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ABSOLUTE YIELD OF DELAYED NEUTRONS FROM PLUTONIUM FISSION, WITH FAST-NEUTRON EXCITATION

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ABSTRACT

The number of delayed neutrons per second per fission has been measured in an absolute sense, as a function of time, for plutonium irradiated for relatively short times with high-energy neutrons. The experimental results can be represented for an infinitely short irradiation time by the expression

$$Y(t) = (0.32\, e^{-t/79.9} + 0.77\, e^{-t/32.4} + 2.57\, e^{-t/7.9} + 8.26\, e^{-t/2.5} + 17.85\, e^{-t/0.45})\times 10^{-4} \text{ delayed neutrons per second per fission.}$$

The total yield of delayed neutrons from plutonium excited by fast neutrons has been found to be

$$Y(\text{total}) = 6.8\ (0.4)\times 10^{-3} \text{ delayed neutrons per fission.}$$

This is 0.23 o/o of the number of prompt neutrons from plutonium fission. The value is considerably lower than that obtained from slow-neutron excitation of plutonium. However there is no conclusive evidence that the difference in yield is attributable to the difference in the primary-neutron energy.

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ABSOLUTE YIELD OF DELAYED NEUTRONS FROM PLUTONIUM FISSION, WITH FAST-NEUTRON EXCITATION

INTRODUCTION

One of the methods used to determine the nuclear efficiency of the Trinity test bomb involved measurement of the intensity of delayed neutrons emitted from the fission products as a function of time.(1) In order to convert the results of the measurements into nuclear efficiency, it is necessary to know the yield of delayed neutrons from plutonium fission as a function of time, particularly over the interval of time covered by the measurements.

Some measurements had been made, as a function of time, of the relative numbers of delayed neutrons from Pu fission, and of the delayed-neutron yield relative to the yield from U-235 fission.(2) However, these measurements were made using slow neutrons to induce fission. Furthermore, the Pu was irradiated to equilibrium activity; and consequently the shorter periods could not be determined. Except for the work of Wilson and Sutton, no measurements were made during the first second after the slow-neutron irradiation.

~~Inasmuch as the plutonium bomb has a high-energy-neutron spectrum,(3) and since the~~ effective irradiation time in the bomb is short, it was considered ~~advisable~~ of interest to determine the delayed neutron yield from Pu fission induced by fast neutrons, particularly during the first few seconds after a short irradiation of the plutonium.

The results of these measurements discussed in Part I indicated the desirability of doing a further experiment to determine (a) the total yield of delayed neutrons from Pu, and (b) the shape of the decay curve over a longer interval than was possible in the first measurements.

~~(1) Blair, J. M.; Frisch, D. H.; and Richards, H. T.; LA 376.~~
(2) Wilson, R. R., and Sutton, R. B. LA 76
Redman, C. and Saxon, D., CF 2318
Feld, B.T. and de Hoffmann, F. LA 251
~~(3) Richards has found that the prompt neutron spectrum from plutonium fission is very nearly the same as that from U-235 fission (LAMS 222, p.8). The latter has an average energy of 2.6 Mev, as determined by Richards. (See LA 200).~~ [redacted]

Part I

The measurements were made using the 2.5- Mev electrostatic generator in W building as a modulated source of high-energy neutrons. For intensity reasons, it was necessary to use a lighium target of about 200-Kev stopping power. Consequently the primary neutrons had energies ranging roughly from 400 to 600 Kev, weighted, however, toward the higher energy, becasue of the resonance in the forward direction yield from the $Li^7 (p,n,) Be^7$ reaction, the maximum of which is at about 600-Kev neutron energy.[4]

The plan of operation was as follows: (1) to irradiate the plutonium with neutrons from the lithium target for a short time T_k, by allowing the proton beam to fall on the target; (2) to measure the fissions induced in the plutonium by the primary irradiation; and (3) to measure the delayed neutrons emitted from the Pu during successive intervals of time after the proton beam was cut off by the modulator.

FISSION MEASUREMENTS:

The plutonium, comprising 118 grams of material ~~from the Clinton~~ pile, was prepared in the form of two short cylinders, each 1.0" diameter by 1/4" long. ~~These were protected by a silver coating.~~ One of the cylinders was placed on an insulating support in an ionization chamber, (Fig. 1), and connected to the collector voltage supply. Opposite this cylinder, and separated from it by about 1/16" was a Pt foil carrying an additional 758 micrograms of Pu to serve as a fission monitor. The other cylinder was then placed behind, and in contact with the foil, and the combination served as the ground electrode of the fission chamber. The chamber was filled with argon to 25 lbs pressure.

(4) See $Li^7 (p,n) Be^7$ yield curve, LA 140, Fig. 28 [redacted]

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Fission pulses from the chamber were fed through an Elmore Model 500 fast pre-amplifier and amplifier (0.2 microsecond clipping time), to a thyratron pulse size discriminator and scale of 64 scaler (see Fig. 2).

The counting efficiency of the chamber was found to be 0.975 $\pm$ .005 by extrapolation of the bias curve to zero pulse size.

The fission chamber was so placed in front of the Li target that neutrons emitted from the target at zero angle would pass through the cylinders and fission foil along the axis of symmetry (Fig.3). Consequently the flux through the foil between the two cylinders will be the mean of the flux averaged over the total volume of the Pu. The average number of fissions per unit mass in the two cylinders is then the same as in the foil, so that the total number of fissions is the mass ratio multiplied by the number of fissions in the foil.

DELAYED-NEUTRON DETECTORS:

Because of the low intensity it was necessary to use sensitive detectors for the delayed activity. For this purpose, two specially sensitive long counters were used. These were 1.0" diameter by 8" long BF_3 proportional counters, filled to 25 cm of Hg with enriched BF_3 (85 o/o B^{10}). The counter was encased in an 8" diameter by 10" long paraffin cylinder, in the usual long counter design.[5] The counting efficiency was about 1 o/o for neutrons incident on the active end of the counter.

These counters were placed as near as possible to the plutonium, (Fig.2). Pulses from the counters were fed through two Sands Model 100 pre-amplifiers and amplifiers to a pulse size discriminator, and thence to a mechanical time discriminator and a set of scalers arranged to count pulses occurring during specified intervals of time after irradiation of the plutonium. The pulses from the discriminator were of uniform size, and at such a high level that electrical pick-up was not a problem, and scales response was uniform.

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(5) Hanson, A. O., LA 276.

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The counting efficiency of the long counters was determined in the geometry used in the delayed-neutron measurement, by placing a natural neutron source (M. F. No. 1) in the normal position of the plutonium, and determining the counting rate in the two counters. The neutron flux from M. F. No. 1 is believed to be known to about 5 o/o,(6) so that the efficiency of the counters for M. F. No. 1 neutrons was determined to approximately that accuracy. However, the source has a high energy spectrum, the maximum in the yield-energy curve being in the region of 1 to 2 Mev,(7) whereas the delayed neutrons from Pu fission probably have energies in the region from 200 to 700 Kev(8). Nevertheless, tests made on the energy characteristics of the Long Counters, in the experimental geometry, indicate that they have an essentially constant detection efficiency from M. F. No. 1 energies down to 100 to 200 Kev. It is therefore believed that the efficiency, as determined by the source M. F. No. 1, is reliable to about 1 o/o for the delayed neutron spectrum.

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(6) The source M. F. No. 1 was compared with a standard Ra + Be source No. 40 by Robert L. Walker. Both a water bath and a graphite column were used in making the comparison. The flux relative to the standard source was determined to an accuracy of 1 o/o. The standard source is believed to be known in an absolute sense to about 5 o/o.

(7) Richards, H. T., (LA 201) gives 1.9 Mev for the average energy.

(8) This is based on the assumption that since the delayed neutron periods for plutonium are essentially the same as for U-235, (L.A. 76; C. K. 2318; L.A. 231), the neutrons are probably emitted by similar fission fragments, and therefore have the same energies as those from U-235. Hughes, Dabbs, and Cahn have measured the delayed neutron energies for U-235, and find that they range from 250 Kev to 620 Kev. (C.P. 3094).

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COUNTER SHIELDING:

With the sizeable masses of paraffin of the long counters located near to the fission chamber, it was necessary to shield the latter from primary neutrons which undergo energy degradation in the paraffin and later diffuse out. Aside from the fact that such neutrons would not fulfill the requirements for fast excitation of fission, they would not be properly monitored by the fission foil, because of their short range in plutonium. The fission chamber was therefore covered with a cadmium shield, plus a 5/16" layer of enriched boron powder, (82 o/o B^{10}), so that only high-energy neutrons could reach the plutonium in appreciable numbers.

In order to keep the long-counter-background counting rate down to a reasonable value, it was found necessary to eliminate, or to shield the counters from several sources of stray neutrons. A rather large background was found to arise from the proton beam when it was allowed to fall upon copper or brass parts in the target tube, due either to (p,n) reactions in these metals, or perhaps to small quantities of lithium on their surfaces. Wherever possible, such parts were either covered with, or replaced by graphite. However, some parts could not be so protected. Furthermore, there was a possibility that the small amount of deuterium in the normal hydrogen used in the ion source could give rise to an appreciable neutron yield from the C^{12} (d,n)N^{13} reaction; and since all parts of the vacuum system upon which the beam falls are always coated with a carbon deposit from diffusion pump oil, it was necessary to shield the Long Counters from all such sources of neutrons. For this purpose, a thick paraffin wall, followed by a 2-inch layer of B_4C, was interposed between these parts and the counters.

