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ABSORPTION OF THERMAL NEUTRONS IN URANIUM

by

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Abstract

A knowledge of the absorption processes for neutrons in uranium is important for planning a chain reaction experiment. The absorption of thermal neutrons in uranium and uranium oxide has been studied. Neutrons from the cyclotron were slowed down by passage through a graphite block. A uranium or uranium oxide sphere was placed at various positions in the block. The neutron intensity at different points in the sphere and in the graphite was measured by observing the activity induced in detectors of uranium oxide or manganese. It was found that both the fission activity in the uranium oxide and the activity induced in manganese was affected by non-thermal neutrons. An experimental correction for such effects was made by making measurements with the detectors surrounded by cadmium. After such corrections the results from three methods of procedure with the uranium oxide detectors and from the manganese detectors were consistent to within a few per cent. If f, the density of thermal neutrons in the absorbing material, is given by $\Delta f = \varkappa^2 f$, the experimental data are shown to give a value $\varkappa = 0.41$ cm^{-1} for a U metal sphere of density 8.6 in contrast to 0.35 cm^{-1} from constants given by Fermi and his collaborators. For a U_3O_8 sphere of density 5.3 the data give $\varkappa = .24$ cm^{-1} in agreement with Fermi's results. Possible sources of error have been discussed.

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Report on thermal neutron absorption for both metal and oxide.

Absorption coefficient depends on
1. Absorption cross section of constituent atoms.
2. Number atoms per c.c.
3. Scattering cross-section of atoms.

From calculated absorption coefficient for thermal neutrons and the measurements of resonance absorption it is the future intention to deduce the optimum dimensions for the typical cell in the proposed lattice.

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In report of June 1, 1941 "The Capture of Resonance Neutrons by a Uranium Sphere Imbedded in Graphite" Fermi and Anderson arrive at figure 4800 cm^3 for volume of black body equivalent with respect to resonance absorptions to a sphere 8.5 cm radius containing 9170 gm. U_3O_8. In Appendix B the proportion of absorption taking place in various portions of the sphere was investigated and it was found that absorption could be divided into two parts: surface effect and volume effect. This resulted in derivations of a formula giving the absorbing power for resonance neutrons of U_3O_8 spheres of radii greater or less than 8.5 cm.

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ABSORPTION OF THERMAL NEUTRONS IN URANIUM

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I. Introduction

Neutron absorption in uranium is believed to be principally of two general types: an absorption by U-238, chiefly without fission, in the thermal region and in a resonance region; and an absorption by U-235 which leads to fission. A detailed knowledge of these absorption processes is of interest in calculating the optimum size and spacing of the spheres to be used in the experiments on the chain reaction. The object of the present series of experiments is the study of the absorption processes in uranium and in particular is the accumulation of data from which can be calculated absorption coefficients for thermal and resonance neutrons.

The absorption of resonance neutrons was dealt with in a report submitted on June 1, 1941. In that report, Appendix A represented results of measurements carried out at Princeton jointly by the Columbia and Princeton groups on "The Capture of Resonance Neutrons by a Uranium Sphere Imbedded in Graphite." Appendix A (written by Fermi and Anderson) arrived at a figure of 4800 cm^3 for the volume of a black body equivalent with respect to resonance absorption to a sphere of 8.5 cm radius containing 9170 gm of U_3O_8. Appendix B investigated what proportion of the resonance absorption takes place in various portions of the U_3O_8 sphere. It was found that the absorption could be divided into a surface effect and a volume effect. This result made it possible to derive a formula giving the absorbing power for resonance neutrons of U_3O_8 spheres of radii greater than or less than 8.5 cm.

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Further studies of the absorption of resonance neutrons have been carried out at Princeton, with the important difference that the absorbing spheres were composed of compressed U_3O_8 and of uranium metal. Results of these studies will be described in a subsequent report. The present report deals with the absorption of thermal neutrons in both uranium metal and uranium oxide. A sphere of the substance to be investigated was placed in the interior of a graphite block. The surface of the block was exposed to an intense source of neutrons of about 4 Mev. The neutrons were slowed to thermal energies in the graphite and penetrated into the absorbing sphere. The relative number of thermal neutrons was measured at various points in the graphite and in the sphere. The measured falling off of the number of thermal neutrons toward the center of the sphere allows the determination of a certain absorption coefficient.

The absorption coefficient is a macroscopic property characteristic of the material of the sphere. It depends not only upon the absorption cross sections of the constituent atoms and upon their number per cubic centimeter but also upon their scattering cross sections. From the calculated absorption coefficient for thermal neutrons and the measurements of resonance absorption already reported for U_3O_8 and in progress for uranium metal and compressed U_3O_8, it is the intention to deduce the optimum dimensions for the typical cell in the proposed lattice.

