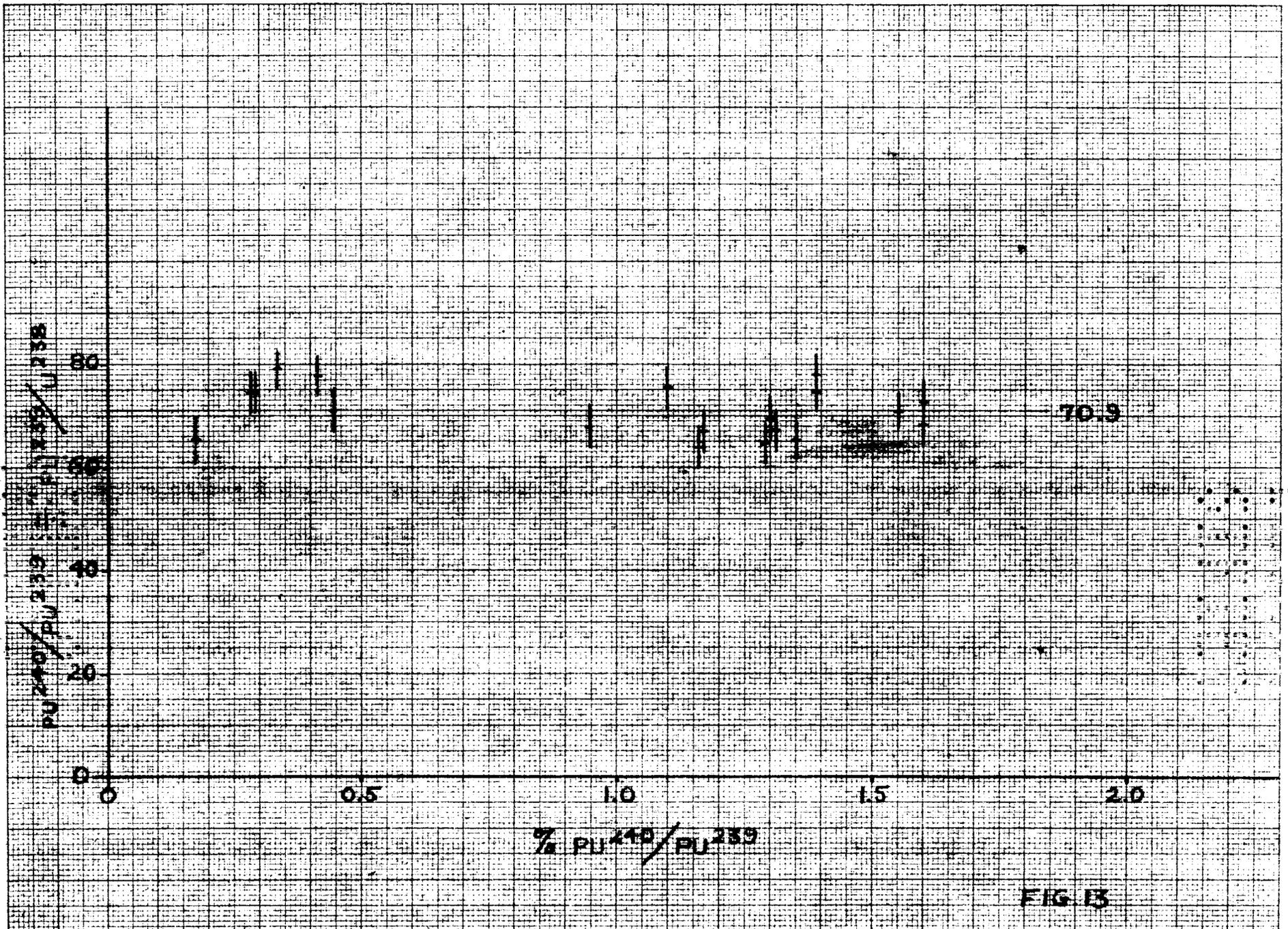
$$l = 1.75$$



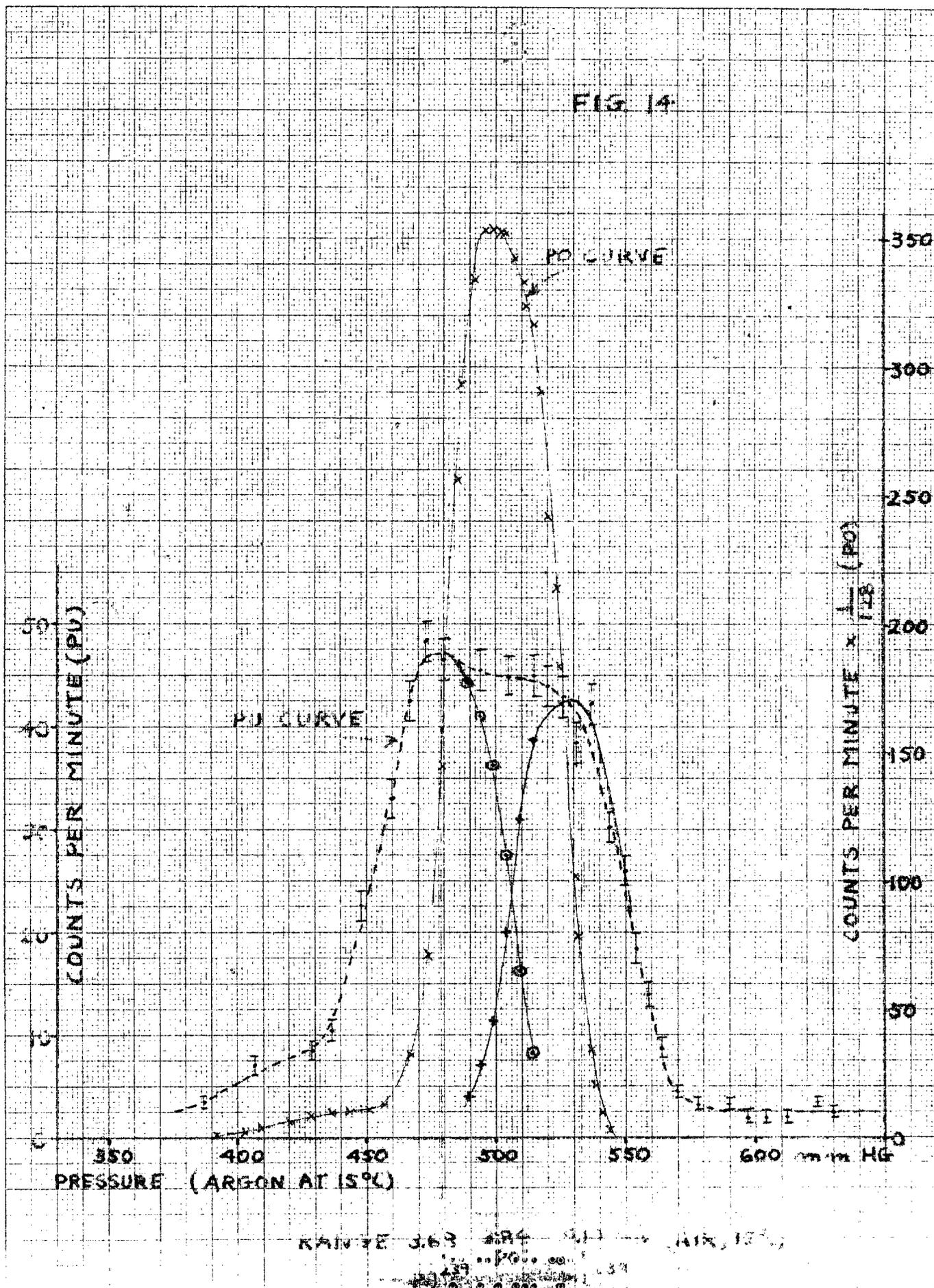
00113

00700

Milimeters, 10th lines heavy.  
MADE IN U.S.A.