MODULATION EQUIPMENT:

Fig. 3 is a vertical section through the target tube, showing the deflector plates (D), mechanical shutter (B), defining aperture (A), and lithium target (T). Normally the proton beam from the electrostatic generator passes through the wide

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slit (S), between the deflector plates, and through the aperture to the lithium target, from which neutrons are emitted. When a positive potential of about 2 kilovolts is applied to the upper deflector plate, the beam is deflected to some point below the aperture, thus cutting off the neutron yield from the target. The mechanical shutter was raised during the counting interval in order to cut off a background due to protons scattered from the slit (S) at such an angle that they passed through the aperture and to the target when the deflector voltage was on.

The modulator circuit, which controls the potential of the upper deflector plate, is shown in Fig. 4. The first two stages were designed primarily for use in other experiments, and are essentially a triggering buffer stage and a variable frequency relaxation oscillator. For the present work, the bias of the 884 thyratron was set to prevent oscillation, and the trigger signal was supplied from another source. The third element is a uni-vibrator which puts out a square wave, or gate, the width of which is variable by means of the variable resistor and condenser in the first grid circuit of the 6SN7. This gate width determines the on-time (T_k) of the proton beam, and therefore the length of the irradiation period. The triode part of the fourth element holds the grid of the gammatron at cut-off except during the passage of the gate signal. The diode is to prevent overshooting of the gammatron. The plate of the gammatron is normally at a potential of 2 Kv, but is reduced to approximately ground potential by the gate signal. The rise time of the deflector plate potential is of the order of a few microseconds.

TIME DISCRIMINATOR:

The essential features of the mechanical time discriminator are shown schematically in Fig. 5. The main parts consist of a set of 12 separate slip rings, mounted on an insulating cylinder, and a non-conventional type commutator, having 12 segments, each of which is connected electrically to one of the slip rings. The

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commutator-slip ring assembly is supported on a shaft, and driven by a synchronous motor through a worm gear of such ratio that the time spent by the commutator brush on each segment is of a specified length, Δt. The commutator segments, which are small brass plates screwed to facets milled into a lucite cylinder, were so machined that the effective angle subtended by each at the central axis is quite accurately the same for all segments. The brush was so shaped that there is a very short dead time between segments. The purpose of the secondary spring and the gum rubber pad is to introduce additional vibration periods into the brush assembly in order to prevent chattering of the contact with the commutator segment, which would result in counting losses. Each of the slip rings is connected through a brush contact to a scaler, or as otherwise indicated.

Two arms, or cams, mounted on one end of the shaft, operate micro-switches S_4 and S_5 at specified times during each revolution of the commutator. Another set of cams, driven by the shaft through a reduction gear, operates a set of three micro-switches, S_1, S_2, and S_3, during one, out of a fixed number of revolutions of the commutator. The purpose of these switches can be understood most easily by referring to Fig. 6 which is a graphical representation of the sequence of events. Time is there plotted along the horizontal axis, and each event in the sequence is indicated in relation to the time at which the commutator brush first makes contact with successive segments No. 1 to No. 12, as indicated by the vertical line above the time axis to the left of each number.

The first event in each sequence is the closing of S_1 and S_2, and the opening of S_3, which occurs while the brush is in contact with segment No. 11. The closing of S_1 completes the circuit from the pulse size discriminator to the commutator brush (Fig. 5). Hence, pulses from the long counters which trigger the thyratron of the discriminator are transmitted to the commutator, and also to the totalizing scaler. The latter, however, records no pulses during this interval, because slip-ring No. 11 is grounded. When segment No. 12 makes contact with the brush, the background scaler

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and also the totalizer are operative. As soon as segment No. 12 is cleared by the brush, S_4 operates, opening the shutter control circuit, and allowing the mechanical shutter to drop. Immediately afterward, S_5 closes momentarily, completing the circuit from the battery, through S_3, to the modulator circuit. This triggers the gate, which allows the proton beam to fall on the target for a time, T_k. The irradiation period ends before the brush clears segment No. 1, and the shutter switch, S_4, closes immediately afterward, raising the shutter in the target tube so that no scattered protons may reach the target during the delayed neutron counting period. Note that No. 1 slip-ring is grounded, so that the totalizer records no counts from the primary neutrons.

The zero of time on the plot is at the end of the irradiation period. Beginning at a time δt, the nine counting channels become operative in succession, each for an interval Δt. The totalizer records the sum of the counts in all channels, and gives a check on counting losses due to commutator or scaler difficulties.

When the brush clears the last counting channel, (segment No. 10), it returns to segment No. 11. During this interval, S_1 and S_2 open, and S_3 closes, so that the sequence cannot be repeated immediately. The reason for this is to allow the long period delayed activity in the plutonium to decay to such a value that the background counting rate will not increase appreciably with time. Furthermore, it will insure that the background rate, measured immediately before the irradiation period, will not change significantly during the following counting period, since the long period activity which remains will be essentially constant during the relatively short counting interval.

The delayed-neutron counting rate was such as to give only a few counts per channel during each counting cycle. It was necessary, therefore, to repeat the cycle a large number of times for statistical accuracy.

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NEUTRON MULTIPLICATION:

One major correction must be made to all the data. This arises from the fact that neutron multiplication occurs in any mass of plutonium. In order to get an estimate of this factor, the following measurements were made: (1) a neutron source of small dimensions (M.F. No. 1) was placed between the long counters, in the normal position of the plutonium, and the counting rate was determined. (2) The source was placed between the Pu Cylinders, (again in their usual position, except separated 1/4 inch). The counting rate was found to be higher than for the bare source by a factor 1.16. (3) With the source between the cylinders, as in (2), the cylinders were turned perpendicular to their normal position, so that the axis of symmetry was parallel with a line between counters. The counting rate ratio to bare source was 1.09. (4) With the cylinder separation again 1/4 inch, the source was placed at the outer end of one cylinder, with the cylinders in their usual position. The counting rate ratio was 1.12. (5) With the source as in (4), and the cylinder separation at the normal 1/16 inch, the counting ratio was 1.13.

On the basis of these measurements, and using single collision theory, Messrs. Gass, Lax, and Schiff calculated the multiplication for a distributed source in the geometry of the plutonium[9]. The multiplication factor thus derived is 1.15 ± .01. All delayed neutron yields must therefore be reduced by this factor.

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(9) Gass, Lax, and Schiff. LAMS 306.

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EXPERIMENTAL DATA

Three separate runs were made, using the time values shown in Table I, where all times are in seconds. The subscript in the term T_k refers to the run number.

TABLE I

Run Number	Irradiation time T_k	Time when counting began δt	Channel width ΔT	Total counting time	Cycle time
1	0.971	0.0112	1.0	9.0	120
2	0.479	0.0087	0.5	4.5	60
3	0.0838	0.010	0.1	0.45	24

The results of run No. 3 revealed a systematic error in the first three counting intervals, which was found to be due to the fact that during the primary irradiation, the ionization in the long counters was so great that the collector voltage was considerably reduced, thus lowering the gas amplification to such a level that the counte were driven off the bias plateau. This was corrected by removing a series resistor in the filtering circuit of the high voltage supply, so that the voltage at the counte could recover immediately after the primary neutrons were cut off. Inasmuch as run No.3 required a great deal of time, a repitition of the run was made on a relative basis only, i.e. by removing the plutonium from the fission chamber, in order to increase the delayed counting rate by improved geometry. Under these conditions, the shape of the decay curve was determined with good statistical accuracy; and on the basis of this, the data of the first three counting intervals of run No. 3 were corrected. Since the shape of the two curves was very closely the same over the remaining portion, it is believed that the corrected points are probably as reliable as other points of

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Although the first counting interval of each of the runs No. 1 and No. 2 was affected by the counter difficulty, the runs were not repeated. However, these points were disregarded in attempting to fit the data by a mathematical expression. It turns out that for run No. 1 the first point falls above the final curve; whereas for run No.2 the first point is considerably below. In estimating the probable limits of error for the latter, a factor is included which is based on the correction found necessary for run No. 3, which is sufficient to raise the point to a reasonable value. Such a correction is not considered accurate because the magnitude of the effect of the primary neutron on the counter response will depend upon how much the gas amplification of the counter is reduced with respect to its normal value, i.e. how far off the bias plateau the counter is driven. In general it will depend in a non-linear manner upon the intensity of the neutron flux, which is proportional to the proton beam current. It was not possible to measure the latter under the experimental conditions; consequently its value for a particular run is not known. However, it is known that the beam current varied by a factor of 2 from one run to another, which may partially explain why run No. 2 and No. 3 were apparently affected more than run No. 1.