The following measurements of density of thermal neutrons have been carried out in some cases by observing the radioactivity of the products resulting from fission by thermal neutrons; in some cases by

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measuring the activity induced in a detector made of manganese, which is sensitive to thermal neutrons; and in a few cases by separating out chemically the radioactive iodine resulting from fission produced by thermal neutrons. The three methods of measurement, when compared, were found to give results in satisfactory agreement with one another. However, certain internal discrepancies which appeared in the first experiments were only eliminated when it became apparent that two unexpected effects were present and would have to be taken into account. One of these effects is apparently fission produced in the uranium detectors by neutrons of energy between 1 ev and 1000 ev. It was corrected for in later experiments by covering the uranium detectors with cadmium and subtracting the activity so obtained from the total. The other important effect revealed in the first experiments was activity produced in the manganese detectors by non-thermal neutrons. Cadmium shielding also made it possible to correct for this effect in later measurements. Neither effect was expected on the basis of what was previously known. The apparent production of fission by epi-thermal neutrons is of considerable interest. Further experiments are in progress to investigate this point in more detail.

The cyclotron was used as the source of neutrons. In the previous experiments* beryllium was bombarded at the target chamber with about one microampere of 8 Mev protons. This corresponded to about 3500 curies radon beryllium equivalent. Lately it has been found that the neutron intensity at the graphite block is increased by a factor of about ten if the beryllium is bombarded internally on a probe; this increase occurs even though the probe is about 50 cm farther away from the block than is the usual target.

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*Preliminary report submitted to NDRC by H. D. Smyth on June 1, 1941.

The neutrons were slowed down, as in previous experiments, by a graphite block (61 x 61 x 91 cm), the front face (61 x 61 cm) of which was 60 cm from the internal beryllium target. The uranium sphere was placed in a spherical cavity which could be located at various distances from the front face of the graphite block. Experiments were made on U_3O_8 spheres of 16.4 and 15.2 cm diameter and of density 4.0 and 5.5 g/cm^3 respectively, and on a pure uranium powder* sphere of 11.4 cm diameter and of density 8.60 g/cm^3. In each sphere was a cylindrical cavity about one cm in diameter which extended somewhat past the center as shown in Fig. 1. As the same procedure was used for all spheres, we shall speak only of the uranium sphere except in the statement of the final results.

II. Absorption of Fission-Producing Neutrons

We used four different methods for measuring the distribution of fission-producing neutrons. These methods will be described in this section. The first will be called the Wide Sample Physical Method, the second the Narrow Sample Physical Method, the third the Manganese Method, and the fourth the Chemical Method.

Wide Sample Physical Method

The cavity in the uranium sphere was filled to the surface with U_3O_8 which had just been purified from U X and which was contained in a cylindrical bag of cellophane that fitted snugly into the cavity. Purification of the U_3O_8 from U X was carried out by the standard ether extraction method recently described by Anderson.[1]

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*We were informed by Dr. Szilard that the U metal contained 50 - 200 parts of boron per 1,000,000 parts of metal.

1. H. L. Anderson, Resonance Capture of Neutrons by Uranium.

The average number of neutrons present just outside the sphere was measured by a thin purified U_3O_8 detector, the center of which was placed 15 cm from the center of the sphere as shown in Fig. 1. In some cases, as a check on the method, two detectors were placed together. To correlate experiments made with the sphere at different positions in the graphite block, a purified U_3O_8 detector was placed each time at the position (M) indicated in Fig. 1. This detector will be referred to as the monitor.

With the sphere placed in the graphite so that the U_3O_8 cylinder was horizontal and parallel to the front face of the block, a neutron exposure of several hours was made. The cellophane tube containing the U_3O_8 was then withdrawn and cut into four or five measured segments. The U_3O_8 from one of these sections was removed and finely ground using an agate mortar and pestle. Several drops of a dilute solution of collodion in amyl acetate were mixed with the ground-up U_3O_8 and about 50 mg of the mixture were then painted thinly and uniformly over an area of 4.5 x 6.5 cm on a piece of paper 6.0 x 6.5 cm which was subsequently covered with Scotch Cellulose Tape. It was in just this way that the monitor and outside neutron detector had been prepared from the purified but unexposed U_3O_8. The U_3O_8 from the other sections was treated in the same way and the activities of the samples prepared from the sections of the cylinder together with the monitor and detector were observed continuously one after another for a period of from five to seventeen hours. The activity of a sample was measured by enclosing it in a lucite tube which could be slipped over a thin-walled silvered glass Geiger-Muller counter. About 20,000 counts were taken for each measurement. The consistency of the Geiger-Muller counter was checked at frequent

intervals using a standard uranium sample and, as a further check, an unexposed sample of purified U_3O_8 was also followed.

