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SECTION I

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REPORT

THE MULTIPLICATION FACTOR FOR PRODUCT DRUMS CONTAINING URANIUM HEXAFLUORIDE

PROBLEM ASSIGNMENT NO. PX14-1

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ABSTRACT

The multiplication factors for 300-lb product drums containing uranium hexafluoride were measured by placing a fission source in the center of the drum and deriving the total number of secondary fissions produced in the hexafluoride per primary fission in the source. The measurements were made using material having isotopic concentrations of U^{235} of 1.1% and 0.536%. The effect of a hydrogeneous reflector around the drum was studied as was the degree of protection to be gained by cadmium and boron jackets. An extrapolation of the results indicates that a drum would become critical if it contained hexafluoride with about 10% isotopic concentration of U^{235}, if it were surrounded with water and had no cadmium cover. If dry or cadmium-covered, the concentration would have to be several times greater for criticality.

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Multiplication Factor for Product Drums Containing Uranium Hexafluorids under Varying Reflector Conditions

We had been asked to make some measurements which would aid in arriving at an indication of the safety against criticality of certain vessels, called "Product Drums", containing enriched uranium hexafluoride in fairly large amounts. Theoretical estimates had been made by E. Teller and others[1], which had been used (together with a generous safety factor) to set limits for the enrichment which could be tolerated in these vessels, but an accurate prediction was difficult to make particularly with regard to the influence of hydrogenous reflectors and the efficacy of cadmium coating on the drums. In view of the importance of the question from the engineering standpoint, it was desired to obtain measurements which would supplement the estimates and add new information about the reflection from surroundings. The experiments were to be done under conditions far from critical, but the measured values for the multiplication constant might serve as a basis for extrapolation to a value for the critical enrichment in the drums. Preferably, this would be done with the help of other measurements taken at higher enrichment levels.

The experiments were simple in principle. If a beam of neutrons is admitted into a product drum through a re-entrant tube and strikes

[1] Konopinski, Metropolis, Teller and Woods, CF-548.

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a plug of fissionable material near the center of the vessel, such an arrangement may be called a fission "source". Each fission in the source will have a probability k of producing directly another fission in the material in the drum. Each of these fissions has again a probability of producing directly a third, and we shall assume that this probability is again k. Continuing under this assumption, one fission in the source will produce $k + k^2 + \ldots = \frac{k}{1-k}$ fissions in the vessel. The assumption of constant k is incorrect insofar as fissions of later generations may take place nearer the surface of the vessel and, therefore, have a reduced chance of reproducing, but under the conditions of low enrichment prevailing in our experiments the later generations contribute so little that the assumption is justified. The ratio of the total number of fissions taking place per minute in the vessel to the number taking place per minute in the source leads, therefore, to a value for the reproduction factor k. In the experiments, the fission rate in the vessel was measured by taking distribution curves with a calibrated fission chamber across two diameters of the vessel at different positions on its axis, and integrating the distributions numerically to give a value for the whole vessel. The source strength was measured in terms of fissions per minute by determining the neutron output in a water tank experiment, and dividing by ν. The experiments were performed in two main series, supplemented by a few miscellaneous tests.

Experiments - First Series

Fig. 1 is a diagram of the product drum totally immersed in a water bath. The water, which was heated, served the double function of providing a hydrogenous reflector as required for most of the readings, and of keeping the hex in a liquid state so as to avoid cavities and consequent localized perturbations in the counting rates. The drum was 12" inside diameter, and had convex ends on a 20" long cylindrical section. The walls were 1/4" thick monel. A cadmium jacket 0.020" thick could be closely fitted over the outside surface. A pressure gauge connected to the interior of the vessel gave a convenient indication of the vapor pressure, and hence the temperature, of the contents. The temperature of the water bath was usually held at 74°C.

Three nickel pipes, one inch inside diameter, passed completely through the water bath and the drum. The upper pipe passed through the center of the drum, and the axes of the other two were, respectively, three inches and six inches below it. A deep, tight cadmium thimble extended into the upper tube from the lefthand side in the figure; the collimated neutron beam entered the open end of this thimble, traveled down its length, and struck the source plug at the closed end. The purpose of the cadmium thimble was to eliminate thermal neutron scattering from the source plug or the walls of the beam pipe into the contents of the drum. As will be seen later, even the epi-cadmium scattering was serious. The source plug was an aluminum container filled with 31 grams of 10.3% enriched UO_3.

The fission chamber is indicated in the middle hole. It was built at the end of a long shaft of 1/2" thin-walled brass tubing, which was connected through a detachable fitting to the housing of the first stage of the amplifier, indicated at the right of the figure. A platinum foil carrying an electroplated film of enriched U_3O_8, as thick as could be made to stick, was wrapped into a cylinder and slipped into the end of the brass tube; it constituted the grounded electrode for the chamber. A stiff wire was run down the axis of the brass tube and was supported there by spaced insulators. It was the high voltage electrode, and it was connected through a high voltage condenser to the grid of the first tube. The space between the brass tube of the fission chamber and the inside of the 1" nickel pipe was filled with appropriate substances to prevent neutron leakage; viz. wood or hydrogenous plastic in the region of the water*, and annular plugs filled with normal UF_4 in the region inside the vessel. Other brass capsules filled with UF_4 occupied unused parts of the other pipes which, if left empty, might have permitted undue leakage. By interchanging plugs, the fission chamber could be inserted into the lower pipe.

Calibration of Fission Chamber

The fission chamber could not be assumed to count with 100% efficiency because (1) it was cylindrical, and (2) the U_3O_8 film was not definitely thin to fission particles. It was necessary, therefore, to obtain a factor relating counts per minute in the chamber to fissions

* These are of course removed when readings are taken without water.

per minute per milligram of 25 on the foil.

This factor was obtained in three independent ways.

(1) Catcher Method. A piece of uranium metal foil was wrapped in a close-fitting cellulose acetate envelope and irradiated for 30 minutes (a) in the test vessel at a point at which the fission chamber was counting at the rate of 2682 per minute; and (b) in a thermal column under a slow flux (measured with small indium foils) of 2.95×10^5 neutrons/cm^2-sec. In the second case, use of the value 545 barns for the fission cross section of 235 leads to the conclusion that fissions were taking place in the uranium foil at a rate of 2.49×10^4 per minute per mg 235. This fission rate resulted in a count on the envelope or "catcher" of 53,600 over the time interval adopted for the catcher counting. The catcher in the vessel gave a similarly obtained count of 4186. Therefore, fissions were taking place in the vessel at a rate of $(2.49 \times 10^4 \times 4186) / 53,600$ or 1940 per minute per mg 235. The chamber counting rate, as noted above, was 2682, so that the counts per minute in the chamber must be multiplied by $1940/2682 = 0.72$ to obtain fissions per minute per mg 235.

The catcher foil was made of the same size as the foil in the fission chamber, so that variation of fission rate over the extent of the foils would not introduce error.

(2) Calibration Against Chamber With Weighed Foil. When the fission chamber was made, no microbalance was available to weigh the

uranium deposit. After the balance had been repaired another chamber was constructed, similar in size and shape, which contained 0.68 mg of 79% enriched U_3O_8 (0.46 mg 235). This chamber was found to count with 0.122 of the sensitivity of the chamber used through this first series of experiments. The 0.68 mg chamber gave a counting rate of 794 per minute in a thermal flux (in thermal column, measured with indium foils) of 53,500 neutrons/cm^2-sec, whence a counting efficiency of 39% can be deduced. The counts in the chamber used for the experiment must, therefore, be multiplied by $0.122 \times \frac{1}{0.39} \times \frac{1}{0.46} = 0.68$ to give fissions per minute per mg 235.