The data from the three runs are presented in Table II, where the yield, $Y_k(t)$, is the number of neutrons per second per fission, for each of the nine counting intervals of run No. k. The values were calculated from the expression

[redacted]

TABLE II

Delayed Neutron Yield - Neutrons per Second per Fission

Counting Interval	$Y_1(t)$	$Y_2(t)$	$Y_3(t)$
1	11.62×10^{-4} $\pm .70$	14.40×10^{-4} $\pm .95$	25.55×10^{-4} ± 1.76
2	$6.81 \pm .41$	$10.85 \pm .65$	22.27 ± 1.53
3	$5.07 \pm .31$	$8.26 \pm .50$	19.55 ± 1.33
4	$3.96 \pm .24$	$6.72 \pm .41$	17.13 ± 1.17
5	$3.17 \pm .20$	$5.63 \pm .34$	15.78 ± 1.08
6	$2.61 \pm .16$	$4.96 \pm .30$	14.26 ± 0.99
7	$2.33 \pm .15$	$4.61 \pm .28$	$12.76 \pm .89$
8	$1.87 \pm .12$	$3.88 \pm .24$	$11.85 \pm .83$
9	$1.56 \pm .10$	$3.32 \pm .21$	$11.55 \pm .82$
Counting Losses	0.6 o/o	0.5 o/o	2.6 o/o

(1) $$Y(t) = \frac{N(t)}{\varepsilon\ \Delta t}\ \frac{e}{F}\ \frac{m}{M}\ \frac{1}{C}$$

where N(t) is the number of delayed neutron counts in a time Δt, corresponding to each interval; ε is the counting efficiency of the long counters; e is the counting efficiency of the fission chamber; F is the total number of fission counts for run k; m is the mass of Pu in the fission foil; M is the mass of the two Pu cylinders; and C is the neutron multiplication factor.

The counting loss shown for each run is the percentage difference between the totalizing scaler and the sum of all channels. It is not certain that the same correction should be applied to all counting channels in a given run. However, as will be shown later, any error due to these losses in run No. 3 is eliminated in the final analysis

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of the data; and since the losses in the other two runs were small, no serious error arises in those cases.

ANALYSIS OF DATA - Part I

The values $Y_k(t)$, from Table II are plotted in Fig. 7. The curves drawn through the experimental points were calculated from the results of the analysis described later. As is to be expected, neither the slopes of the curves, nor the yields are the same for the three sets of data, since the irradiation time, T_k, is different for each. It is therefore necessary to obtain an expression which will represent the data, taking this factor into account.

The irradiation intervals, T_k, were neither long enough to saturate the delayed activity, nor yet short enough to be considered as instantaneous irradiation. Consequently, neither type of representation is possible. For infinitely short irradiation time, the delayed neutron activity would be of the form

$$(2) \quad A_1(t^*) = \sum_i A_{oi} \; e^{-t^*/\tau_i}$$

where (i) modes of decay are assumed to exist, each mode having an initial activity, A_{oi}, and a mean life, τ_i. For an irradiation time, T_k the magnitude of the activity will be

$$(3) \quad A_{T_k}(t^*) = \int_0^{T_k} A_o(t^* - \tau)\,d\tau = \sum_i A_{oi} \int_0^{T_k} e^{-(t^*-\tau)/\tau_i} \, d\tau = \sum_i A_{oi}\, \tau_i \, e^{-t^*/\tau_i} \left(e^{T_k/\tau_i} - 1\right), \quad (t^* > T_k).$$

where the zero of time is the beginning of the irradiation interval. It is convenient to make the zero of time coincide with the end of the irradiation period, by making the substitution, $t = t^* - T_k$, so that for $t = 0$, $t^* = T_k$.

The activity will then be represented by

$$(4) \quad A_{T_k}(t) = \sum_i A_{oi} \tau_i \left(1 - e^{-T_k/\tau_i}\right) e^{-t/\tau_i} \quad \therefore \quad A_{T_k}(0) = \sum_i A_{oi} \tau_i \left(1 - e^{-T_k/\tau_i}\right).$$

In order that the expressions (3) and (4) may be dimensionally the same as (2), the

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term τ_1, which arises from the integration in (3) should be considered as a non-dimensional constant.

The only available data on delayed neutrons from plutonium indicates that there are four modes of decay, the periods of which were found to be practically identical with those found for the delayed neutrons from U-235.[10] More recent data on the delayed activity from U-235 has been resolved into five modes of decay.[11] It is therefore assumed that the same five periods exist for plutonium, with perhaps different relative intensities than were found for U-235. In view of the fact that the counting time in the present work extended only to 9 seconds, it is not possible to determine the periods and relative intensities directly from the data. Moreover, in the experiment described under Part II, the background counting rate was too high to allow an accurate determination of the longer half-lives directly. It was therefore decided to try to fit the data by using the values for the decay periods found by de Hoffmann for U-235, and adjusting the coefficients where necessary.

In Table III, the mean lives, and relative activities corresponding to irradiation to equilibrium, are reproduced from de Hoffmann's results for U-235.[12]

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(10) C. F. 2318; LA 231.
(11) de Hoffmann, F. LA 252. Hughes, D. J., Dabbs, J., and Cahn, A., C.P. 3094.
(12) L.A. 252, p.8

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TABLE III

Period	1	2	3	4	5
τ_i	79.9 ± 1.0	32.4 ± 0.6	7.9 ± 0.5	2.5 ± 0.3	0.52 ± 0.1
R_i	.054	.294	.297	.279	.076

where the values of R_i were determined by

$$(5) \qquad R_i = \frac{A_{oi}\tau_i}{\sum_i A_{oi}\tau_i} \qquad \text{and} \qquad \sum_i R_i = 1.$$

In order to determine the relative intensities of the two longer periods we have made use of the results of the second experiment described under Part II. Fig. 9 shows the shape of the decay curve from 0 to 250 seconds after the Pu had been irradiated to equilibrium activity. After 60 seconds, only the two longer periods have sufficient intensity to be detectable. At the end of four minutes the second period has almost disappeared. It is therefore possible to determine values for the coefficients of these two periods which will fit the experimental points to a fair approximation in the time interval from 60 to 160 seconds.

Now from equation (3) we obtain the following relation for the relative intensities of the periods after an irradiation time T_k:

$$(6) \qquad R_i(T_k) = \frac{A_{oi}\tau_i(1-e^{-T_k/\tau_i})}{\sum_i A_{oi}\tau_i(1-e^{-T_k/\tau_i})}, \qquad \sum_i R_i(T_k) = 1.$$

Since the value of R_i for saturated activation is proportional to $A_{oi}\tau_i$, from (5) and (6) we obtain a relation between the relative intensities for saturated activation and the relative intensities corresponding to irradiation time T_k.

$$(7) \qquad R_i\,(T_k) = \frac{R_i(1-e^{-T_k/\tau_i})}{\sum_i R_i(1-e^{-T_k/\tau_i})}$$

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Making use of (7), the relative coefficients of the two longer periods, corresponding to run No. 1, were determined. Then values of the relative coefficients for periods 3 and 4 were chosen in such a way as to give a reasonable fit to the experimental points of run No. 1 in the time interval 2 to 9 seconds, where the shortest period makes no contribution to the activity.

Again using relation (7) the relative coefficients of the first four periods were calculated for the case of run No. 3. With these values fixed, the relative value for period 5 was also fixed. However, it was found that the slope of the resulting curve was not sufficiently steep to fit the experimental points of run No. 3. Furthermore, the values for the intercept at t=0, could not be harmonized for the three runs, when normalized to the same irradiation time. It was therefore necessary to determine a new value for the mean life of period 5 which would give the proper slope to fit the data of run No. 3. The value thus obtained was τ_5 = 0.45 sec. which is within the 20 o/o uncertainty limits of de Hoffmann's value 0.52 ± 0.10.

Table IV shows the values of the mean lives, τ_i, and the relative intensities, R_i, of the five activities used in the final analysis, together with the relative values, $R_i(T_k)$ corresponding to each of the three runs, No. 1, No. 2 and No. 3.

TABEL IV

Period	1	2	3	4	5
τ_i	79.9	32.4	7.9	2.5	0.45
R_i	.033	.327	.265	.270	.105
$R_i(T_1)$	.0018	.0438	.1390	.3941	.4212
$R_i(T_2)$	.0014	.0352	.1143	.3450	.5041
$R_i(T_3)$	.0011$_4$	.0278	.0920	.2926	.5865

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Before proceeding to determine the actual coefficients for the individual activities, it is necessary to determine the value of the intercept at $t = 0$. This can be done most accurately by an integration which eliminates the counting loss error. If we designate the total activity at $t = 0$, by $A_o(T_3)$ for the case of run No. 3, the activity of each period, $A_i(T_3)$ will be given by

(8) $A_i(T_3) = A_o(T_3)\, R_i(T_3)$.

The total yield, Y_3(total), for the nine counting intervals of run No. 3, as determined from the totalizing scaler, will be equal to the integral of the sum of all activities, taken over the counting interval, 0.01 to 0.91 second. Using the right hand side of equation (8) for the actual coefficients, we have

$$(9) \qquad Y_3(\text{total}) = A_o\,(T_3) \int_{0.01}^{0.91} \sum_i R_i(T_3)\, e^{-t/\tau_i}\, dt$$

The value obtained from the totalizing scaler is Y_3 (total) $= 1.52 \times 10^{-3}$ neutrons/fission, so that we obtain from the calculation, (9), the initial intensity $A_o(T_3) = 27.77 \times 10^{-4}$ neutrons/second/fission. The values for $A_i(T_3)$ are then obtained from (8), using the values of $R_i(T_3)$ from Table IV. The results are given in Table V.

We wish now to find the actual coefficients corresponding to an infinitely short irradiation interval, but with the same total amount of induced activity as for run No. 3. If there were no decay during the irradiation period, the final intensity would be, for each mode, $A_{oi}T_k$, where A_{oi} is the activity induced in an infinitessimal irradiation time. The true activity, in terms of A_{oi} and T_k, is given by the coefficient in equation (4). Taking the ratio of these terms, and multiplying by the experimentally determined values, $A_i(T_k)$, will therefore give the correct coefficients for the case where all the activity is induced in a short irradiation time. Thus,

$$(10) \qquad A_{oi} = A_i(T_k) \frac{A_{oi}\, T_k}{A_{oi}\,\tau_i\,(1-e^{-T_k/\tau_i})} = A_i(T_k) \frac{T_k}{\tau_i\,(1-e^{-T_k/\tau_i})}$$

[redacted]

Using the values obtained for $A_i(T_3)$, the coefficients, A_{oi}, corresponding to short irradiation were calculated by the use of (10), and are shown in Table V, together with their sum, which is the total intensity, A_o, for short irradiation time. The relative values, R_{oi}, normalized to unity, are given in the last column of Table V.