In some of the earlier experiments, the recording equipment, a hard tube scale of thirty-two and a Cenco mechanical counter, missed counts at counting rates above 5000 counts per minute. This was ascertained by observing the decay of a sample of iodine which had been exposed to neutrons. The decay curve is plotted in Fig. 2. It is seen that, at low counting rates, the activity decays with a 25-minute half-life as it should, but that, at higher counting rates, the curve deviates from the straight line drawn through the points at low rates. The difference between the straight line and the observed curve is a measure of the counts lost, and was applied as a correction to the data. Usually the correction amounted to but a few per cent. In the later experiments the resolving time was decreased so that no counts were lost below 20,000 counts per minute.

In Fig. 5 the measured activities of a typical run are plotted on semi-log paper as a function of the time after the end of the exposure. Presumably the measured activities consist of three parts, that from U-239 of half-life 24 minutes, that from the fission products, and that from U X newly generated by the uranium. The first two activities will die out, the 24-minute activity rapidly, the fission activity more gradually, but the U X activity will grow, giving the upward turn to the curves. To obtain the amount of U X activity to be subtracted, the samples were measured several days later when the U X had grown to such an extent that the fission products were negligible. The U X should grow as $A\ (1-e^{-\lambda t})$ where A is a constant proportional to the mass of uranium in the sample, λ is the decay constant of U X, and t is measured

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from t_o which is approximately the time of U X separation — actually a few hours earlier as the separation is not complete. For about a day after the separation the growth is very nearly linear. The activity of an unexposed sample was followed, and when these measurements were plotted as a function of time, the straight line obtained could be accurately extrapolated to obtain t_o. After a day or so the growth is no longer linear but can be closely approximated by $At\ (1 - 0.59 \times 10^{-3} t)$ where t is measured in hours. It was found expedient to plot the observed activity plus the product of it by $0.59 \times 10^{-3} t$ as this should reduce the data to a straight line continuation of that which would be obtained at earlier times. Such growth curves are shown in Fig. 4. It is seen that the curves are essentially straight lines after a couple of days, and that all of these lines can be extrapolated back to very nearly the same zero time given by the unexposed sample. In some cases, the activities of the monitor and detector were measured before the neutron exposure. The slopes of the straight line part of the build-up curves for all samples were measured. These are proportional to the masses of U_3O_8 in the samples, and enable one to calculate how much to subtract from the earlier measurements in allowing for the U X activity.

In Fig. 5, the activities measured within the first twenty hours after exposure are plotted after the subtraction of the U X contribution. If the neutron energy distribution at each sample had been the same, one would expect these curves to be parallel, i.e., the ratios of the activities of different samples to be independent of time. Actually the neutron energy distribution was not the same and therefore the relative proportions of the 24-minute U-239 activity and of fission products of various life-times will not be the same. The effect of the

24-minute U-239 can be ignored after about four hours when it has fallen to less than one tenth of one per cent of its first measured value. After this time, if the only activity were from fission products created by thermal neutrons, the ratios of the activities of various samples should remain constant in time and should give the relative intensities of thermal neutrons at the positions of the various samples. For samples taken from the sphere these ratios did remain constant to within a few per cent over a range of about 15 hours as can be seen from Fig. 5. However, the ratio of the detector and monitor intensities to those of the sphere samples increased in the same time range by as much as ten per cent, although the ratio of the detector activity to the monitor activity remained constant. This result indicates that we are dealing with an effect that does not arise entirely from thermal neutrons and, as might be expected, that the neutron energy distribution at the monitor and detector are about the same but differ from that prevalent throughout the sphere. Since this effect does not enter into the question of the relative activities within the sphere, its further consideration will be postponed until after the final reduction of data has been explained and the results of this group of experiments presented.

In Fig. 5 we have the activity of various samples corrected for U X and the 24-minute U-239. In Fig. 4 we have the U X activity build-up curves of the various samples. The slopes of these build-up curves are proportional to the amount of uranium in the samples so that dividing the activities at a given time (Fig. 5) by the slopes of the corresponding build-up curves gives the relative activity per mg of the various samples. Fig. 6 exhibits the result of three separate runs made with the center of the sphere located 20 cm from the front face of

the graphite block. The results are presented in the form of histograms where the relative fission produced (taken four hours after the neutron exposure) per mg is plotted as a function of the distance from the center of the sphere. The relative strengths have been adjusted to unity at the center of the sphere; and the monitor and detector activities are indicated by the horizontal lines. It is seen that the agreement is to within three per cent for all points within the sphere, and that the agreement with the results of the chemical method, which is also shown and which will be explained presently, is about the same except for the detector activity which is about seven per cent lower.