(3) Calibration Against an Alpha-counted Foil. A flat fission chamber was built, which could be expected to count with 100% efficiency. It contained a foil with a thin deposit of normal uranium, which was found to emit 581 ± 5 alphas per minute in a 52% geometry, and hence contained 0.0052 mg of 235. In a certain neutron flux, this chamber gave 143.5 counts per minute, or (assuming 100% efficiency) 27,600 fissions per minute per mg 235. In the same flux, the "effective"* counting rate with the cylindrical chamber was 44,000 per minute. 44,000 counts per minute in the chamber, therefore, corresponded to a fission rate of 27,600 per minute per mg 235, and all readings in the chamber had to be multiplied by 27,600/44,000 = 0.63 to obtain the fission rate.

*Actually, a third chamber was used as an intermediary in this inter-calibration, so that inordinately high counting rates could be avoided by using two flux levels.

Of these three results the third is based on the more reliable types of measurement, and we adopted it for use.

Effects of Scattered Neutrons.

The fission chamber in the pipes running through the vessel would register counts arising from neutrons of the following categories:

(1) Neutrons from thermal fissions in the source plug;

(2) Neutrons from epi-cadmium fissions in the source plug;

(3) Epi-cadmium neutrons scattered from the beam by the source plug;

(4) Stray neutrons from the beam or from the room.

A clean way to eliminate the undesired neutrons and at the same time to retain a well-defined meaning for the "source strength" seemed to be to take the difference between counts with and without a cadmium disk over the face of the source plug through which the beam entered. Such a difference would represent the effects of neutrons of the first kind only. A similar difference measurement would be taken in measuring the source strength. We would thus be dealing only with thermal fissions in the source. As will be seen in our description of the results, the fissions from neutrons of categories 2, 3 and 4 comprised about three-quarters of the total, and there resulted a serious loss in the accuracy of the difference measurements both for the source strength and for the fissions in the chamber caused by thermal fissions in the source plug. A curve taken with no source plug present (upper pipe empty) showed that there were few stray neutrons; undoubtedly the epi-cadmium scattered neutrons were the main contributors to the counts obtained with the

cadmium disk over the source face. Attempts were made to improve this situation by thermalising the primary beam with graphite plugs placed at the optimum position in the hole through the pile reflector. The cost in terms of intensity was, however, so great that the attempts were abandoned in favor of the alternative approach used in the second series of experiments.

Source Strength Determinations.

The source arrangement adopted after some preliminary work was a cylindrical aluminum cartridge 7/8" ID x 15/16" OD x 2½" in length, containing 31.150 gm of UO_3 enriched to 10.3% U^{235}.

The neutron beam entering the source from the X-pile was monitored by 7/8" disks of 0.006" indium held normal to the beam in a dummy aluminum cartridge interchangeable with the source. The difference between saturated activities of such foils with and without a 0.02" cadmium shield was taken as an index to the slow neutron flux in the beam.

Foil measurements at both ends of the source indicated that about 15% of the slow flux entering the source was transmitted.

One method of determining the effective source strength is that of relating monitor-foil activities in the beam and in the standard pile to determine the flux entering the source, and calculating the fission rate which should result. The fraction (q) of slow neutrons not transmitted through the source that produces fissions of U^{235} is:

$$q = (U^{235} \text{ fissions})/(U^{235} \text{ absorptions} + U^{238} \text{ absorptions} + \text{U scattering} + \text{O scattering})$$

$$= \frac{.103 \times 545}{.103 \times 645 + .897 \times 2.7 + 1 \times 8.5 + 3 \times 4} = 0.63$$

In attempting to evaluate the flux in the beam in terms of activations in the standard pile, it was found that the 0.006" monitor foils are relatively "thick" with regard to activities induced by random and by collimated slow fluxes, and also with regard to β-absorption within the foil. It was, therefore, necessary to evaluate activities in the beam and in the standard pile with foils of various thicknesses (.001", .002", .004", .006") and extrapolate to zero thickness from these data to determine the ratio of beam to standard-pile flux. This ratio, corresponding to a pile power of about 3600 kw, was found to be 1180. The slow (C-neutron) flux in the standard-pile position used is 8570 neutrons/cm^2-sec. Since the capture cross-section of indium for slow neutrons is approximately proportional to $1/v$, an additional correction must be made to take account of the difference between neutron temperatures in the beam and in the standard pile -- about 550°C and 330°C, respectively, -- by the factor $\sqrt{\frac{550}{350}} = 1.25$. The slow (C-neutron) flux in the beam at 3600 kw is then:

$$nv = 1181 \times 8570 \times 1.25 = 1.264 \times 10^7 \text{ neutrons/cm}^2\text{-sec.}$$

Since the cross-sectional area of the source (7/8" diameter) was 3.87 cm^2, the fraction of incident slow neutrons not transmitted through the source was 0.85, and the fraction of this portion producing U^{235} fissions was 0.63, the fission-source strength is given by:

$$Q_f = 1.264 \times 10^7 \times 3.87 \times .85 \times 0.63 \times 60 = 15.8 \times 10^8 \text{ fissions/min.}$$

An alternate method of calibrating the source is the usual substitution procedure in a tank of manganese sulfate solution, by means of which the average activations of the manganese by the fission source and by a standard source are compared.

An open cylindrical steel tank was used, of height 30", diameter 27", and wall thickness about 1/8"; the wall and bottom of the tank were covered with 0.02" cadmium. A 1" ID nickel tube 1/8" thick, identical with those forming the measuring channels in the vessels of UF_6, passed through a central diameter of the tank. The solution used was composed of about 1% by weight of $MnSO_4 \cdot 4H_2O$ in tap water.

The sample of manganese to be counted was precipitated as the carbonate by taking a sample of about 1 liter from the tank after thorough stirring, filtering the sample, adding 15% by volume of saturated NH_4Cl solution, heating to about 85°C, and adding an excess of $(NH_4)_2CO_3$ solution. The precipitate was filtered out, washed with water, and pressed level while damp into a 1/2" x 2½" x 1/16" pan milled into the surface

of an aluminum tray, and dried. The tray supported the sample axially underneath the G-M counter tube.

Ordinarily several duplicate samples were run, because individual fluctuations of 5% were not unusual. Also, long-life activities -- chiefly iron -- built up gradually in the solution because of the considerable stray neutron background in the vicinity, and were mixed with the precipitate. It was, therefore, necessary to follow the decay of both the intentionally-activated sample and a background sample taken immediately before irradiating the solution. The difference of these two curves agreed closely with the expected manganese decay curve, and was used as the basis for evaluating the saturated activity of the manganese resulting from the desired irradiation.

The standard source used was a 0.5 gm Ra-α-Be unit (C.L.#9) of strength 5.27×10^6 neutrons/sec. Standardization runs were made by placing this source in an aluminum cup attached to the outside of the nickel tube at the middle, so that the source capsule itself was in the solution; a lining of 0.02" cadmium extended from the middle of the tube to one end, duplicating the arrangement in the test vessel. An irradiation time of 2 to 3 hours was found satisfactory.

A calibration run with the fission source was made by aligning the nickel tube with the neutron beam, placing the cartridge of enriched UO_3 at the middle of the tube in its cadmium thimble, and

irradiating for 1 to 2 hours at a constant pile power level. After the sample was taken, the cartridge was removed and monitor foils were run at the source position. Two runs had to be made, one with a cadmium shield in front of the source, to determine the activation resulting from slow-neutron fissions.