KEUFFEL & ESSER CO., N. Y. NO. 359-14  
Millimeters, Ink film heavy.  
MADE U.S.A.





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ADDENDA TO LA-490

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Per J. Brown, FSS-16 Date: 6-2-96  
By Marlene Lyons CIC-14 Date: 5-23-96

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This report describes in detail the work on spontaneous fission in plutonium which was done subsequent to the early investigations reported in LAMS-131 by Chamberlain, Farwell, and Segre. LAMS-131 reported the discovery of Pu<sup>240</sup> and gave a rough estimate of its spontaneous fission rate. The present report (LA-490) gives spontaneous fission data on Pu<sup>238</sup> as well as more recent data on Pu<sup>240</sup> and described methods developed for more accurate determination of spontaneous fission rates. (An overall picture of the spontaneous fission measurements made on a number of isotopes of heavy elements is given by E. Segre in Chapter V of the Los Alamos Technical Series.)

The work described herein was guided and supervised by E. Segre. T/3 A. Spano of the Special Engineering Detachment at Los Alamos gave considerable assistance in the routine conduct of much of the experimental work. C. Wiegand was responsible for the design and construction of the first battery-operated linear amplifier units.

Following suggestions made by S. K. Allison and E. Fermi, several changes and additions have been made since the report was printed at Los Alamos as LA-490.

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1. Add attached introduction before abstract
2. p.3, line 14, in parentheses (n,  $\gamma$  ).  
line 17, change woth to worth.  
footnote, add initial O. before Chamberlain.
3. p 4, line 20, change paraellal to parallel.
4. p 6, line 2, change superosition to superposition.  
line 5, make addition as follows:

...average time interval between alpha pulses, fluctuations in the alpha activity background...

line 6, change he to be.

5. p 11, line 13, insert as follows:

....0.3 volt drop in filament voltage....

line 22, change partically to practically.

6. p 17, line 19, insert equal sign as follows:

...where n v t = total neutron....

line 20, insert equal sign as follows:

...r(28) = radiative capture...

line 26, capitalize letter T in then.

line 29, change (3) to (e); also correct forumla:

$$\frac{N_{40}}{N_{49}} = \frac{1}{2} \frac{N_{49}}{N_{28}} \frac{\sigma_r (49)}{\sigma_r (28)}$$

7. p 18, delete top half of page down to (1) Calculation...;  
change (1) to (2);  
also delete footnotes at bottom of page;  
insert four pages designated Insert I at top of  
page 18 to replace deleted material.
8. p 22, delete bottom on page starting with  
  
...and equation (g) may be written... (line 7);  
also delete footnotes; Insert five pages designated Insert II to replace deleted material.
9. p 23, delete top half of page down to paragraph starting  
Results of spontaneous....;  
also delete footnote

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10. p 24, Capitalize first word of footnote.
11. p 25, delete lines 15 through 20  
Comparing equation.....agreement reasonable.  
Insert material marked Insert III to replace deletion.
12. p 26, remove 2nd and 3rd lines from bottom of page which read:

For the mass assignment see

<sup>14</sup>Kennedy, Perlman, Segre, Wahl, A-207

R Rewrite as follows:

<sup>14</sup>For the mass assignment see Kennedy, Perlman, Segre, Wahl, A-207.