The values for the actual coefficients corresponding to runs No. 1 and No. 2, can be found from either the values, A_{oi}, or $A_i(T_3)$, using equation (10). The results, together with their sums, are shown in Table V.

TABLE V

Period	$A_i(T_1)$ Neutrons/sec. per Fission.	$A_i(T_2)$ Neutrons/sec. per Fission.	$A_i(T_3)$ Neutrons/sec. per Fission.	A_{oi} Neutrons/sec. per Fission	R_{oi}
1	0.031×10^{-4}	0.31×10^{-4}	0.32×10^{-4}	0.32×10^{-4}	.0011
2	0.76	0.77	0.77	0.77	.0262
3	2.42	2.49	2.55	2.57	.0870
4	6.85	7.52	8.13	8.26	.2802
5	7.32	10.98	16.29	17.85	.6055
$A_o(T_k)$	17.37×10^{-4}	21.79×10^{-4}	27.77×10^{-4}	29.48×10^{-4} n/sec/f.	

From the values, $A_i(T_k)$, from Table V, the activity as a function of time was calculated for each of the irradiation times, T_k, using the relation

$$(11) \qquad A_k(t) = \sum_i A_i(T_k)\, e^{-t/\tau_i} .$$

The results are shown as the three curves in Fig. 7.

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In order to see more clearly how well the three sets of data match the analytical results, the activity as a function of time was calculated for short irradiation time, and the experimental points were normalized for this case by the relation

$$(12) \qquad Y'_k(t) = Y_k(t) \quad \frac{A_o(t)}{A_k(t)}$$

where the activities are evaluated for times, (t) appropriate to each of the experimental points. These results are plotted in Fig. 8. The Limits of error shown are the total probable error limits as discussed in Part I. The solid curve was calculated from the expression

$$(13) \qquad A_o(t) = \sum_i A_{oi}\, e^{-t/\tau_i}.$$

TOTAL DELAYED NEUTRON YIELD:

It is of interest to determine the total yield of delayed neutrons, by integration of equation (12) over infinite time. The result of this integration is given in Table VI, together with the integrated yields for 10,5, and 1 seconds, and the fraction of the total yield emitted during these times .

TABLE VI

Time in seconds	Delayed Neutron Yield per fission	Fraction of total Delayed Neutron Yield
	7.65×10^{-3}	1.0000
10	4.98	.6512
5	3.92	.5118
1	1.72	.2249

[redacted]

PART II

EXPERIMENTAL

The plan of operations in this experiment was (a) to measure the number of fissions per second occuring in the plutonium, during the time that it was irradiated by a constant flux of fast neutrons; (b) to determine the number of delayed neutrons per second emitted by the plutonium at the instant when the primary irradiation ended, by extrapolation of the decay curve to zero time; (c) to determine the shape of the decay curve over as long a time as possible after equilibrium irradiation.

The experimental arrangement was in general the same as that described in Part I. Only minor changes in the beam modulation control, and in the method of counting were necessary. The Long Counter efficiency, and the counting efficiency of the fission chamber were re-measured, and found to be very nearly the same as in Part I. The neutron multiplication was assumed to be the same as previously determined, since the geometry was the same.

The following sequence of operations was followed:

1. The proton beam was kept on the target for a period of 6 minutes.
2. During the last four minutes of irradiation, the fission counter was turned on, so that the average number of fissions per second was easily determined.
3. Beginning at t = 0.11, the scalers connected to the time discriminator recorded the delayed neutron counts for a period of 1 second. Five counting channels were used, each with a width of 0.2 second.
4. Beginning at t = 2.0 sec the delayed neutron counts were recorded manually, by reading a scaler at the end of fixed intervals. This was carried out to 300 seconds.
5. The background counting rate was measured during a 10 second interval at the end of the sixth minute after the beam was turned off.

The six-minute irradiation period was sufficiently ~~long~~ to saturate all the periods

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and the six minute off-time was sufficient for all the delayed activity to decay well below the background level.

Because of the fact that the high-voltage supply used with the long counters in Part I was not available, it was necessary to use another supply which had such characteristics that the long counters did not recover immediately after the beam was cut off. Hence it was not possible to count the delayed neutrons during the interval 0.01 to 0.01 sec. In fact the interval 0.11 to 0.21 appears to have been slightly affected by thi trouble, since the first experimental point is a few per cent below the curve shown in the insert on Fig. 9.

A total of 46 runs were taken, which gave upwards of 2500 counts per channel for each of the five channels in the 1-second interval. The statistical accuracy is therefore better than 2 o/o. The counting losses were about 1 o/o. In general the proton beam was fairly constant during a given irradiation period, so that the average number of fissions per second is given by the total number of fissions divided by the total counting time for all 46 runs. Nevertheless, in a few cases the beam decreased noticeably near the end of a run, below the average value. This would have an appreciable effect on the number of delayed neutrons recorded during the first second since the short periods would be reduced in intensity. It is not possible to determine accurately the magnitude of this effect. As a matter of fact there is also some compensation from the opposite effect of the beam being slightly above the average value at the end of the irradiation period. A reasonable estimate seems to be that in about 10 o/o of all cases the beam was more than 10 o/o below average and in some cases as much as 20 o/o below average during a few seconds before the irradiation period ended. The total net effect, uncompensated by the opposite effect, would then be between 1.0 o/o and 2.0 o/o, i.e. the number of delayed neutrons per second at t = 0 should be 1.5 o/o higher than indicated by the counters. The delayed neutron count has therefore been increased by 1.5 o/o for the channels in the 1.0 second counting interval.

[redacted]

The corrected data for the 1.0-second counting interval are given in Table VII.

TABLE VII

Counting interval	Delayed Neutrons per second.	Delayed Neutrons per Fission
1	319.3	6.07×10^{-3}
2	318.9	6.06
3	294.3	5.59
4	286.4	5.44
5	281.1	5.34

Average Fissions/second = 296.34

The values in the last column of Table VII were calculated from the expression

$$(14) \qquad n/f = \frac{N/\Delta t}{\varepsilon} \; \frac{e}{F/\Delta t} \; \frac{m}{M} \; \frac{1}{C}$$

where N is the number of delayed neutron counts in a time Δt; ε is the counting efficiency of the long counters; F is the number of fissions in a time Δt; e is the counting efficiency of the fission chamber; m is the mass of the Pu in the fission foil; M is the mass of Pu in the cylinders; and C is the neutron multiplication factor.

The values in the last column of Table VII are plotted on an expanded time scale in Fig.9 (insert), the limits of error being only the statistical limits based upon the number of counts. The solid curve drawn through the points was calculated on the basis of the relative intensities R_i, and the mean lives τ_i, given in Table IV, for the five decay periods.

TOTAL DELAYED NEUTRON YIELD:

Since all the activities are saturated at the end of the irradiation period, the rate of emission of delayed neutrons divided by the fission rate at the time t = 0, gives the number of delayed neutrons per fission. Hence, the total delayed neutron yield

[redacted]

is given by the intercept of the curve, Fig. 9, at t = 0. This value is found to be

$$Y = 0.0068 \pm .0004 \text{ delayed neutrons per fission.}$$

Taking the value (49) 2.95, (LA-140-A), we find that the total number of delayed neutrons is 0.23 o/o of the number of prompt neutrons.

The uncertainty in the above yield value is a composite of several uncertainties involved in the experiment. The largest uncertain value is in the absolute counting efficiency of the long counters, which depends ultimately upon the absolute neutron flux from the Ra + Be source No. 40 with which the M. P. No. 1 source was compared. The uncertainty here is of the order of 5 o/o. Hence the Long Counter efficiency is not known, with certainty to better than about 5.2 o/o. The counting efficiency of the fission chamber is uncertain to about 0.5 o/o. The mass of the fission foil is known to about 0.5 o/o. The neutron multiplication factor may be uncertain to about 2 o/o; and the statistical uncertainty in the delayed neutron counting rate is about 2 o/o. The total number of fission counts was of the order of 10^6, so no statistical uncertaint is included for this term. The mass of the Pu cylinders is given to three decimal place i.e. 118.034 grams. Hence the uncertainty is presumably small in this case. Taking the square root of the sum of the squares of these uncertainties gives 6.0 o/o for the total probable uncertainty.

SHAPE OF THE DECAY CURVE:

The results of the long time counting of the delayed activity are plotted in Fig. 9. The first point, plotted at 0.6 second, was taken from the sum of all channels in the automatic recording interval. The remaining points were from the manually record data. Obviously the timing in the latter data was not precise. However, over the 46 runs the timing errors probably averaged out to a small net error for any given point. After 170 seconds the background count per channel was greater than the effect. Hence th

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data have little meaning beyond that time. For this reason, as mentioned in Part I, it was not possible to make any precise determination of the half-lives nor of the absolute intensities of the various activities. The analysis of these data has already been discussed in Part I. The curve in Fig. 9 was computed from the values for the relative intensities R_i given in Table IV, for equilibrium irradiation, and the total yield value previously derived.