The irradiation with the standard source was repeated several times. Low intensity, iron contamination, high backgrounds, and other troublesome factors caused considerable inconsistency among the results. The last determination was made with three 0.5 gm Ra - α - Be sources at the center of the tank; the increased intensity, together with the lowered background after rust-proofing the tank, resulted in a value which is believed to be reliable within about 5%. Usable results for the saturation activity, $\bar{A}_s$, are as follows:

	642	counts/min	(1 source)
	595	"	"
	580	"	"
	691	"	"
	598	"	" (3 sources)/3
Average	621	"	"

Because of the greater weight to be given the last determination, together with other experimental factors, the value $\bar{A}_s$ = 600 counts/min is probably as good as can be assigned on the basis of these data.

A determination of $\bar{A}_s$ with the fission source in the tank yielded the results:

A_s (uncovered source) = 18,950 counts/min

A_s (Cd-covered source) = 14,270 counts/min

A_s (slow fissions) = 4,680 counts/min

The fission source strength on the basis of these results is:

$$Q_f = \frac{5.27 \times 10^6 \times 4680}{600} = 4.11 \times 10^7 \text{ neutrons/sec.}$$

which corresponds to 9.86×10^8 fissions/min.

The above-mentioned background irradiation, however, was made while the pile was running at half the usual power; and the extrapolation to full power conditions is not very satisfactory in view of the small difference between background and total activity. Another determination was made in the same way, but with nearly constant pile power during both irradiations. The results are:

A_s (uncovered source) = 10,510 counts/min

A_s (Cd-covered source) = 6,680 counts/min

A_s (slow fissions) = 3,830 counts/min

The corresponding evaluation of source strength is:

$$Q_f = 3.36 \times 10^7 \text{ neutrons/sec.}$$

or 8.1×10^8 fissions/min.

The discrepancy between this value and that obtained by flux measurements with foils, 15.8×10^3 fissions/min., has not been explained. Because of the relative simplicity of the manganese-sampling method, the result obtained by use of it is probably the more reliable.

Neutron Captures in the Water Reflector

An attempt was made to determine the rate of emission of neutrons from the product drum into the surrounding water reflector. The method used was similar to that for calibrating the fission source by activation of manganese; the water bath was filled with a weak $MnSO_4$ solution and the fission source irradiated in the vessel.

The relation of the rate of capture in the bath to that in the standardizing tank with the standard source involves both the respective saturated activities of the manganese and the ratio of the volumes of water in the two tanks. Assuming that all neutrons emitted from the vessel are captured in the manganese solution at a total rate Q, the average rate of capture in an element of volume ΔV is:

$$\overline{\Delta Q} = Q \frac{\Delta V}{V} .$$

If the density ρ of manganese in solution is such that [(Mn capture rate)/Q]<1 then the manganese capture rate and hence average activation $\bar{A}_s \propto \rho$.

If uniform samples weights w are taken for counting,

$$\Delta V = \frac{w}{\rho} \text{ and } \bar{A}_s = \mathcal{H} \rho \quad \overline{\Delta Q} = \mathcal{K} \rho \left(\frac{w/\rho}{V} \right) Q$$

where $\mathcal{K}$ includes counter calibration and other constant factors ,

[redacted]

then $\frac{Q}{Q'} = \frac{\bar{A}_s V}{\bar{A}'_s V'}$

Data were taken both with and without cadmium enclosing the vessel of UF_6. In each case it was necessary to determine the activation of the manganese by two irradiations, one with a cadmium shield in front of the sourcecartridge to exclude the effects of slow-neutron fissions. Actually, since the saturated activity of the managanese was not much different with or without the cadmium cover on the vessel, only one background run was made; the resulting data were extrapolated to the other case by the ratio of total saturated activities with and without the cadmium shield on the vessel. Results, monitored to about 3600 kw., are:

	A_s(total)	A_s(Bkgnd)	A_s(slow fissions)
Uncovered vessel	6770	4830*	1940
Cd-covered vessel	5730	4090	1640

The capture rate im the jacket Q_j is given by:

$$Q_j = \frac{Q_s (A_s)_j V_j}{(A_s)_s V_s}$$ where j and s refer to values in the water jacket and in the standardization tank respectively.

Without Cd covering on the vessel, $Q_j = \frac{5.27 \times 10^6 \times 1940 \times 709}{600 \times 282} = 4.28 \times 10^7$ neutrons/sec

With Cd covering on the vessel, $Q_j = 3.62 \times 10^7$ neutrons/sec.

It will be noted that both of these results exceed the fission-source strength as determined by manganese-sampling technique. Since the slow-fission activity in each case is obtained as a small difference of large numbers, these results probably indicate only that the capture rate

* Extrapolated value

[redacted]

in the jacket is of the same order as the strength of the source.

Results of Fission Counts - First Series of Experiments

Fig. 2 shows the fission distribution curves as obtained with and without cadmium on the source face, in the hole three inches below the source plug, with a cadmium jacket on the vessel and the water reflector present. The lower curve is the difference of the upper two. The figure is given as an example of the nature of the experimental data. In Figs. 3 and 4 only the difference curves are given, and accordingly no experimental points are indicated. Figure 3 refers to measurements in the hole three inches below the source, and Figure 4 to the hole six inches below the source. Three conditions of surroundings for the drum are treated in each figure. The large effect of a water reflector and its partial neutralization by the cadmium are evident even on casual inspection.

The asymmetries in the distributions on either side of the axis were unexpected, and we have not found an adequate explanation for them. They look like a scattering effect, but our method of taking differences was intended to eliminate that sort of thing.

Fig. 5 shows the difference curves for the distributions taken with the source displaced from the center to a position near the wall of the drum.

In describing the evaluation of k from these data, one should first observe that the fission chamber detector had finite width, and, therefore, the sharp rise in counting rate near the walls of the vessel when the reflector and cadmium were present must be partly discounted

because of overlapping into the reflector region. Only such regions of these curves were used as corresponded to the chamber being entirely within the drum. A mean was struck between the halves of the curves on either side of the axis, the averaged values of the ordinates were multiplied by $2\pi r$, and the resulting curves were separately plotted using a smooth extrapolation to the drum wall when it was necessary to bridge the uncertain region mentioned just above. The areas under these plots then gave the product (fission rate per mg. 25) x (cross-sectional area).

To perform the integration in the vertical direction, these products were plotted against the distance along the axis, giving two points which, together with zeros at the bottom and top of the hex in the drum and the consideration of symmetry about the equatorial plane, sufficed to give a good integration for the product (fission rate per mg. 25) x (volume of hex in cubic inches). When multiplied by the amount of 235 in one cubic inch of hex (442 mg for 1.1% enrichment), and corrected for neutron absorption in the walls of the nickel pipe, (see Appendix A), this yielded the required value for the total number of fissions taking place per minute in the whole drum. The height of the liquid hex in the vessel was estimated from the weight (304 lbs.), the density (59.1 g.per cu. in.) and the inside diameter of the vessel, with reasonable approximation for the convex bottom and correction for the volume occupied by the three internal pipes.

Treatment of the experimental data in this manner led to the figures in the fifth column of the first section of Table II for the total number of fissions taking place per minute in the drum.

[redacted]

Measurements in Neutron Capture in 238

In addition to the fission rate measurements, some measurements were attempted of the rate of capture in 238 in the hexafluoride drum. These were done in the middle and lower holes with no reflector around the drum, and with the water reflector but no cadmium jacket. As with the fission measurements, differences had to be taken between results obtained with and without a cadmium disk over the source face.