13. p 27, line 4, change the figures 375 to 460;  
in equation (1') change 0.9 to 0.7.
14. p 29, line 2 below table, change  $7.7 \times 10^{-6}$  to  
 $7.7 \times 10^0$ .  
line 7 below table, change gotten to gotten.
15. p 30, line 6, change 0.9 to 0.7;  
to the equation in the middle of the page add:  
 $= 4.6$   
line 10, change 0.9 to 0.7.
16. p 31, line 12, change G. R. Seaborg to G. T. Seaborg  
change furnished to furnished  
line 21, delete the word "to"

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Insert I, 1

(for top p. 18)

Before this formula is applied to the calculation of the ratio  $N_{40}/N_{49}$  in a rod of material irradiated in the Hanford pile, the following points must be considered:

- (1) An estimate of the effect of the destruction of  $\text{Pu}^{239}$  and  $\text{Pu}^{240}$  must be made.
- (2) A correction must be made for the non-uniformity of the neutron flux over the length of a rod and from outside to center within a rod.
- (3) Values used for  $\sigma_r(28)$  and  $\sigma_r(49)$  must take into account resonance capture of neutrons in  $\text{U}^{238}$  and  $\text{Pu}^{239}$ .

Equation (e) may be rewritten as

$$(g) \quad \frac{N_{40}}{N_{49}} = \frac{1}{2} \frac{N_{49}}{N_{28}} \frac{\sigma_r(49) \text{ eff.}}{\sigma_r(28) \text{ eff.}} \beta \delta$$

where  $\delta$  = correction factor for destruction of  $\text{Pu}^{239}$  and  $\text{Pu}^{240}$ ,  
and  $\beta$  = correction factor for non-uniformity of neutron flux.

(1) Calculation of the correction factor  $\delta$  for destruction of  $\text{Pu}^{239}$  and  $\text{Pu}^{240}$ :

$$\begin{aligned} \text{Let } N_1 &= N_{49}/N_{28} \\ N_2 &= N_{40}/N_{28} \\ \lambda_1 &= nv\sigma(49), \sigma(49) = \sigma f(49) + \sigma_r(49) \\ \lambda_2 &= nv\sigma(40), \sigma(40) = \sigma f(40) + \sigma_r(40) \\ k &= nv\sigma_r(28), \sigma_r(28) = \text{effective (thermal + resonance) radiative capture cross section of } \text{U}^{238}. \\ d &= \frac{\sigma_r(49)}{\sigma + (49)} \end{aligned}$$

The differential equations for  $N_1$  and  $N_2$  are:

$$(1) \quad dN_1/dt = k - \lambda_1 N_1$$

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I,2

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$$(2) \quad dN_2/dt = \frac{\alpha}{1+\alpha} \lambda_1 N_1 - \lambda_2 N_2$$

For  $N_1$  and  $N_2$  initially zero, the solutions for  $N_1$  and  $N_2$  are:

$$(3) \quad N_1 = \frac{k}{\lambda_1} (1 - e^{-\lambda_1 t})$$

$$(4) \quad N_2 = \frac{\alpha}{1+\alpha} k \left( \frac{1 - e^{-\lambda_2 t}}{\lambda_2} + \frac{e^{-\lambda_2 t} - e^{-\lambda_1 t}}{\lambda_2 - \lambda_1} \right)$$

The ratio  $N_{40}/N_{49}$  is:

$$(5) \quad N_2/N_1 = \frac{\alpha}{1+\alpha} \lambda_1 \frac{1 - e^{-\lambda_2 t}}{\lambda_2 (1 - e^{-\lambda_1 t})} + \frac{e^{-\lambda_2 t} - e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1) (1 - e^{-\lambda_1 t})}$$

In the special case of  $\lambda_1 t$  and  $\lambda_2 t$  very small, these equations reduce to:

$$(3') \quad N_1 = kt = nvt \sigma_r(28)$$

$$(4') \quad N_2 = \frac{\alpha}{1+\alpha} k \left( \frac{1}{2} \lambda_1 t^2 \right) = \frac{1}{2} (nvt)^2 \sigma_r(49) \sigma_r(28)$$

$$(5') \quad N_{40}/N_{49} = N_2/N_1 = \frac{\alpha}{1+\alpha} \left( \frac{1}{2} \lambda_1 t \right) = \frac{1}{2} \frac{\sigma_r(49)}{\sigma_r(28)} \frac{N_{49}}{N_{28}}$$

Equation (5') is the approximate equation (e) already given.