As an additional check on the method, in a few of the experiments, two outside detectors were placed together. The relative fission activity per mg of the two samples was the same to within one per cent. The average results of runs made with the uranium sphere at 10, 20, 30, 40, and 60 cm from the front face of the graphite block are shown in Fig. 7a. In Fig. 7b are shown the results of runs made with two different uranium oxide spheres. Thus, Figs. 7a and 7b present the data from this first group of experiments. Before reporting other methods and results, we will consider further the curves in Fig. 5.

It was mentioned before that the decay curves of the samples inside the sphere were not parallel to those of the detector and monitor samples (Fig. 5). Furthermore, it is noted in Fig. 7a that the absorption of neutrons appears appreciably less when the sphere is closer to the front face of the graphite block. This effect might be attributed to the 2.3-day activity of element 93, which is the daughter substance of U-239. An estimate indicates the number of 93 disintegrations per minute comparable to those of the fission products, after four hours, but they

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will contribute considerably less to the measured activity because the maximum energy of the electrons of 93 is .47 Mev while the maximum energy of those of the fission products is about 1.2 Mev. Therefore a much greater fraction of the 93 electrons will be absorbed in the counter wall. To see how much of the activity was due to element 93 or fission products resulting from fast neutron collisions, we enclosed in cadmium the uranium oxide cylinder and the detector but not the monitor. The usual exposure was made and the samples were prepared, measured, and the data worked up as before. The curves for the samples enclosed in cadmium were flatter than that for the unenclosed monitor, but still showed the slow change of half-life that is characteristic of fission products instead of a simple composition of 24-minute and 2.3-day component curves. This would indicate fast neutron fission and its nature will be discussed later. The relative activities per mg four hours after the end of the exposure, due to whatever cause, were reduced to the same monitor activity in the experiments with and without cadmium. Then a corrected absorption curve was obtained by subtracting the cadmium-covered relative activities from the non-cadmium-covered values. The relative values so obtained with the sphere at 20 cm from the front face are shown in Fig. 8. When the above experiment was repeated, but with the sphere 60 cm from the front face of the graphite block, no measurable activities except those due to U X were observed in the samples covered with cadmium. Furthermore, the difference curve obtained with the sphere at 20 cm is in reasonable agreement with the absorption curve obtained with the sphere at 60 cm if the width of the samples is taken into account. This was to be expected since both activities are caused by thermal neutrons produced outside the sphere. The result was checked also by the Mn method (see

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below) and forms the basis of the theoretical interpretation.

Narrow Sample Physical Method

A modification of the general method described above was to prepare all the samples in a form suitable for measurement before the exposure. These samples were prepared as before except they were one cm wide and 5.5 cm long. They were then wrapped around a thin-walled brass tube so that they could be slipped snugly into the cavity in the sphere. The brass tube was filled with uranium to the surface of the sphere, the rest of the space being filled with a carbon plug. The advantages of this arrangement are that the positions of the samples are more definite and that more points can be taken. Indeed, many samples were placed in the graphite at intervals of a few centimeters on a horizontal line parallel to the front face and extending from the center of the sphere to the edge of the block.

In this modified procedure, as in the earlier method, the samples were measured after the neutron exposure. However, the weight of the U_3O_8 in the samples could now be obtained by igniting and then weighing them rather than by the more tedious device of following the build-up of U X. This method was not possible with the larger sized samples used before because the counter wall did not have uniform thickness over its length. This is illustrated in Fig. 9 where the counts per minute of a narrow source of uranium in equilibrium with the U X is plotted as a function of the position of the source along the counter. It is seen that only over a short distance near the middle of the tube is the thickness uniform. The samples were placed in this region. By counting small weighed uranium samples in equilibrium with U X, it was possible to calculate the amount of U X activities to subtract from activities of the

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exposed samples. To determine t_o, the growth of an unexposed sample of the purified uranium was also followed. Results of two exposures using this method are shown in Fig. 10.

Manganese Method

Thermal neutron distributions in and around the sphere were also obtained by substituting MnO_2 in place of the uranium oxide in the narrow samples used in the method just described. The decay curves of these samples after exposure all displayed a pure 2.59-hour half-life as is shown in Fig. 11 for a typical experiment. The relative neutron density was then obtained by dividing the intensity of a sample by its weight. The weight was obtained by piling all the samples at one place in the graphite block, exposing them to neutrons, and then measuring the activity induced. To obviate absorption by the samples themselves, several other exposures were made with the samples piled in different orders. An average of the resultant intensities was taken as the relative weight.

When the manganese samples were exposed covered with Cd, considerable activity was still noticed. Hence, in some cases, a second exposure was made with the samples in the same positions except that all but the monitor sample were covered with cadmium. Thus the non-thermal component could be subtracted. The non-thermal part will be discussed later. Figs. 12a, b, c, d, e show thermal neutron distributions so obtained in the graphite block in various directions with and without the uranium spheres. Fig. 8 shows the agreement with the Physical Method in those cases when the non-thermal components have been subtracted.