Five capsules of U_3O_8 were placed at intervals in the hole under investigation. They were spaced with UF_4 plugs. After irradiation for one hour they were removed and dissolved in nitric acid. After evaporating nearly to dryness, the resulting nitrate was dissolved in ether and two extractions of the fission products and the UX were made with water. Following this the uranium was precipitated as sodium uranyl acetate, and counted as samples infinitely thick for beta rays. The decay of the 23-minute U^{239} was followed by growth of beta activity arising from UX, and the 239 activities were all normalised to an equal UX growth rate. This whole procedure was calibrated by carrying it out in a known thermal flux, so that the results could be expressed in absolute terms; viz., "captures per minute per mg. 238".

The results are given in Table I. They are inaccurate because of (1) the low activities involved, and (2) the necessity of taking differences to eliminate the effects of scattered neutrons. These results were obtained with material of 1.1% isotopic concentration of 235, and the thermal fission source strength was 8.1×10^8 fissions per minute. The results give only a

[redacted]

rough idea of the rate of capture; it will be noticed that when no water was present the effect due to thermal fissions in the source was too small to show above the errors in the measurements.

TABLE I

Captures per minute per milligram of U^{238}

	Hole	Source Face	Distance from axis(inches)				
			-4	-2	0	+2	+4
Water, no cadmium jacket	Middle	Uncovered	18.7	25.6	30.2	-	25.9
		Cd-covered	14.4	18.3	26.3	27.8	21.8
Difference: captures due to thermal fissions in source			4.3	7.3	3.9	-	4.1
Water, no Cd jacket	Lower	Uncovered	8.0	13.8	11.5	12.1	14.1
		Cd-covered	7.7	6.8	6.7	9.8	8.5
Difference			0.3	7.0	4.8	2.3	5.6
No water	Middle	Uncovered	15.8	20.7	25.6	27.6	16.8
		Cd-covered	17.4	21.0	25.2	27.4	17.6
Difference			-1.6	-0.3	+0.4	+0.2	-0.8

Second Series of Experiments

Because of the troublesome effects of the scattered neutrons from the beam source used in the first series of experiments, it was thought desirable to repeat the fission chamber measurements with a "mock fission" source of neutrons in the center of the vessel. The results of such measurements should be more reliable because (1) no differences would have to be taken to eliminate the effects of undesired neutrons; (2) the fission distribution curves might be expected to be more symmetric; (3) the source strength could be determined more accurately. A Po-Be source was borrowed from Chicago for a few days, and during this time we not only repeated all the fission chamber measurements on the vessel containing 1.1% material, but added a similar set of data for a drum containing depleted (0.536%) material and also investigated the effect of a protective jacket of calcium boride.

As the Po-Be source was much weaker than the beam source used earlier, it was necessary to compensate for the corresponding loss in intensity by increasing the sensitivity of the detector. This was done by building a new fission chamber, in which the diameter of the grounded outer electrode was increased to one inch, and the high-voltage electrode was expanded to a cylinder 1/2" in diameter. Uranium-plated foils were mounted on both electrodes. This chamber was calibrated against the old one by direct comparison in the same neutron flux. The result was that counts per minute in the new chamber had to be multiplied by 0.135 to give fissions per minute per milligram of 235. It may be noted that this calibration rests upon the same data as did the old one, namely measurement

against the alpha-counted foil in the flat fission chamber independently backed by checks by the catcher and weighing methods.

The cadmium thimble in the upper or source tube was naturally dispensed with in this series of measurements, and the hole was symmetrically filled on either side of the Po-Be source with UF_4 plugs.

Results of Fission Counts - Second Series of Experiments

Figures 6 and 7 give the experimental data for the middle and lower holes, respectively, in the drum containing the 1.1% material. Fig. 8 gives "displaced source" distributions for the case with reflector and no protection. Figures 9 and 10 give centered source distribution curves for the vessel containing 0.536% material.

These data were all subjected to the same type of numerical integration as was described above. The results are given in the second section of Table II.

Source Strength

We did not have time to measure this for ourselves, but instead we accepted the value 4.75×10^6 neutrons per second on July 28 as obtained by Friedman.

Use of Radium-Boron Source

A two-gram radium-boron source was available, and a measurement was taken with this source in the center of the depleted tank, with water but no cadmium jacket. The result is given in the third section of Table II, and it will be noticed that the k-value obtained is in close agreement

with that obtained in like circumstances with the Po-Be source. This agreement may be taken as an indication that the multiplication is not a sensitive function of the energy spectrum of the neutrons from the source.

Discussion of Results

The results obtained with the radioactive sources as given in Table II are probably accurate to about 15%. Those obtained with the beam source are less reliable, and should be considered as corroborating figures, independently obtained.

A few rather obvious features of the results can be seen by comparison of the fission distribution curves, and in the figures of Table II. They include (1) the very large effect caused by the presence of the water reflector; (2) how this is very substantially reduced by the use of a cadmium jacket; (3) that boron is slightly better protection than cadmium. Within the precision of the measurements, the multiplication factor is proportional to the isotopic concentration of U^{235}.

The readings with the displaced source were taken to check the assumption inherent in the simple theory that a fission has a uniform chance of producing another fission, irrespective of the position in the vessel. For the 0.536% material with water but no cadmium present, it is seen that a value of k of 0.089 was obtained when the source was near the drum wall, as compared with $k = 0.10$ for the centered source. Apparently, then, a fission near the wall was 89% as effective in producing other fissions as was a fission near the center; if the reflection from

TABLE II

[redacted]

Isotopic Concentration of 235	Source Position	Shielding and Reflector Conditions	(Fissions in Chamber/min/mg 235) x (Volume of drum in cu. in.)	Total Fissions per min. in drum	k
---- Beam Source ;		Equivalent Strength: 8.1×10^8 fissions per minute			
1.1%	Centered	Water, cadmium jacket	11.3×10^4	5.6×10^7	0.065
"	"	Water, no jacket	30.6×10^4	20.3×10^7	0.20
"	"	No water	2.97×10^4	1.5×10^7	0.018
"	Displaced	Water, no jacket	15.3×10^4	10.2×10^7	0.11
---- Polonium-Beryllium Source;		Equivalent Strength: 1.06×10^8 fissions per minute			
1.1%	Centered	Water, cadmium jacket	8.6×10^3	4.3×10^6	0.039
"	"	Water, no jacket	32.0×10^3	21.2×10^6	0.167
"	"	No water	3.94×10^3	2.0×10^6	0.019
"	"	Water, calcium boride jacket	6.00×10^3	2.9×10^6	0.027
0.536%	"	Water, cadmium jacket	10.5×10^3	2.54×10^6	0.023
"	"	Water, no jacket	37.3×10^3	12.0×10^6	0.10
"	"	No water	3.27×10^3	0.79×10^6	0.0069
"	Displaced	Water, no jacket	32.0×10^3	10.3×10^6	0.089
---- Radium-Boron Source;		Equivalent Strength: 1.05×10^8 fissions per minute			
0.536%	Centered	Water, no jacket	37.0×10^3	11.9×10^6	0.10

[redacted]

the surroundings were absent this figure would probably be much reduced. However, a large correction is not involved even for the dry and the jacketed drums, because at these low concentrations the second generation fissions already are making only a very small contribution.