In the special case  $\lambda_2 = 0$ , i.e.  $\sigma(40) = 0$ ,

the equations are

$$(3'') \quad N_1 = \frac{k}{\lambda_1} (1 - e^{-\lambda_1 t})$$

$$(4'') \quad N_2 = \frac{\alpha}{1+\alpha} \frac{k}{\lambda_1} (e^{-\lambda_1 t} - 1 + \lambda_1 t)$$

$$(5'') \quad N_2/N_1 = \frac{\alpha}{1+\alpha} \left( \frac{\lambda_1 t}{1 - e^{-\lambda_1 t}} - 1 \right)$$

In the special case  $\lambda_2 = \lambda_1$ , i.e.  $\sigma(40) = \sigma(49)$ , the equations are:

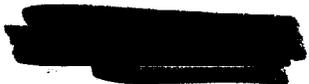
$$(3''') \quad N_1 = \frac{k}{\lambda_1} (1 - e^{-\lambda_1 t})$$

$$(4''') \quad N_2 = \frac{\alpha}{1+\alpha} \frac{k}{\lambda_1} (1 - e^{-\lambda_1 t} - \lambda_1 t e^{-\lambda_1 t})$$

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I,3

$$(5''') \quad N_2/N_1 = \frac{\alpha}{1+\alpha} \left( 1 - \frac{\lambda_1 t e^{-\lambda_1 t}}{1 - e^{-\lambda_1 t}} \right)$$

To calculate  $N_2/N_1$  for a typical production case, the following data may be used:

$$nv = 1.0 \times 10^{13} \text{ (a) or } nv = 2.0 \times 10^{13} \text{ (b)}$$

$$t = 94 \text{ days}$$

$$\sigma_{r(28)} = 3.5 \times 10^{-24} \text{ cm}^2$$

$$\sigma(49) = 1070 \times 10^{-24} \text{ cm}^2$$

$$\alpha = 0.49, \quad \frac{\alpha}{1+\alpha} = 0.329$$

Case (a) gives  $N_{49}/N_{28} \approx 300$  ppm.

Case (b) gives  $N_{49}/N_{28} \approx 600$  ppm

Calculation of  $N_{40}/N_{49} = N_2/N_1$  by equations (5'), (5''), and (5''') gives the following results:

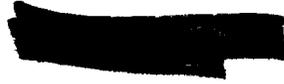
	$\lambda_1 t \lambda_2 t$ very small (5')	$\lambda_2 = 0$ ( $\sigma(40) = 0$ ) (5'')	$\lambda_2 = \lambda_1$ ( $\sigma(40) = \sigma(49)$ ) (5''')
(a) $N_{40}/N_{49} =$	$1.62 \times 10^{-2}$  ( $nv = 1.0 \times 10^{13}$ )	$1.65 \times 10^{-2}$	$1.60 \times 10^{-2}$
(b) $N_{40}/N_{49} =$	$3.24 \times 10^{-2}$  ( $nv = 2.0 \times 10^{13}$ )	$3.41 \times 10^{-2}$	$3.13 \times 10^{-2}$

It has been shown<sup>I</sup> that for thermal neutrons  $\sigma_f(40)$  is zero or a small fraction of  $\sigma_f(49)$ . Recent experiments<sup>II</sup> indicate a value of from  $40 \times 10^{-24}$  to  $500 \times 10^{-24}$  cm<sup>2</sup> for

<sup>I</sup> Farwell and Kahn, L.A. 444; also recent Chicago work.

<sup>II</sup> Personal communication, W. Manning, Metallurgical Laboratory.