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Chemical Method

In the chemical method of measuring the fission production, a cylinder of purified U_3O_8 was prepared for exposure in the same way previously described. The outside detector sample of several grams was contained in a thin cellophane envelope. It was found necessary to make a neutron exposure of about six hours in order to get measurable activities. After the exposure, the cellophane cylinder was taken from the sphere and cut into measured segments as before. The U_3O_8 from each segment was then weighed. The U_3O_8 which constituted the outside detector was mixed and then divided into two parts which were also weighed. To each sample was added an equal weight of iodine to serve as a carrier, and then a chemical separation of iodine was made. This separation, ~~which is described in the appendix,~~ yielded silver iodide which was finely ground up, mixed with alcohol, and painted on a weighed piece of aluminum over an area of 2 x 6 cm. A second weighing yielded the weight of the silver iodide which indicated the efficiency of the separation. The chemical separation of all the samples was made as nearly simultaneously as possible. The order of the samples was the same for every operation! First we took part of the detector sample, then the samples from the sphere, and finally the other fraction of the detector sample. Thus any error introduced because of not making the separations exactly simultaneously would be indicated by different activities per mg in the two detector fractions.

After the samples were prepared for measurement in the usual way, their activities were followed for several hours. These were plotted on semi-log paper, and the smooth curves so obtained were closely parallel. The activities of the samples at a given time were read from

SUMMARY OF FIGURES

Fig. No.	Exp. No.	Type	Sphere Material	Sphere Density	Sphere Radius	Sphere Position (Z)	Bomb. Time	Method	Remarks
1									Experimental arrangement
2									Correction for finite resolving time of counter
3	4	Fission	U Metal	5.7	8.6	20 cm	2-1/3 H	Wide Sample	Total activity curves
4	4	Fission	U Metal	5.7	8.6	20 cm	2-1/3 H	Wide Sample	Growth of UX
5	4	Fission	U Metal	5.7	8.6	20 cm	2-1/3 H	Wide Sample	Activity with UX subtracted
6	3 4 9	Fission	U Metal	5.7	8.6	20 cm	2-1/3 H 2-1/3 H 6-1/4 H	Wide Sample	Comparison of physical and chemical methods
7A	14) 17)	Fission	U Metal	5.7	8.6	10 cm	2 H	Wide Sample	Summary of wide sample results, with U metal sphere, uncorrected for non-thermal fission
7A	3) 4) 9) 18)	Fission	U Metal	5.7	8.6	20 cm	2-1/3 H 2-1/3 H 6-1/4 H 2 H	Wide Sample	
7A	10) 15) 22)	Fission	U Metal	5.7	8.6	30 cm	? 2 H 2-2/3 H	Wide Sample	
7A	6	Fission	U Metal	5.7	8.6	40 cm	2 H	Wide Sample	
7A	5) 16)	Fission	U Metal	5.7	8.6	60 cm	2-1/2 H 4 H	Wide Sample	

Fig. No.	Exp. No.	Type	Sphere Material	Sphere Density	Sphere Radius	Sphere Position (Z)	Bomb. Time	Method	Remarks
7B	8) 12) 13)	Fission	U_3O_8	8.2	4.0	20 cm	1 H 2 H 4 H	Wide Sample	Results with two U_3O_8 spheres
7B	38	Fission	U_3O_8	7.6	5.3	60 cm	4-1/6 H	Narrow Sample	
8	5) 4) 9)	Fission	U Metal	5.7	8.6	20 cm	2-1/3 H 2-1/3 H 6-1/4 H	Wide Sample	Thermal fission and thermal manganese activities in U metal sphere
	18						2 H		
8	5) 16)	Fission	U Metal	5.7	8.6	60 cm	2-1/2 H 4 H	Wide Sample	
	22						2-2/3 H		
8	49	MnO_2	U Metal	5.7	8.6	60 cm	1-1/2 H	MnO_2	
	35						1 H		
9									Counter sensitivity
10A	50	Fission	U Metal	5.7	8.6	40 cm	4-1/12 H	Narrow Sample	Narrow sample results
10B	38	Fission	U_3O_8	7.6	5.3	60 cm	4-1/6 H	Narrow Sample	
11	43	MnO_2						MnO_2	MnO_2 decay curves
12A	43 28	MnO_2	No Sphere U Sphere	5.7	8.6	20 cm	1-3/4 H ?	Narrow Sample Narrow Sample	Mn neutron distribution in block along Z (thermal + resonance)
12B	29	MnO_2	U Metal	5.7	8.6	20 cm	2/3 H	Narrow Sample	Mn neutron distribution across block with sphere (thermal + resonance)