It is of course of interest to see how one can use these results in estimating critical concentrations for the product drums. It has been pointed out by Teller that the probability P_f of a neutron producing a fission as it passes through the material is not as closely proportional to the concentration p of U^{235} as is the quantity $P_f/(1-P_f)$.* Accepting this, we can write

$$\frac{P_f}{1 - P_f} = \text{const.} \times p$$

Now $P_f = k/\nu$ so that if k_o and p_o are experimental values for a particular arrangement,

$$\text{const.} = \frac{1}{p_o} \cdot \frac{k_o/\nu}{1 - k_o/\nu} \simeq \frac{k_o}{p_o \nu}$$

---

* This can be seen as follows: Neglecting radiative capture in U^{235},

$$P_f = \frac{N_{25}\ \sigma_f}{N_{25}\ \sigma_f + N_{28}\ \sigma_c} = \frac{1}{1 + \frac{N_{28}}{N_{25}} \cdot \frac{\sigma_c}{\sigma_f}}$$

where N_{28}/N_{25} is the ratio of the numbers of U^{238} and U^{235} atoms, viz., (1-p)/p; and σ_c/σ_f is the ratio of capture to fission cross sections, viz., 1/10. Substitution gives $P_f = 10p/(1 + 9p)$ from which it is clear that P_f is not proportionsl to p. On the other hand, we have

$$P_f/(1-P_f) = 10p/(1-p)$$

the values of which are nearly proportional to p when p is fairly small.

In the case of criticality, $\frac{P_f}{1-P_f} = \frac{1/\nu}{1-1/\nu} = -\frac{1}{1-\nu}$

so that $$P_{crit} = \frac{\nu}{1-\nu} \cdot \frac{p_o}{k_o}$$
$$= 1.7 \frac{p_o}{k_o}$$

Substitution of the experimental p - and k - values from Table II then gives the critical concentrations. Taking the most dangerous case, viz., the unjacketed drum surrounded with water, one thus obtains a critical enrichment of 11% from the experiments on the 1.1% material, and 9.1% from the 0.536% material. The average of these corresponds to 9 kg of U^{235} in the drum for criticality. For the jacketed and dry drums, the critical amount is several times larger.

In conclusion, it must be emphasized that both the measurements and the method of extrapolation to critical concentrations are inaccurate. The need for further work at higher concentrations is clearly indicated.

Acknowledgment

The suggestions and continued interest of Frank C. Hoyt are gratefully acknowledged.

APPENDIX A

Correction for Neutron Absorption in the Nickel Pipe

Inasmuch as all of these measurements were taken with the fission chambers in nickel pipes inside the drums, it was important to evaluate the correction required to take into account the neutron absorption in the pipe wall. A sample piece of the pipe was obtained, and measurements were taken by placing the fission chamber in our standard graphite pile at a distance of sixteen inches from the Ra-Be source. Counts were taken under the following conditions, with results as indicated:

(1) Fission chamber alone 420.9 counts per min.

(2) Fission chamber in cadmium 3.2 " " "

(3) Fission chamber in nickel pipe 247.3 " " "

(4) Fission chamber in nickel pipe in Cd. 2.8 " " "

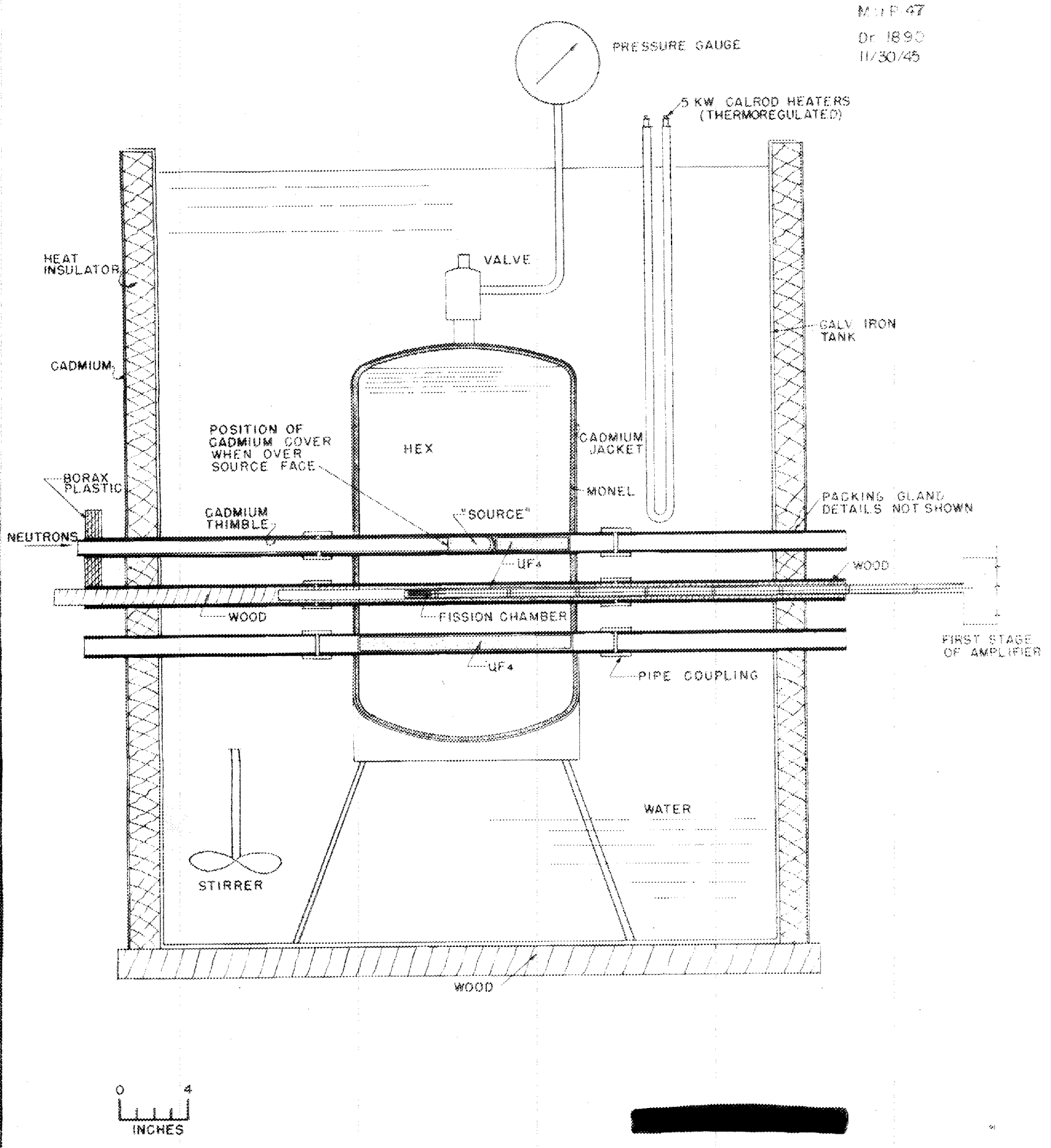
From these figures it is apparent that 57% of the cadmium-absorbable neutrons and about 88% of the epi-cadmium neutrons are transmitted by the tube wal The extent of the correction to be applied would seem to depend on the experimental conditions. For the case of water and no cadmium jacket about 73% of the fissions in the chamber were caused by the thermal neutrons; this can be seen in Table II by a comparison of the fissions per minute in the chamber with and without the cadmium jacket, in the presence of the water. Applying the pipe wall transmission corrections to the thermal and epi-cadmium neutrons separately, one arrives at an overall correction factor of 1.5. This factor was used for the corrected figures given in Table II for the cases of the bare drum surrounded with water. For the

other cases, the correction will be smaller because there are many fewer thermal neutrons. An accurate correction is impossible without more knowledge of the energy spectra of the neutrons, and we have merely applied a correction factor of 1.12.

These corrections were applied in evaluating the figures given in column 5 of Table II from those given in column 4.

[redacted]

M.U.P. 47
Dr. 1890
11/30/45



[redacted]

FIG. 1

FIG. 2.

EXPERIMENTAL POINTS AND DIFFERENCE CURVE

CONCENTRATION 1.1%
MIDDLE HOLE. CADMIUM JACKET.
WATER. SOURCE CENTERED.