DISCUSSION

The best check on the over-all accuracy of these measurements and the analysis is to compare the total delayed neutron yield derived from the two experiments. The value 0.00765 arrived at by integration in Part I is about 12.5 o/o higher than the value 0.0068 obtained by direct measurement. It is true that the latter value involves an extrapolation method which makes use of the results of the analysis of Part I. However, the curve is so nearly linear in the interval under question that the intercept value would not be changed appreciably except by very great changes in the values of the coefficients used. On the other hand the value arrived at in Part I by integration of the analytical expression, involves not only possible errors in the assigned values for the coefficients, but also the uncertainties in the values of the mean lives of all five modes of decay. In view of this fact, it is rather surprising that the discrepancy is not greater than 12.5 o/o.

There is a rather large discrepancy between the above value for the total yield, and that derived from other experiments where slow neutrons were used to induce fission in plutonium. The most recent value for the absolute yield of delayed neutrons from U-235 is that determined by D. Hall, who found it to be 0.73 o/o of the prompt yield.[13] The ratio of the delayed yield from Pu to the delayed yield from U-235 was found by Wilson and Sutton [14] to be 0.38. Feld and de Hoffmann found the ratio to be 0.47.[15]

(13) Hall, D., CF- 3209, p. 12.
(14) Wilson, R. R., and Sutton, R. B., LA 76
(15) Feld, B. T., and de Hoffmann, F., LA 231

[redacted]

[redacted]

Using the mean of these two values, and Hall's delayed-neutron yield for U-235, we find that 0.31 o/o of the neutrons from plutonium are delayed when thermal neutrons are used to induce fission. This result is 35 o/o higher than the value 0.23 o/o obtained from the present measurements.

Feld and de Hoffmann indicate that their ratio is uncertain to 15 o/o. Furthermore their measurements include only delayed neutrons emitted 2 seconds or longer after irradiation ceased. Thus the short periods would not be properly weighted in their measurements. However it is difficult to see how so large a discrepancy can be accounted for on the basis of the experimental uncertainties. One possible answer to this discrepancy is that there may be a real difference in the delayed yield from fast-neutron excitation and slow-neutron excitation of plutonium.

An attempt was made to repeat the present experiment with slow neutrons by slowing down the neutrons from the lithium-p,n source in paraffin. However it was found impossible to obtain a sufficiently strong thermal flux in the region of the plutonium, and still retain a reasonable geometry for detecting the delayed activity. Consequently it was not possible to determine whether or not the total delayed neutron yield depends upon the energy of the exciting neutrons.

On the basis of these measurements, it is not possible to assign any limits of accuracy to the values derived for the intensities of the different periods. It is, however, of interest to compare the relative intensities R_i, shown in Table IV with the corresponding values derived by de Hoffmann for U-235, as shown in Table III. The chief differences are seen to be in the relative coefficients of the longest and the shortest periods. The value for period No. 1 in Table IV is only 61 o/o of de Hoffmann's value, while the value for period No. 5 is 38 o/o higher than the corresponding figure for U-235. In view of the fact that the same mean lives were used, except for No. 5, these large differences might be interpreted as an indication that the branching ratio for the particular fission fragments involved may be different for U-235 and the plutonium.

 [redacted]

[redacted]

Had de Hoffmann used the shorter mean life for period No. 5, the difference would probably have been larger in this case. On other possible reason for this difference may lie in the fact that the U-235 was excited by slow neutrons, whereas the plutonium was irradiated with fast neutrons. Further work will be necessary to determine whether or not the branching ratio changes with the energy of the primary neutrons.

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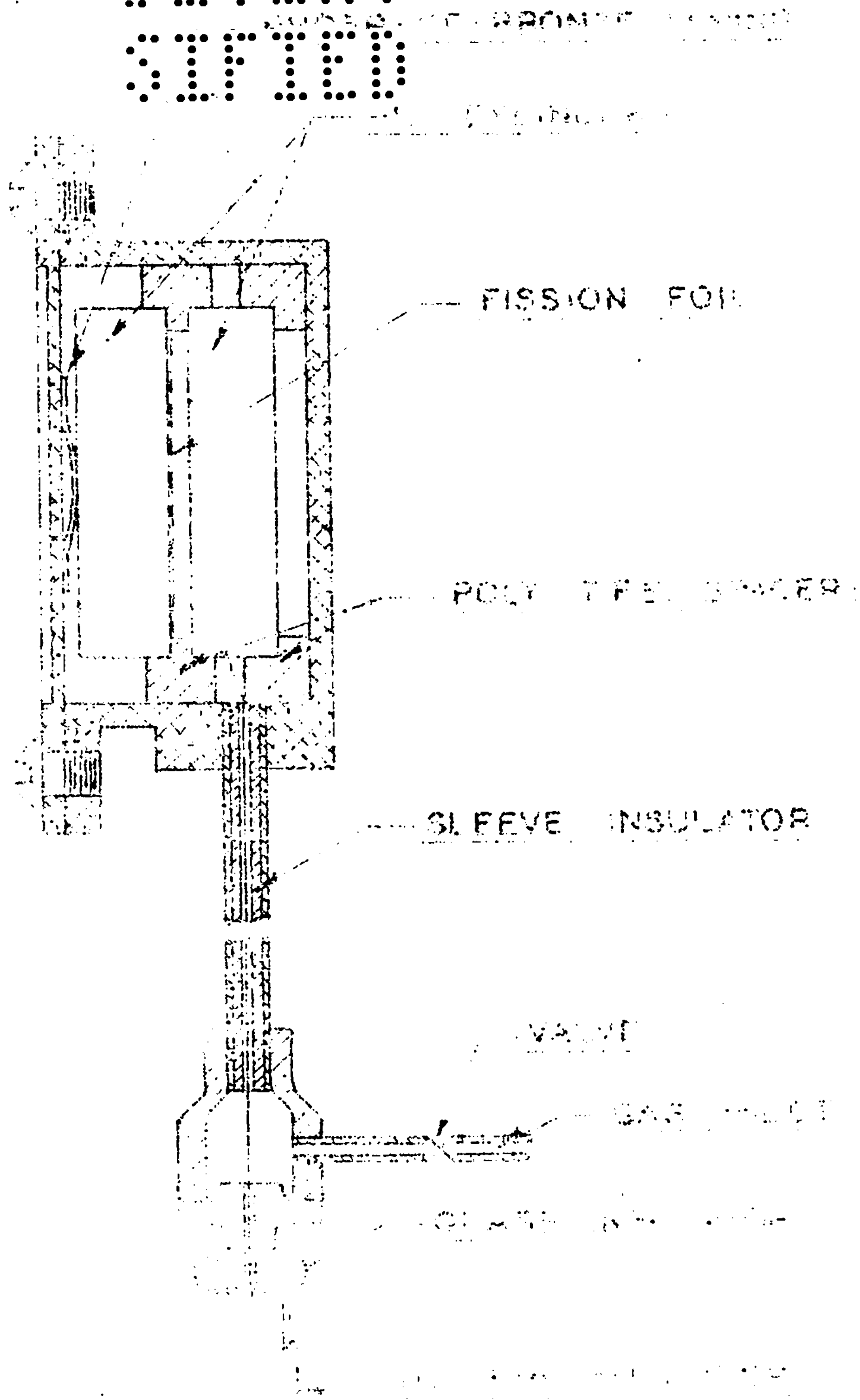