Fig. No.	Exp. No.	Type	Sphere Material	Sphere Density	Sphere Radius	Sphere Position (Z)	Bomb. Time	Method	Remarks
12C	33	MnO_2	U Metal	5.7	8.6	40 cm	1 H	Narrow Sample	Mn neutron distribution across block with sphere (thermal + resonance)
12D	35 49	MnO_2	U Metal	5.7	8.6	60 cm	1 H 1-1/2 H	Narrow Sample	Mn neutron distribution, corrected for non-thermal part.
12E	37	MnO_2	U_3O_8	7.6	5.3	60 cm	1-1/2 H	Narrow Sample	

the curves. The relative fission activity per mg of a U_3O_8 sample was then found by dividing the activity obtained from the curve by the corresponding weight of the U_3O_8 and by the weight of the silver iodide of the measured sample. The results of such a run made with the uranium sphere at 20 cm from the front face of the graphite block have been shown in Fig. 6, where it was seen that they agreed to within seven per cent with the results obtained by the physical method. The two parts of the outside detector agreed with each other to within five per cent.

Because the internal consistency of the results of the chemical method was not so good as the results given by the physical methods, and because the labor and time of bombardment involved was much greater, it was decided not to perform more extensive experiments using the chemical method.

III. Evaluation of the Macroscopic Absorption Coefficients

The thermal neutrons obey in an absorbing material, apart from the immediate neighborhood of the surface, the equation

$$\Delta f = \varkappa^2 f \qquad (1)$$

where f is the density of neutrons and $\varkappa$ is given, with a sufficient accuracy, by the equation

$$\varkappa = \sqrt{3\sigma\sigma_a}\left(1 - \frac{2\sigma_a}{5\sigma}\right). \qquad (1a)$$

Herein σ and σ_a are total and absorbing cross sections contained in unit volume. The above described experiments will be used to determine $\varkappa$ for our U metal of density 8.6 to be

$$\varkappa = .41\ cm^{-1} \qquad (2)$$

This is sufficient for the calculation of the neutron density within a sphere in the "final arrangement" where the disposition of material can be considered to be spherically symmetric about the center of the sphere. Thus the density of thermal neutrons in the final arrangement will be, in the inside of the sphere, given by

$$f = \text{Const.} \frac{1}{\varkappa r} \operatorname{Sh} \varkappa r \approx \text{Const.} \left(1 + \frac{\varkappa^2 r^2}{6} + \frac{\varkappa^4 r^4}{120}\right). \tag{5}$$

For our experimental arrangement, (5) is not accurately obeyed because the distribution of the neutrons may deviate from spherical symmetry. This effect will be discussed and shown not to be large enough substantially to influence our results. It will be shown, furthermore, that the validity of (1) (and hence of (5) in the final arrangement) should extend practically to the surface, i.e., that the surface effect is very small. It will be noted finally that the energy distribution of thermal neutrons is not very sharp but extends over a considerable range. This necessitates some modification of (1) and (5) which will be considered in some detail in a later report.

Our result (2) is a good deal higher than that which follows from the constants given by Fermi and collaborators, i.e.,

$$\sigma = 18 N \cdot 10^{-24} \qquad \sigma_a = 6 N \cdot 10^{-24} \tag{4}$$

where $N = 2.2 \cdot 10^{22}$ is the number of atoms per cm^3. The $\varkappa$ derived from (4) is 0.35 cm^{-1}.

Discussion of Experiments with U Metal Sphere

The density of thermal neutrons was obtained from three essentially independent experiments: fission activity inside the sphere when

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TABLE I

Ratio to activity at center	Distance from center in 60 cm exp	20 cm exp	Mn exp		Derived value of μ in cm^{-1} from 60 cm exp	20 cm exp	Mn exp
1	0	0	0	0	--	--	--
1.1	1.6 cm	1.7 cm	1.65 cm	0.765	0.48	0.455	0.465
1.2	2.25	2.35	2.3	1.065	0.47	0.455	0.465
1.3	2.85	2.9	2.95	1.29	0.45	0.445	0.440
1.4	3.3	3.45	3.55	1.475	0.445	0.430	0.415
1.5	3.65	3.95	3.95	1.62	0.445	0.410	0.410
1.6	4.3	4.4	4.3	1.76	0.41	0.400	0.410
1.7	4.65	4.75	4.65	1.87	0.40	0.395	0.410
1.8	4.95		4.8	1.975	0.40		0.410
1.9			5.0	2.05			0.410
2.0			5.2	2.17			0.415
2.1			5.4	2.25			0.415

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sphere was at 60 cm from source; same activity for 20 cm distance of sphere from source; activity of Mn detectors inside of sphere for 60 cm distance. The "source" means, in this connection, the front of the block, i.e., the place where the fast neutrons enter.