DETECTOR WIDTH

SOURCE

NO CADMIUM ON SOURCE FACE

CADMIUM ON SOURCE FACE

DIFFERENCE

DRUM WALL

FISSIONS PER MINUTE PER MILLIGRAM OF U

1000
900
800
700
600
500
400
300
200
100

-7 -6 -5 -4 -3 -2 -1 0 1 2 3 4 5 6 7

DISTANCE IN INCHES FROM CENTER OF VESSEL

FIG. 3

DIFFERENCE CURVES

CONCENTRATION 1.1%
MIDDLE HOLE. SOURCE CENTERED.

Mon.P-47
Dr. 1892
11-30-45

DETECTOR WIDTH

N's → SOURCE

WATER REFLECTOR, NO CADMIUM JACKET

WATER REFLECTOR, CADMIUM JACKET

NO REFLECTOR

FISSIONS PER MINUTE PER MILLIGRAM OF 25

700
600
500
400
300
200
100

-6 -5 -4 -3 -2 -1 0 1 2 3 4 5 6

DISTANCE IN INCHES FROM CENTER OF DRUM

[redacted]

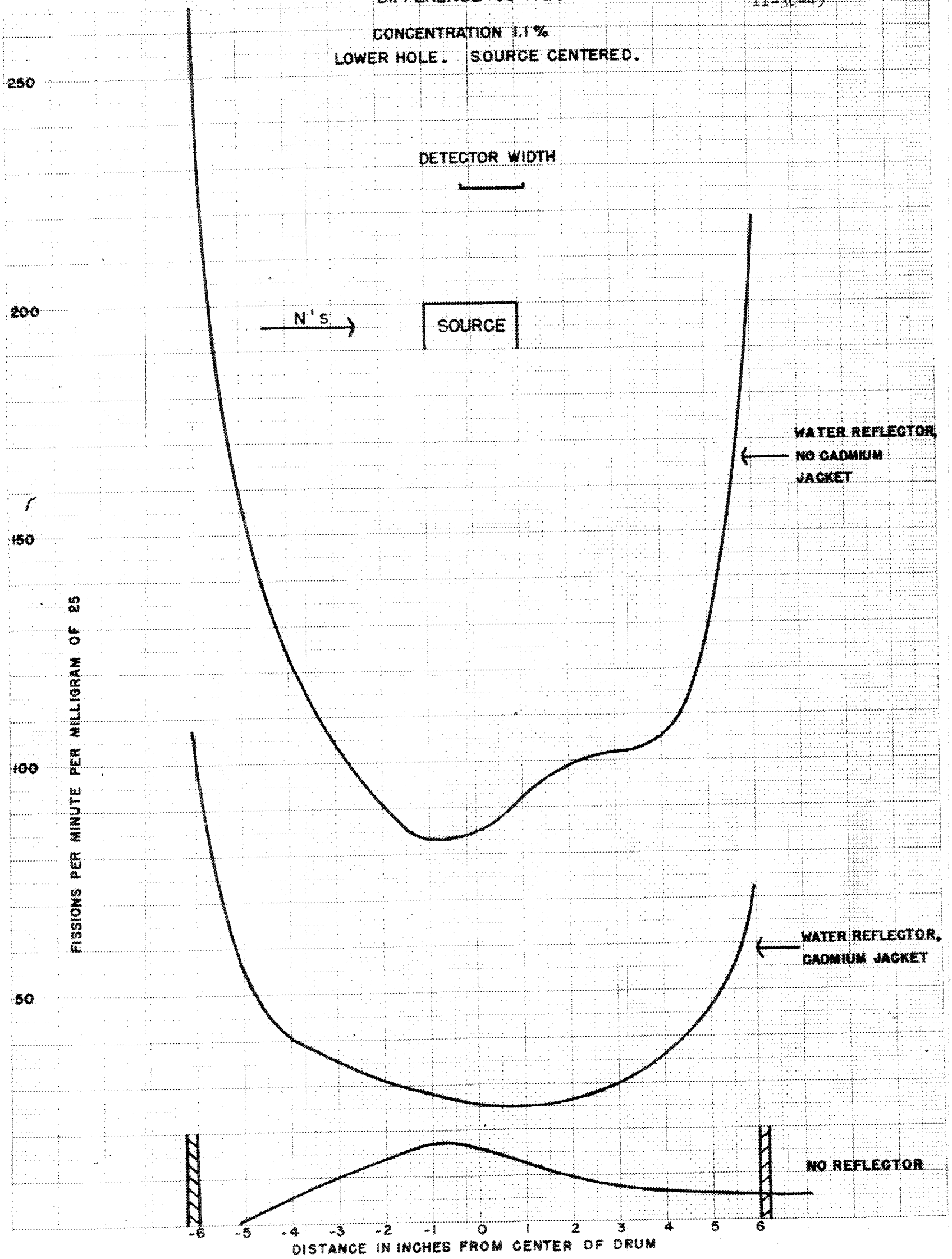
FIG. 4

MonP-47
Dr. 1893
11-30-45

DIFFERENCE CURVES

CONCENTRATION 1.1 %
LOWER HOLE. SOURCE CENTERED.

DETECTOR WIDTH



[redacted]

FIG. 5

DIFFERENCE CURVES

CONCENTRATION 1.1 %
CADMIUM JACKET. WATER.
SOURCE DISPLACED.