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$\sigma_r(40)$  for thermal neutrons, so that the total cross section  $\sigma(40) = \sigma_f(40) + \sigma_r(40)$  must be less than about  $800 \times 10^{24} \text{ cm}^2$  and may be very small. Reference to the above table of values of  $N_{40}/N_{49}$  indicates that for material in which  $N_{49}/N_{28} \simeq 300 \times 10^{-6}$ , the correction factor  $\delta$  is somewhere between 1.02 ( $\sigma(40) = 0$ ) and 0.99 ( $\sigma(40) = \sigma(49)$ ). For the materials on which data are given in this paper, the highest ratio  $N_{49}/N_{28}$  is  $233 \times 10^{-6}$ , so that using the value  $\delta = 1.00$  can introduce no appreciable error into the calculation of the average ratio  $N_{40}/N_{49} \frac{N_{49}/N_{28}}{\dots}$  for the materials investigated.

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Insert II,1

(for pp. 22-23)

(3) a. Calculation of  $\sigma_{r(49)}$  (effective):

The effective capture cross section of  $\text{Pu}^{239}$  for neutrons in a pile may be considered as the sum of the capture cross section as measured at thermal energies and an additional contribution due to resonance capture, principally in the region of a few tenths of an electron volt.

It may be expressed as

$$\sigma_{r(49)} \text{ eff.} = \sigma_{r(49)} \text{ thermal} + \frac{\lambda}{\Lambda \xi} \int \sigma(E) \frac{dE}{E} \times \frac{1}{D}$$

where  $\lambda$  = scattering mean free path,

$\Lambda$  = absorption mean free path (thermal),

$\xi$  = average logarithmic energy decrement for neutron scattering,

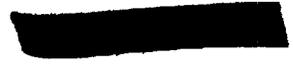
$D$  = disadvantage factor =  $\left( \frac{\overline{nv} \text{ (graphite)}}{\overline{nv} \text{ (uranium)}} \right)^{-1}$ .

and where the integral  $\int \sigma(E) \frac{dE}{E}$  is taken over the energy ranges in which appreciable resonance capture occurs.

The principal contribution to the resonance integral is given by an integration over the resonance occurring at about 0.3 electron volts. From data given in LA-266 on the total (capture plus fission) cross section of  $\text{Pu}^{239}$  in this region, the radiative capture cross section can be calculated and plotted as a function of neutron energy. A numerical integration gives a value of  $1470 \times 10^{-24} \text{ cm}^2$  for  $\int \sigma(E) \frac{dE}{E}$ ,

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II,2

which becomes  $1130 \times 10^{-24} \text{ cm}^2$  when corrected for the  $1/v$  tail of the thermal cross section.

The disadvantage factor D is given in CL697 as  $1/2.005$  for the W pile. Estimating 5% as the dip in the average resonance neutron flux in the metal rods as compared to the graphite, the disadvantage factor becomes  $1/1.905$ .

$\sigma_r(49)$  is given in CL-697 as  $350 \times 10^{-24} \text{ cm}^2$  for thermal neutrons.

Taking  $\lambda = 2.6 \text{ cm}$ ,  $\Lambda = 313 \text{ cm}$ ,  $\xi = 0.158$ , the resonance correction is found to be  $\frac{2.6}{313 \times 0.158} (1130 \times 10^{-24})$  (1.905) or  $113 \times 10^{-24} \text{ cm}^2$ .

$$\sigma_r(49) \text{ eff. is then } (350 + 113) \times 10^{-24} = \underline{463 \times 10^{-24} \text{ cm}^2}.$$

(3) b. Calculation of  $\sigma_r(28)$  (effective):

This calculation is made in the same manner as the calculation for  $\sigma_r(49)$  eff. However, the very heavy absorption of resonance neutrons by the surface layers of the uranium must be taken into account. The integral  $\int \sigma(E) \frac{dE}{E}$  is about  $240 \times 10^{-24} \text{ cm}^2$ .<sup>III</sup> Its effective value for lumps or rods or uranium is<sup>IV</sup>

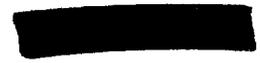
$$\int \sigma(E) \frac{dE}{E} = 9.25 \times 10^{-24} \text{ cm}^2 (1 + 8/3 S), \text{ where } S = \frac{\text{surface (cm}^2\text{)}}{\text{mass (gm)}}.$$

For the W pile the resonance contribution to the capture cross section is

$$\frac{\lambda}{\Lambda \xi} \int \sigma(E) \frac{dE}{E} \text{ eff.} \times 1/D = \left( \frac{2.6}{313 \times 0.158} \right) (9.25 \times 10^{-24}) (1 + 0.164)(2.005)$$

III Anderson and Fermi, A-2

IV Plass and Wigner, CP-372



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Insert II,3  
 or  $1.14 \times 10^{-24} \text{ cm}^2$ .