A separate experiment showed that there is no observable non-thermal fission activity induced in the sphere at the 60 cm position. In this case, the experimental material can be used without any correction. In the two other cases, the activity induced by non-thermal neutrons had to be subtracted from the whole activity.

In all cases, first of all, the histograms were replaced by smooth curves. Then the activity of the Cd covered samples (giving the non-thermal neutron's activity) was reduced to the same monitor intensity which was observed in the experiment with the uncovered sample. The reduced activity of the Cd covered sphere was subtracted from the activity of the uncovered sphere and the distances r determined at which the activity is 1.1, 1.2, 1.5 etc. times greater than the activity at the center. This gave, for the above described three experiments, the three sets of radii given in the second, third, and fourth columns of Table I.

The values of $\varkappa r$ for which $Sh\,\varkappa r / \varkappa r$ assumes the values of the first column are given in the fifth column and the ratio of this $\varkappa r$ to the r of the second, third, and fourth columns is given in the sixth, seventh, and eighth columns. These ratios should be all the same, i.e., $\varkappa$. It is seen that in all three experiments the values of $\varkappa$ decrease considerably from the center. However, the experimental accuracy is least ~~in these cases~~ near the center and the uncertainty of the magnitude of the activity at $r = 0$ may partly be responsible for this

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discrepancy. At higher r, the values rather consistently tend to .41. The agreement within the last values of one column does not mean, of course, much more than that they were all taken from a smooth curve, but the agreement between the three curves must be considered to be significant.

Experiments with U_3O_8 Sphere

The procedure of evaluation was the same as in the experiments with the metal sphere. Table II summarizes some of the results, viz. Exp. 10B and 12E. In the first one, the fission activity was measured directly at 60 cm from the front of the block; in the second, the Mn activity was measured at the same position and corrected by subtracting the fission produced by non-thermal neutrons. It is seen that the Mn experiment agrees with the $Sh\,\varkappa r/\varkappa r$ curve very satisfactorily. In fact, in view of the limited accuracy of the measurements, the agreement must be, to a large extent, accidental. The $\varkappa$ in the fission experiment increases with increasing r, a behavior which must be expected on theoretical grounds but which is surprising in view of the opposite behavior of the more accurate data used in Table I. On the whole, the data indicate a

$$\varkappa = .24\ cm^{-1} \qquad (5)$$

for this U_3O_8 sphere of density 5.8.

Discussion of Errors

The chief difficulty in the interpretation of the experimental data is the persistent increase of $\varkappa$ with increasing r. This behavior is manifested in the very accurate experiments summarized in Table I. It is, of course, possible that this behavior is due to the

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TABLE II

Ratio to activity at center.	Distance from center in Fig. 10	Distance from center in Fig. 12E		κ derived from Exp. 10B	κ derived from Exp. 12E
1	0	0	0	---	---
1.1	3.9	3.3	0.765	0.195	0.23
1.2	5.0	4.6	1.065	0.215	0.23
1.3	6.0	5.7	1.29	0.215	0.225
1.4	6.3	6.5	1.475	0.235	0.23
1.5	6.8	7	1.62	0.24	0.23
1.6	7.2	7.4	1.76	0.245	0.24
1.7	7.4-7.5		1.87	0.25	

inhomogeneity of the material, but it is impossible to discuss this possibility on theoretical grounds.

One might think, next, that the brass tube which contained the U_3O_8 for the analysis may be responsible for the effect. This brass tube had a diameter of 1.25 cm and a wall thickness of .08 cm. The absorbing cross section of Cu is 1.6×10^{-24} (F. Rasetti, Phys. Rev. 58, 869) that of Zn is about 1×10^{-24} (Coltman, ibid, 59, 917). Since the total concentration of Cu and Zn atoms is about four times higher in brass than that of U atoms in our metal, this gives a σ_a for brass which is just about equal to the σ_a of U. The total σ is, on the other hand, higher in brass than in our U metal by about 40 per cent. This corresponds to an increase in $\varkappa$ by 20 per cent. However, the oxide powder contained in the tube decreases the average $\varkappa$ of the center to an even greater extent than the brass increases it, and the cover of the tube has too little effect to explain the variation of $\varkappa$ by much more than 1 per cent.

The solution (5) holds only if the distribution of neutrons is spherically symmetric within the sphere. It is evident from the figures reproduced in the experimental part that this condition is not realized in the experiments. One has to consider, therefore, other solutions of (1), corresponding to higher spherical harmonics. It follows from the symmetry of the problem that among the spherical harmonics with $\mathcal{L} = 1$, only the $m = 0$ can enter corresponding to an angular distribution $\sim \cos\vartheta$. This, however, vanishes in the direction $\vartheta = 90^\circ$ in which the measurements were made. The effect of the next spherical harmonics ($\mathcal{L} = 2$) can be estimated as follows.