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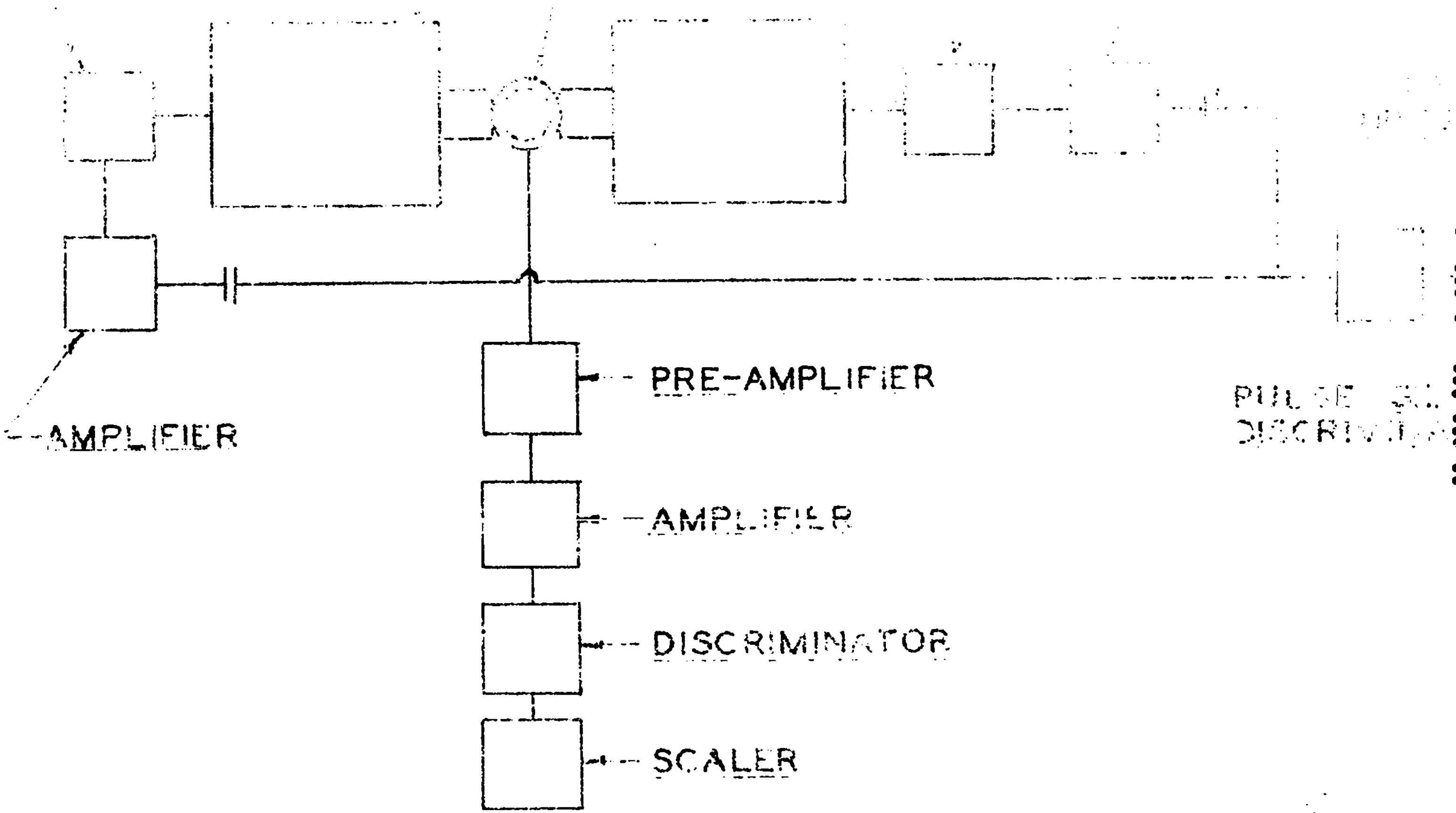
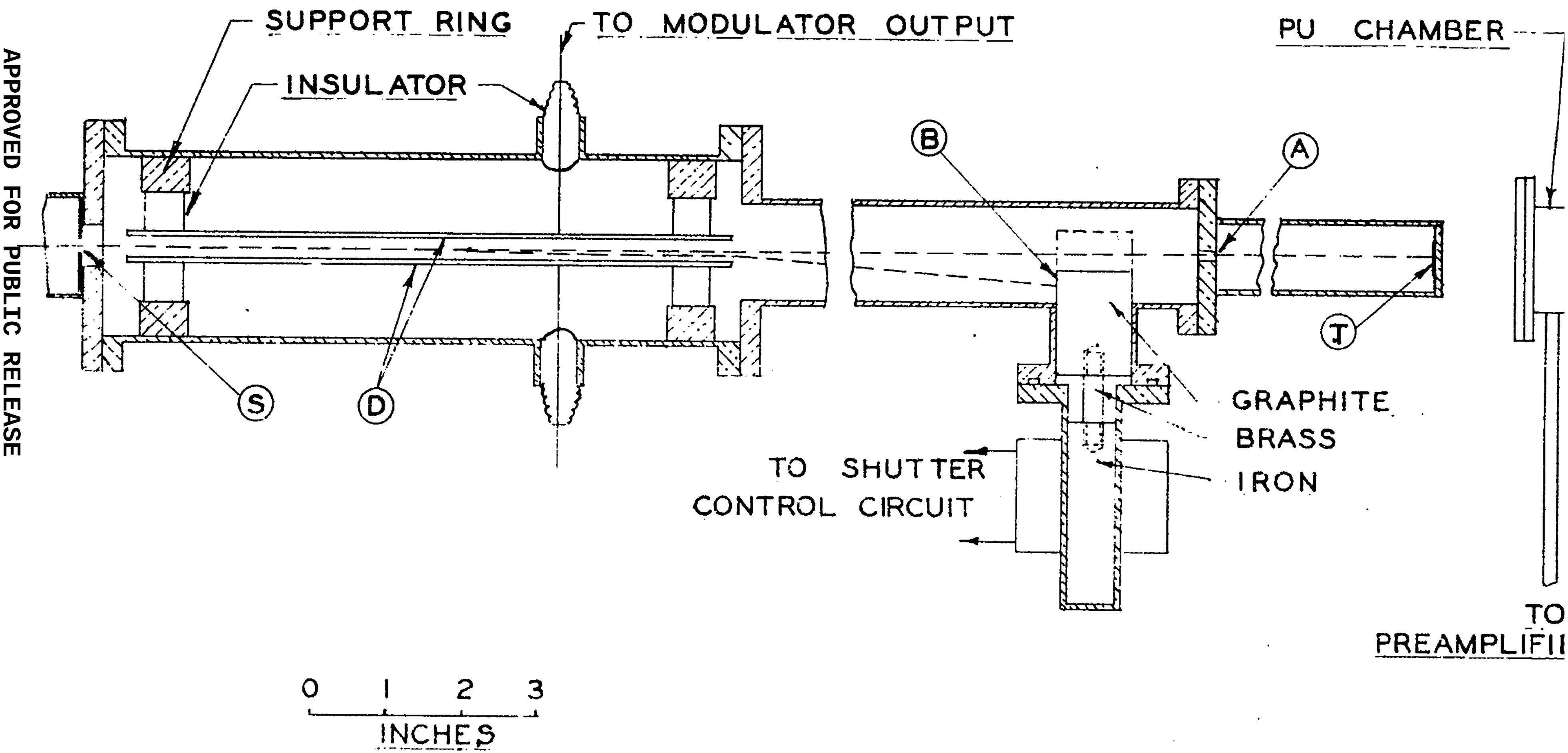


COUNTER ARRANGEMENT



TARGET TUBE ASSEMBLY

FIGURE Nº 3

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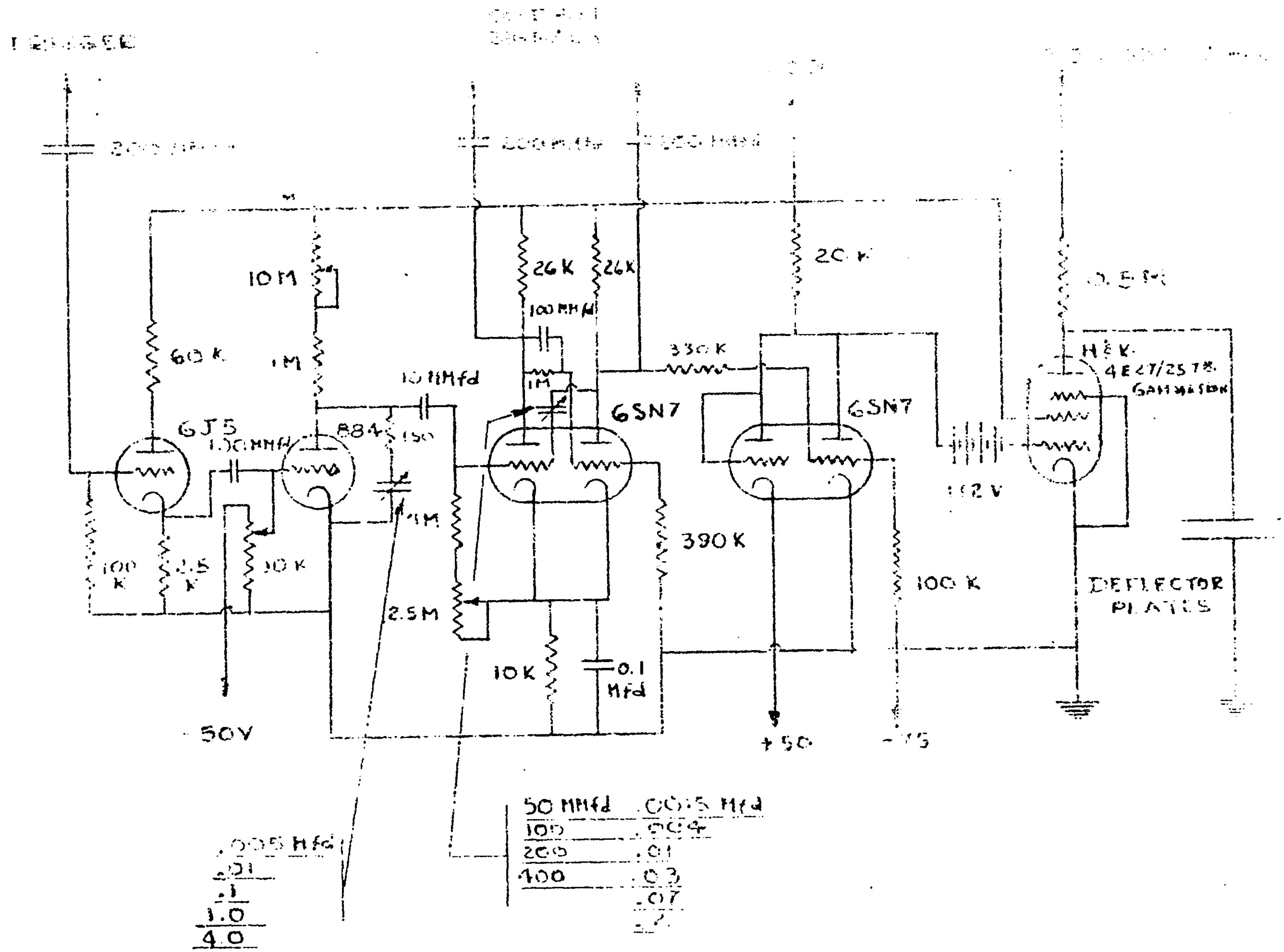