MonP-47
Dr. 1894
11-30-45

DETECTOR WIDTH

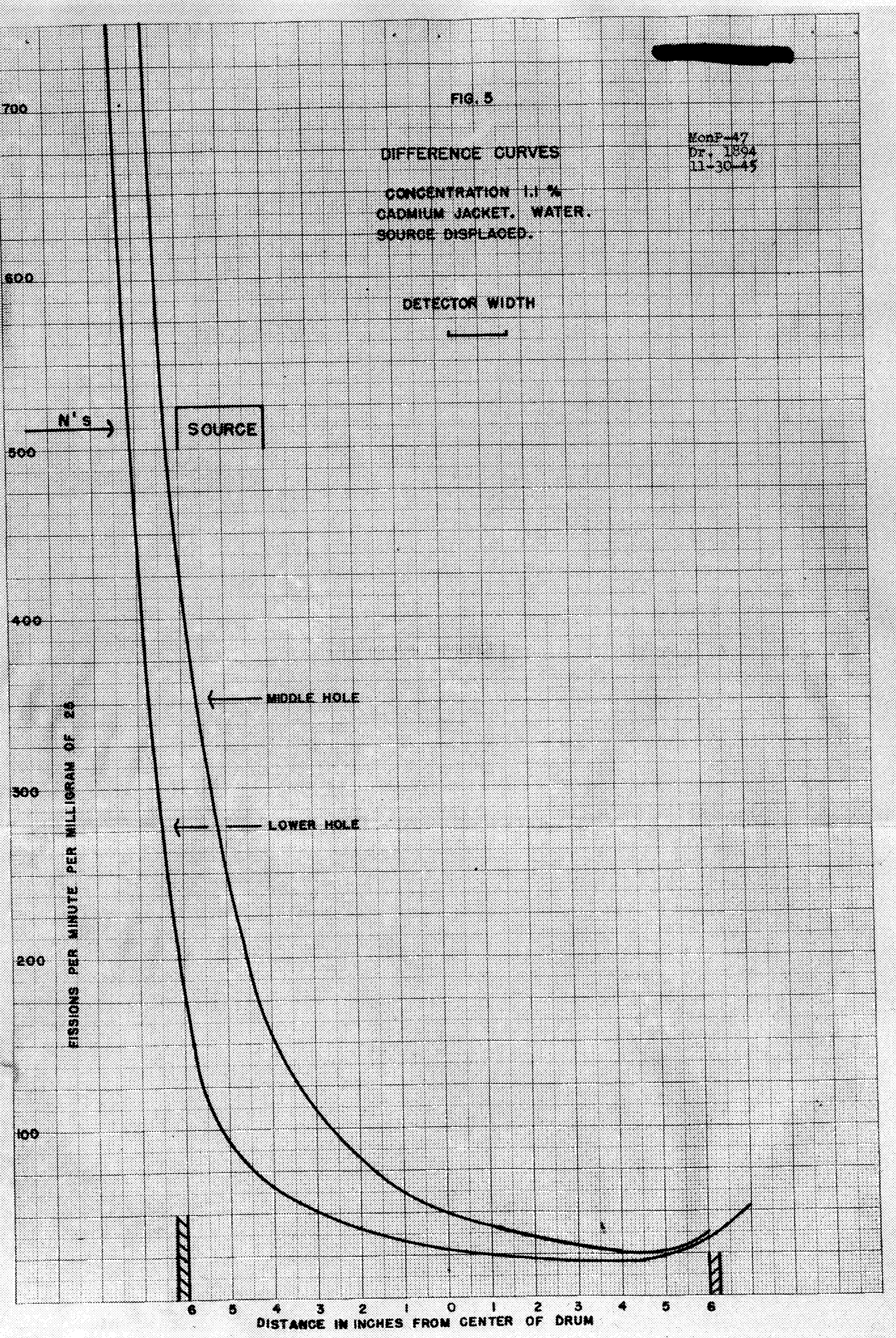


Fig 6

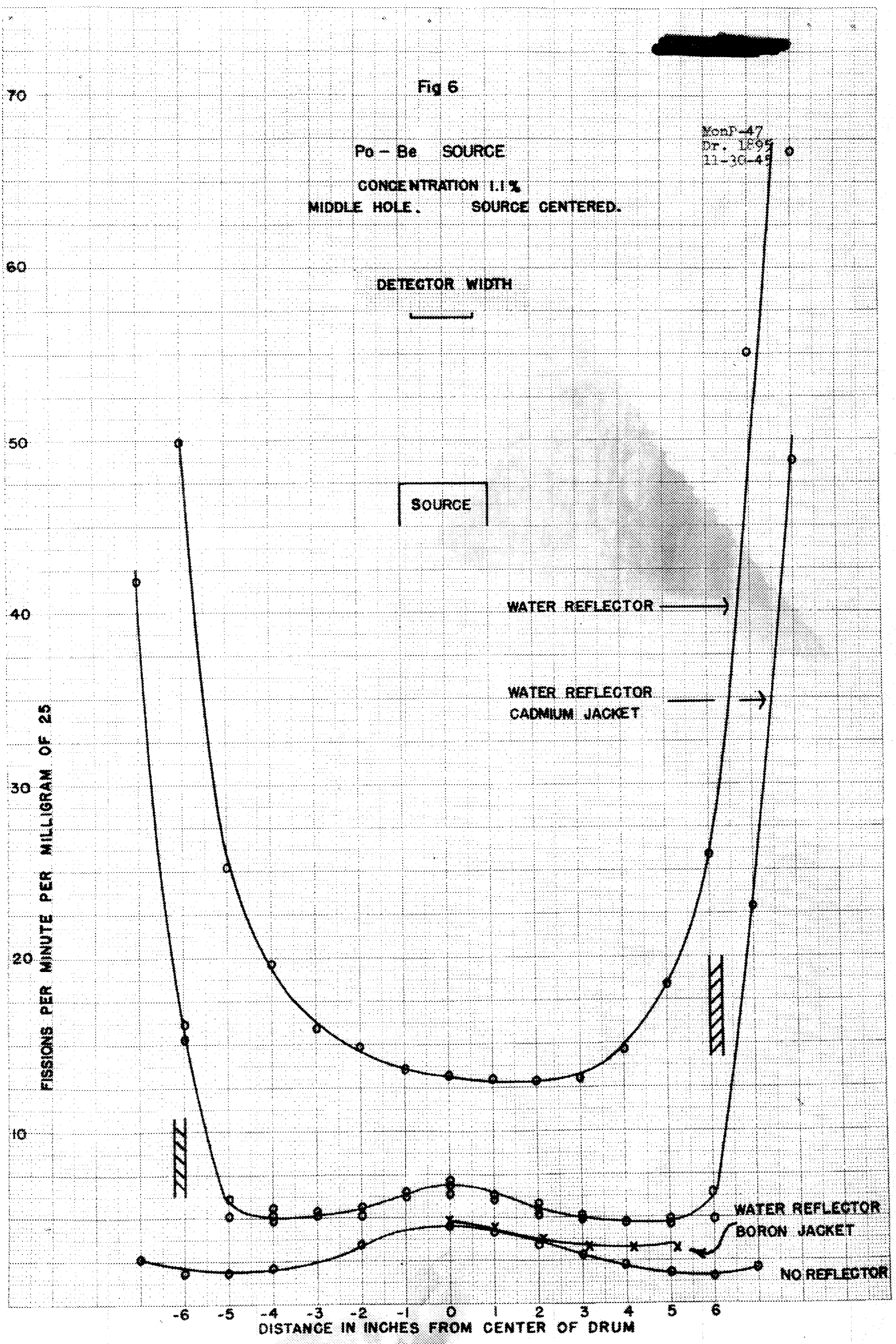

Po – Be SOURCE
CONCENTRATION 1.1 %
MIDDLE HOLE. SOURCE CENTERED.
DETECTOR WIDTH
SOURCE
WATER REFLECTOR
WATER REFLECTOR
CADMIUM JACKET
WATER REFLECTOR
BORON JACKET
NO REFLECTOR
FISSIONS PER MINUTE PER MILLIGRAM OF 25
70
60
50
40
30
20
10
-6 -5 -4 -3 -2 -1 0 1 2 3 4 5 6
DISTANCE IN INCHES FROM CENTER OF DRUM

Fig. 7

MonP-47
Dr. 1896
11-30-45

Po — Be SOURCE

CONCENTRATION 1.1 %

LOWER HOLE, SOURCE CENTERED.

DETECTOR WIDTH

SOURCE

← WATER REFLECTOR

WATER REFLECTOR
CADMIUM JACKET

WATER REFLECTOR
BORON JACKET

NO REFLECTOR

FISSIONS PER MINUTE PER MILLIGRAM OF 25

25

20

15

10

5

[redacted]