$\sigma_{r(28)}$  is given in LA-140A as  $2.56 \times 10^{-24} \text{ cm}^2$ .

Then  $\sigma_{r(28) \text{ eff.}} = 10^{-24}(2.56 + 1.14) = 3.70 \times 10^{-24} \text{ cm}^2$  for the W pile. That this method of calculation is a reasonable one is established by the following comparison of calculated and experimental results:

Data on the ratio of capture to fission in Clinton pile uranium are reported by Engelkmeir and Freedman<sup>V</sup> and by I. Perlman<sup>VI</sup>. Estimates from fission product and plutonium assays of slugs of metal irradiated in the X pile give for the ratio of capture to fission values of  $0.93 \pm .10^V$  and  $0.883^{VI}$ . A value of  $\frac{\text{capture}}{\text{fission}} = 0.90 \pm .06$  seems reasonable.

The slow neutron fission cross section of normal uranium is  $(542 \times 10^{-24} \text{ cm}^2)^{VII} \times 1/140$  or  $3.87 \times 10^{-24} \text{ cm}^2$ . Assuming 3% of the total fissions to be due to fast neutron fission<sup>VIII</sup> in  $U^{238}$ , this gives for the effective thermal neutron capture cross section of  $U^{238}$ .

$$\sigma_{r(28) \text{ eff.}} = (3.87 \times 10^{-24}) (0.90 \pm .06) (1.03)$$

<sup>V</sup> Engelkmeir and Freedman, CN-1917

<sup>VI</sup> I. Perlman, CN-2044

<sup>VII</sup>  $\sigma_f(U^{235})$  as given in LA-140A.

<sup>VIII</sup> CL-697

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Insert II,4

or  $(3.6 \pm 0.2) \times 10^{-24} \text{ cm}^2$  Calculation of  $\sigma_r(28)$

for the X pile by the method outlined for the W pile gives

$$\sigma_r(28) = (2.56 + 1.0) \times 10^{-24} \text{ cm}^2 = 3.56 \times 10^{-24} \text{ cm}^2,$$

in good agreement with the value calculated from the experimental data.

The ratio  $N_{40}/N_{49}$  can now be calculated from equation

(g):

$$\begin{aligned} \frac{N_{40}}{N_{49}} &= \frac{1}{2} \frac{N_{49}}{N_{28}} \frac{\sigma_r(49) \text{ eff.}}{\sigma_r(28) \text{ eff.}} \beta \delta \\ &= \frac{1}{2} \frac{N_{49}}{N_{28}} \frac{(463 \times 10^{-24})}{(3.70 \times 10^{-24})} (1.14) (1.00) \end{aligned}$$

$$(g') \quad \frac{N_{40}}{N_{49}} = 71.4 \frac{N_{49}}{N_{28}}$$

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Insert III, 1  
(for p. 25)

Reference to equation (g') which gives

$$\frac{N_{40}}{N_{49}} = 71.4 \frac{N_{49}}{N_{28}}$$

shows the experimentally determined constant 70.9 to be in excellent agreement with the calculated constant 71.4. The agreement is in fact closer than one has a right to expect, considering the uncertainties present in the calculation of the effective capture cross sections of Pu<sup>239</sup> and U<sup>238</sup> and the possibility of fairly large errors in the Pu<sup>239</sup>/U assays made at Hanford, as well as the experimental uncertainties of the spontaneous fission measurements.

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