SLOW MODULATION CIRCUIT
FOR SHORT TANK

ROLLAND PERRY

FIGURE NO 4

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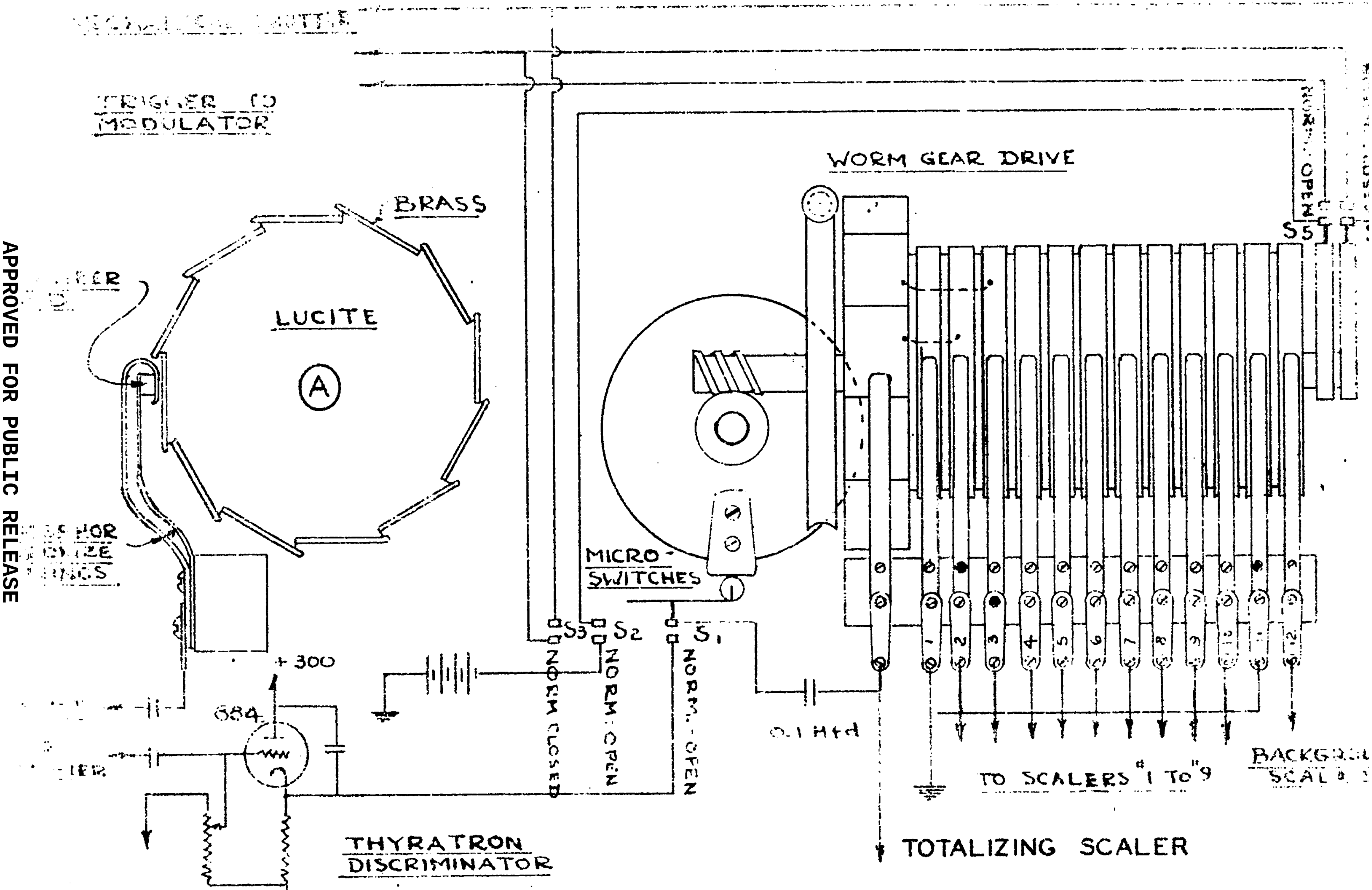

SCHEMATIC OF MECHANICAL TIME DISCRIMINATOR

ROLLAND PERRY

FIGURE Nº 5

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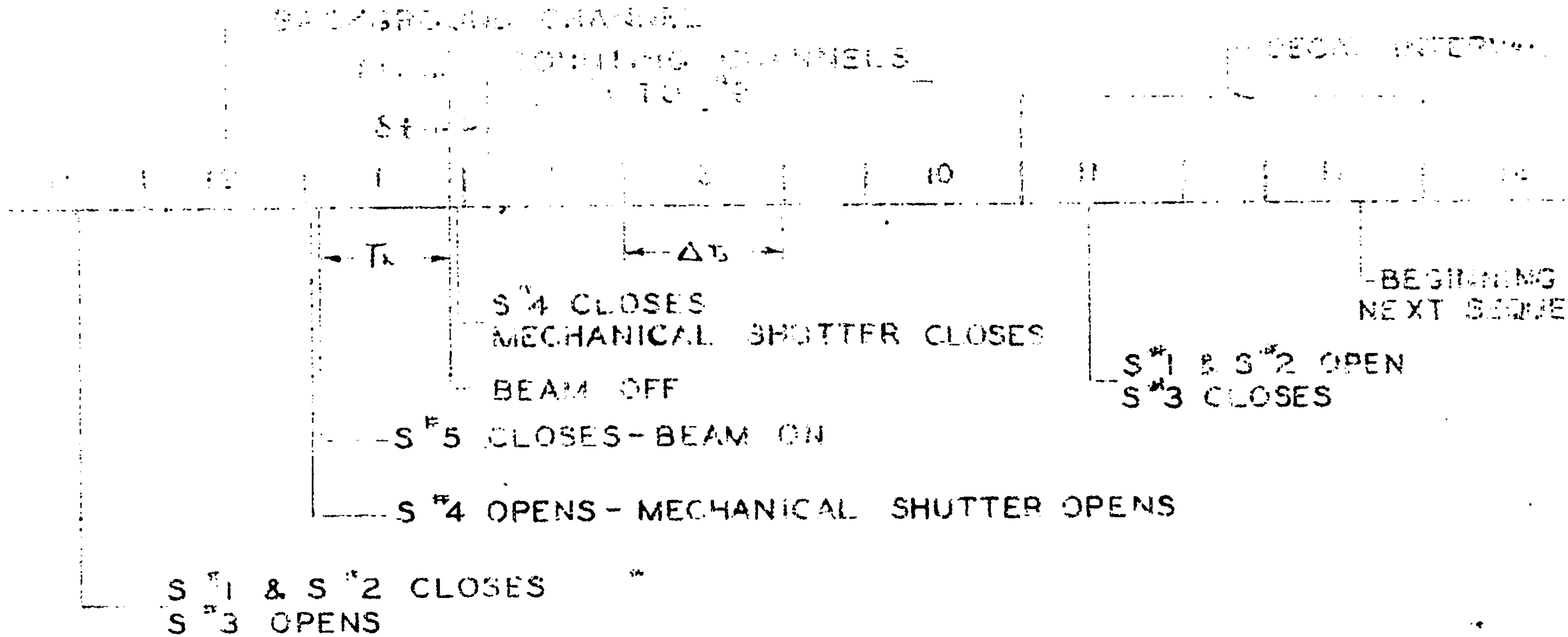

TIMING SEQUENCE

FIGURE Nº 6

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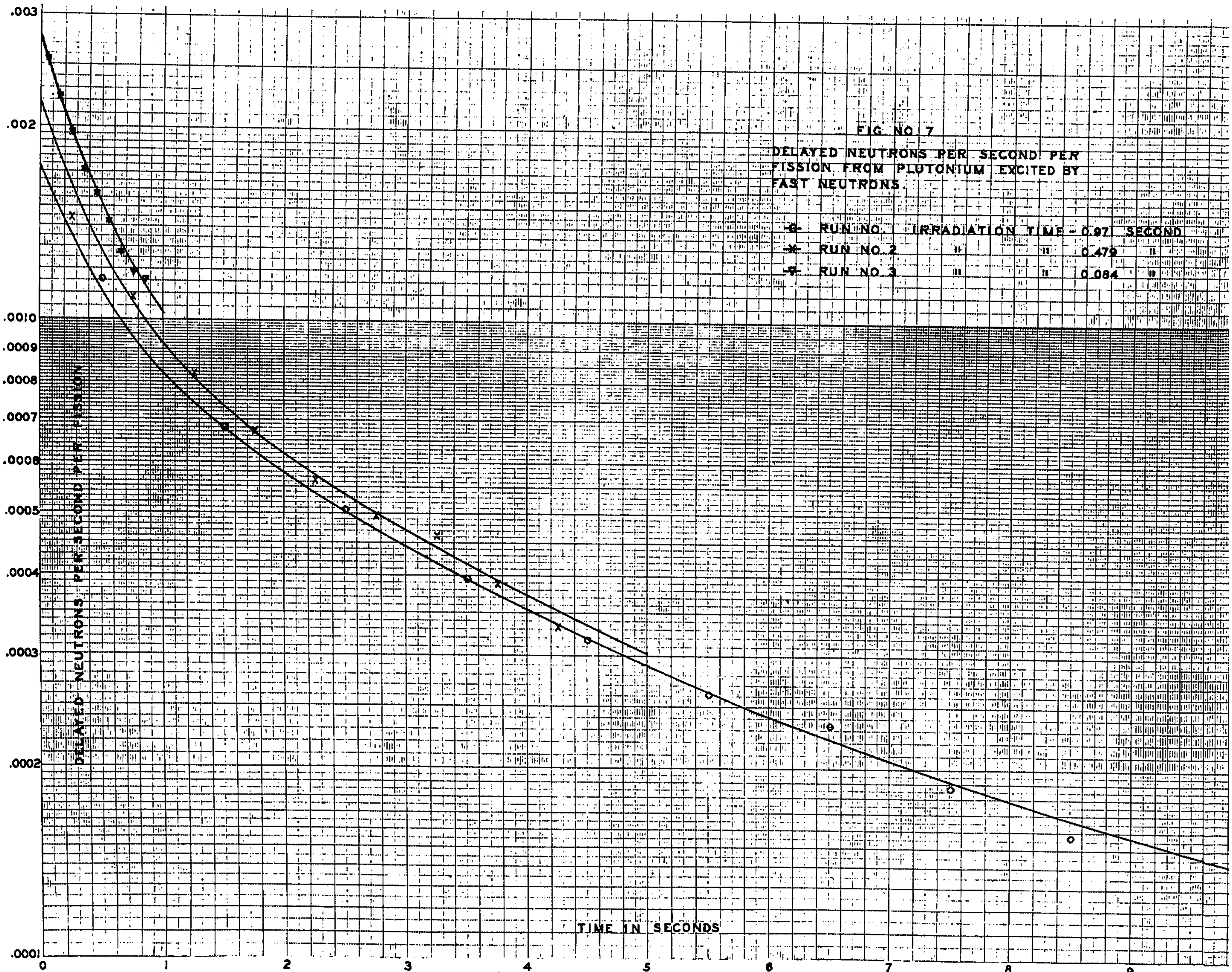