Fig. 8

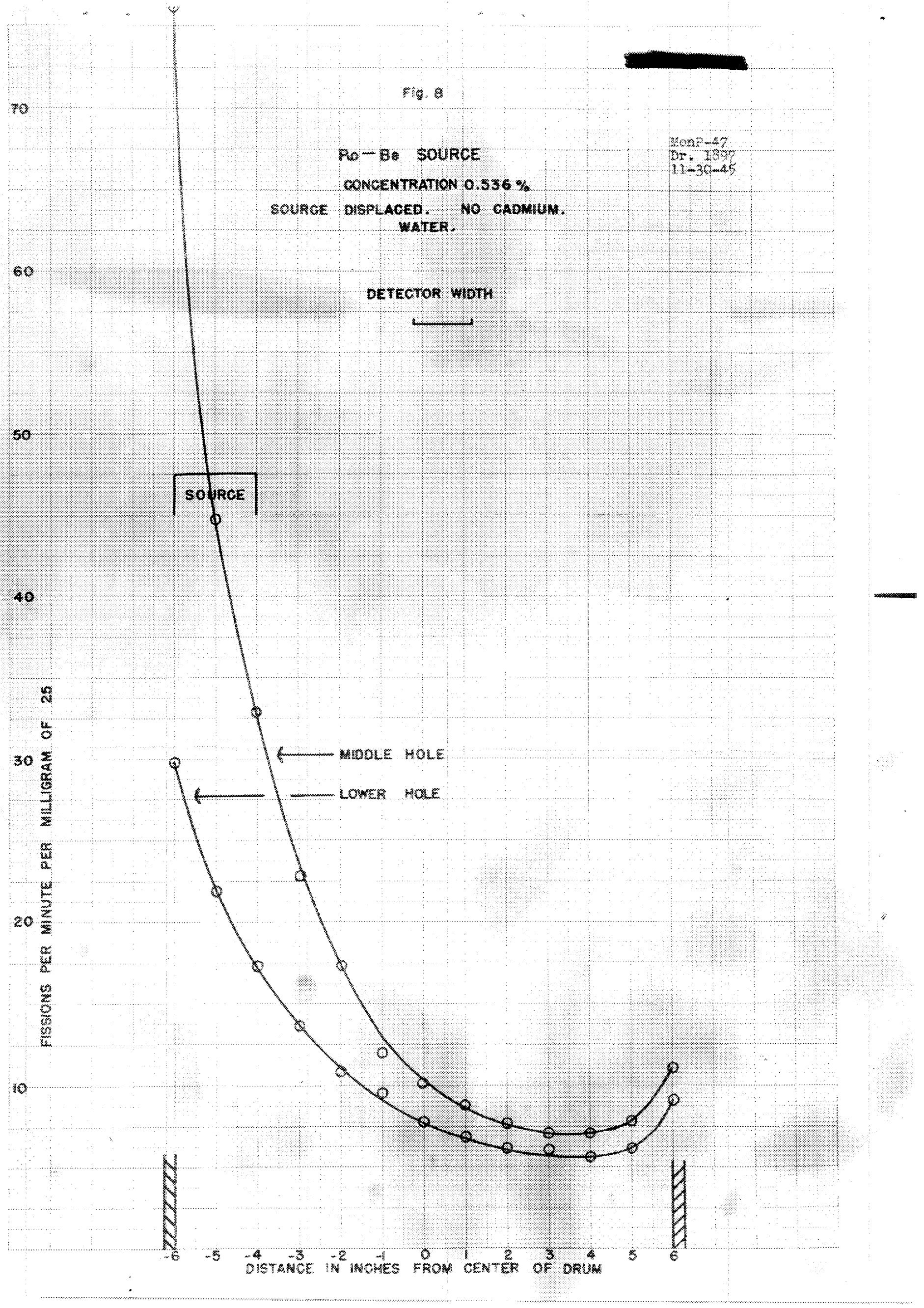
MonP-47
Dr. 1897
11-30-45

Po — Be SOURCE

CONCENTRATION 0.536 %

SOURCE DISPLACED. NO CADMIUM. WATER.

DETECTOR WIDTH



[redacted]

Fig. 9

MonP-47
Dr. 1898
11-30-45

Po - Be SOURCE

CONCENTRATION 0.536 %

MIDDLE HOLE. SOURCE CENTERED.

DETECTOR WIDTH

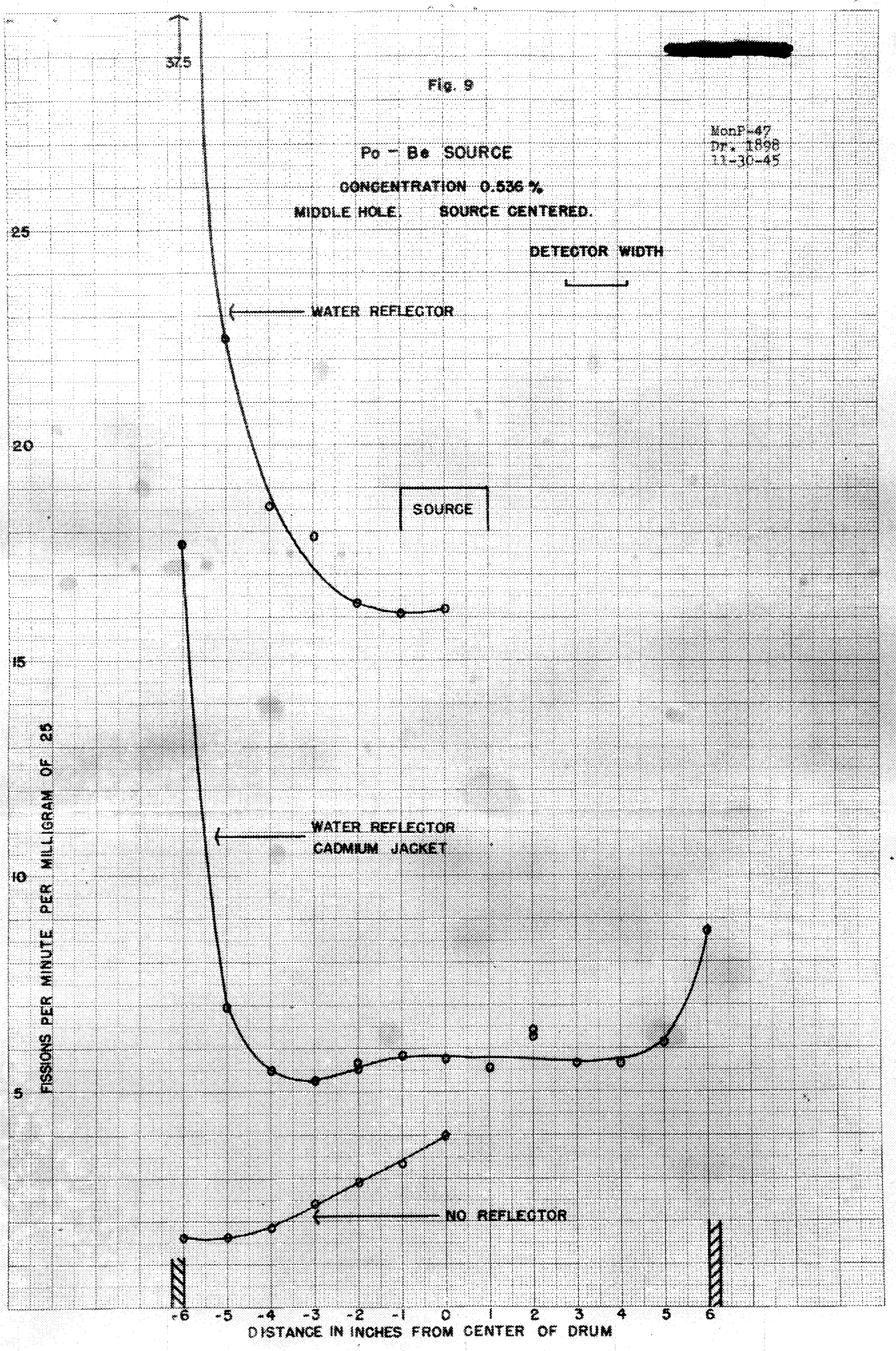


Fig. 10

MonP-47
Dr. 1899
11-30-45

Po – Be SOURCE

CONCENTRATION 0.536 %

LOWER HOLE, SOURCE CENTERED.

DETECTOR WIDTH

SOURCE

WATER REFLECTOR

WATER REFLECTOR
CADMIUM JACKET

NO REFLECTOR

FISSIONS PER MINUTE PER MILLIGRAM OF 25

25
20
15
10
5

-6 -5 -4 -3 -2 -1 0 1 2 3 4 5 6

DISTANCE IN INCHES FROM CENTER OF DRUM