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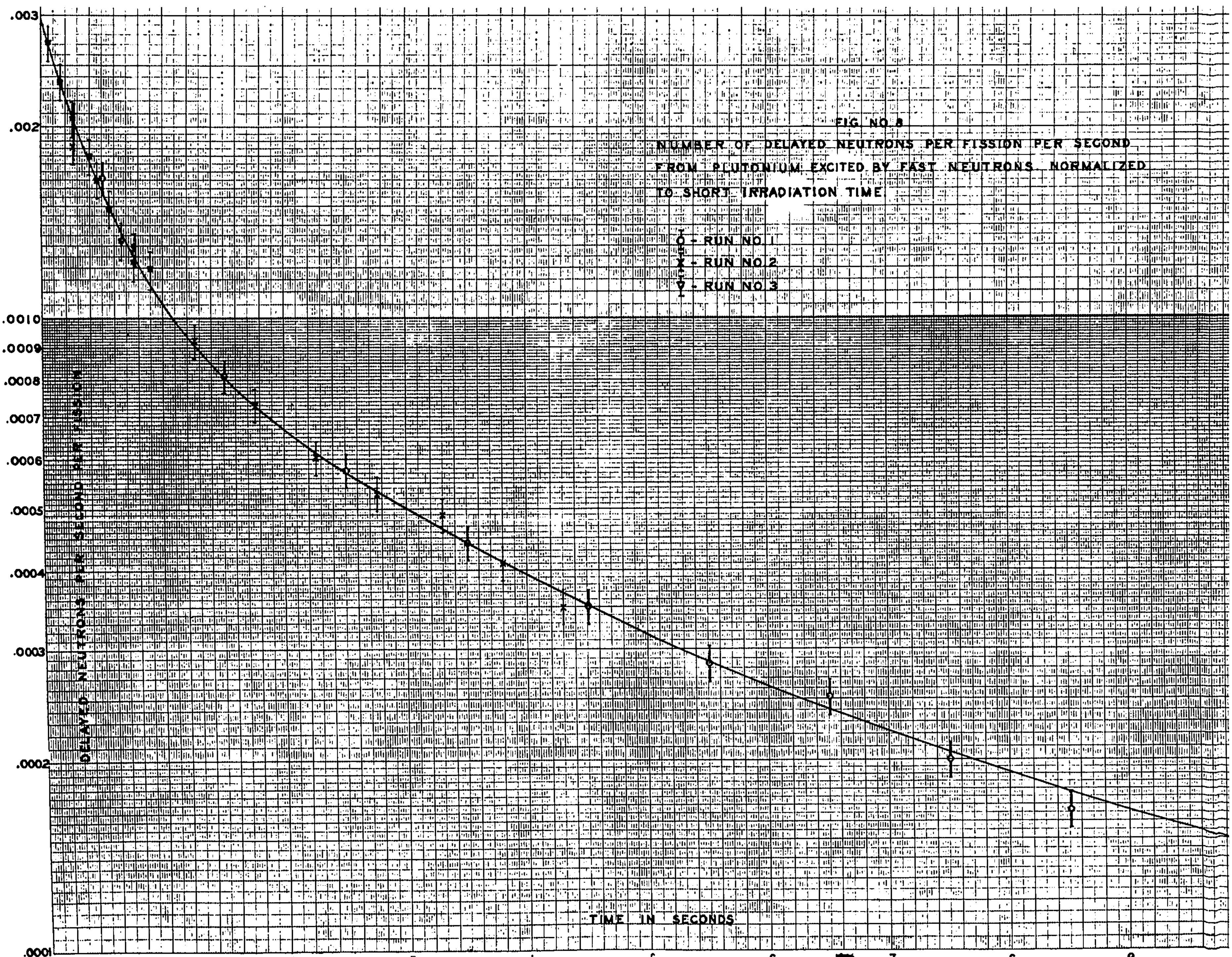

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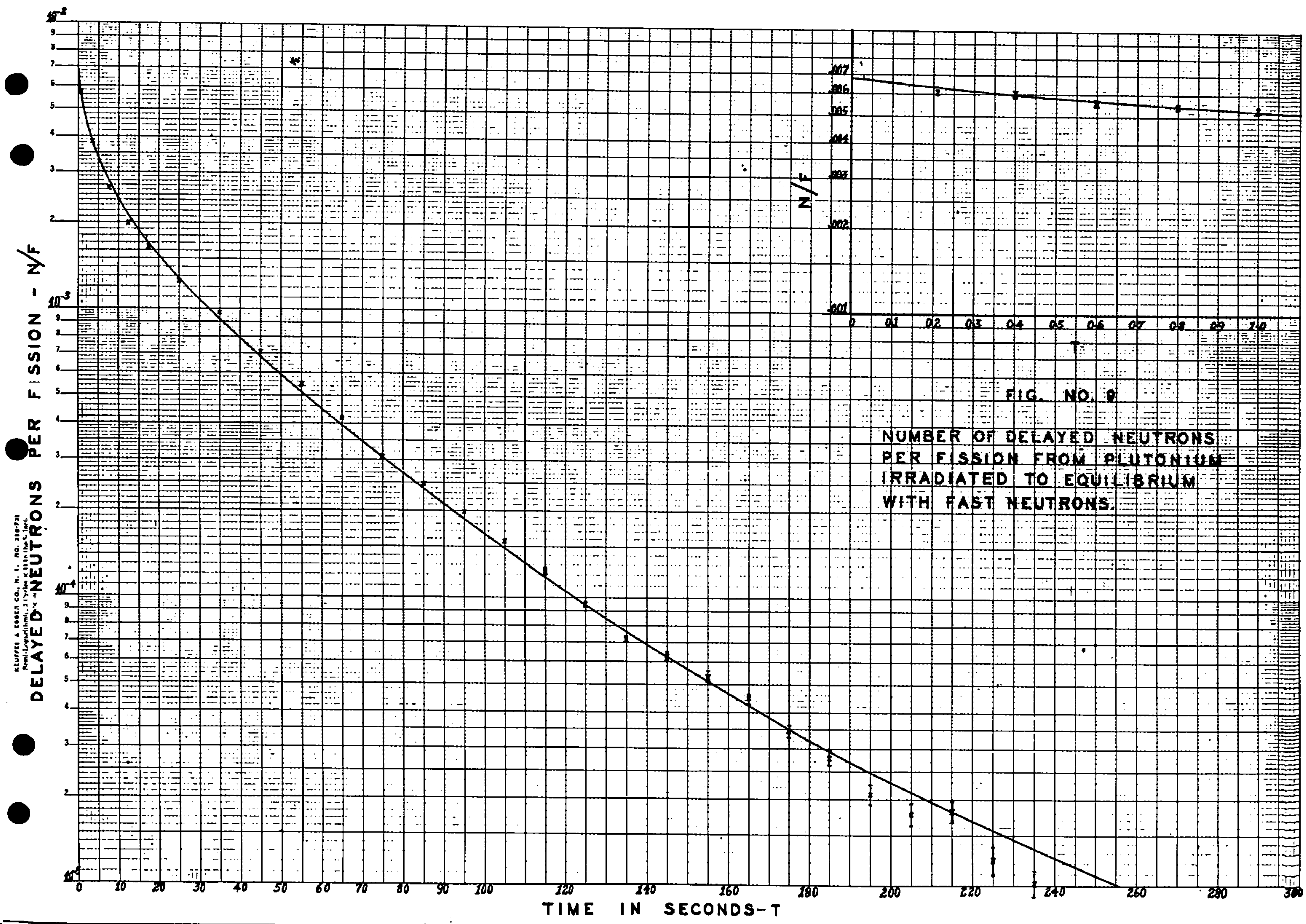

FIG. NO. 9

NUMBER OF DELAYED NEUTRONS PER FISSION FROM PLUTONIUM IRRADIATED TO EQUILIBRIUM WITH FAST NEUTRONS.

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