The solution in question is

$$C\,15\,(3\cos^2\vartheta - 1)\left(\frac{Sh\,\varkappa r}{\varkappa r} - \frac{3\,Ch\,\varkappa r}{\varkappa^2 r^2} + \frac{3\,Sh\,\varkappa r}{\varkappa^3 r^3}\right)$$

$$\approx C(3\cos^2\vartheta - 1)\left(\varkappa^2 r^2 + \frac{\varkappa^4 r^4}{14}\right) \qquad (6)$$

where C is a constant. Since $\varkappa r$ is about 2 on the boundary (6) gives $C \cdot 2\ (4 + 16/14) = 10.2\ C$ for $\vartheta = 0$; $-5.1\ C$ for $\vartheta = 90^\circ$; $10.2\ C$ for $\vartheta = 180^\circ$. The experimental values are, in one particular case (Fig. 12A, 12B) 2.0, 1.75, 1.6. The combination $f(0) - 2f(90^\circ) + f(180^\circ)$ gives 0 for the solutions with L = 0 and L = 1 and can be attributed to the L = 2 solution. It gives $0.1 = (10.2 - 2 \cdot 5.1 + 10.2)C = 10.2\ C$, i.e, $C = 0.01$. This will give the correct order of magnitude of C, but the experimental values are too inaccurate to exclude even a three times higher value for C.

Adding (6) to (5) gives

$$1 + \frac{\varkappa^2 r^2}{6} + \frac{\varkappa^4 r^4}{120} - C\left(\varkappa^2 r^2 + \frac{\varkappa^4 r^4}{14}\right) \qquad (7)$$

for the dependence upon distance of the activity at $\Theta = 90^\circ$. If one calculates for this function the product of the constant term and of the coefficient of r^4 and divides the result with the square of the coefficient of r^2, one obtains a quantity which is independent of $\varkappa$, viz.

$$.5(1 + 3.43\ C) \qquad (8)$$

The experimental value of this quantity, as derived from the figures underlying Table I, is around 0 and certainly not larger than .04. This would involve a $C \approx -1/3$ — which seems impossible.

There are two further effects which should be discussed in detail, viz. the surface effect and the effect that not all neutrons have the same velocity. Both discussions involve a fair amount of theoretical work and will be given, therefore, in another report. It should be mentioned, however, that it does not seem that they can explain the trend exhibited by $\varkappa$ in Table I and the only suggestion

we can offer for explaining this trend, therefore, is a possible inhomogeneity of the metal. The absence of the trend for the oxide sphere (Table II) appears to give some support to this view.

Discussion of Results

The value of $\varkappa$ given by (1) is incompatible with a total cross section $18 \cdot 10^{-24}$ cm^2. The minimum value of σ which is compatible with our $\varkappa$ is just this $\varkappa$ itself, which gives with $N = 2.8 \cdot 10^{22}$ the value $18.5 \cdot 10^{-24}$ cm^2 as the smallest possible value for the total cross section. However, this would involve $\sigma_a = \sigma$, $\sigma_{sc} = 0$, i.e., that U is black toward thermal neutrons. It is more reasonable, therefore, to use a somewhat higher σ, such as corresponding to a total cross section of $20 \cdot 10^{-24}$ cm^2. This gives

$$\sigma = 20\ N\ 10^{-24}; \qquad \sigma_a = 8.2\ N\ 10^{-24} \tag{9}$$

From these figures, the $\varkappa$ for the oxide of density 5.3 can be derived if one assumes $4 \cdot 10^{-24}$ cm^2 as the scattering cross section of oxygen and further assumes that oxygen has no appreciable absorption ($\sigma_a < 10^{-25} N\text{cm}^{-1}$) for thermal neutrons. This gives

$$\sigma = 0.35\ \text{cm}^{-1} \quad \text{and} \quad \sigma_a = 0.095\ \text{cm}^{-1} \tag{10}$$

for the oxide of density 5.3. These figures do not take possible contaminations of the metal into account. This gives a $\varkappa = 0.28$ cm^{-1} for the oxide, a value considerably larger than given by (5). This indicates either an even higher σ (and lower σ_a) than adopted in (9) or a considerable amount of absorption by contaminations in the metal. It is not necessary to discuss this since only the immediately measured $\varkappa$ enter into the consideration of the "final" arrangement.

The absorption coefficient of the oxide is in substantial agreement with the value which can be derived from the cross sections (4) given by the Columbia group.

Our thanks are due to Professors H. D. Smyth and J. A. Wheeler for their invaluable help in connection with the formulation of our results and the preparation of this manuscript.

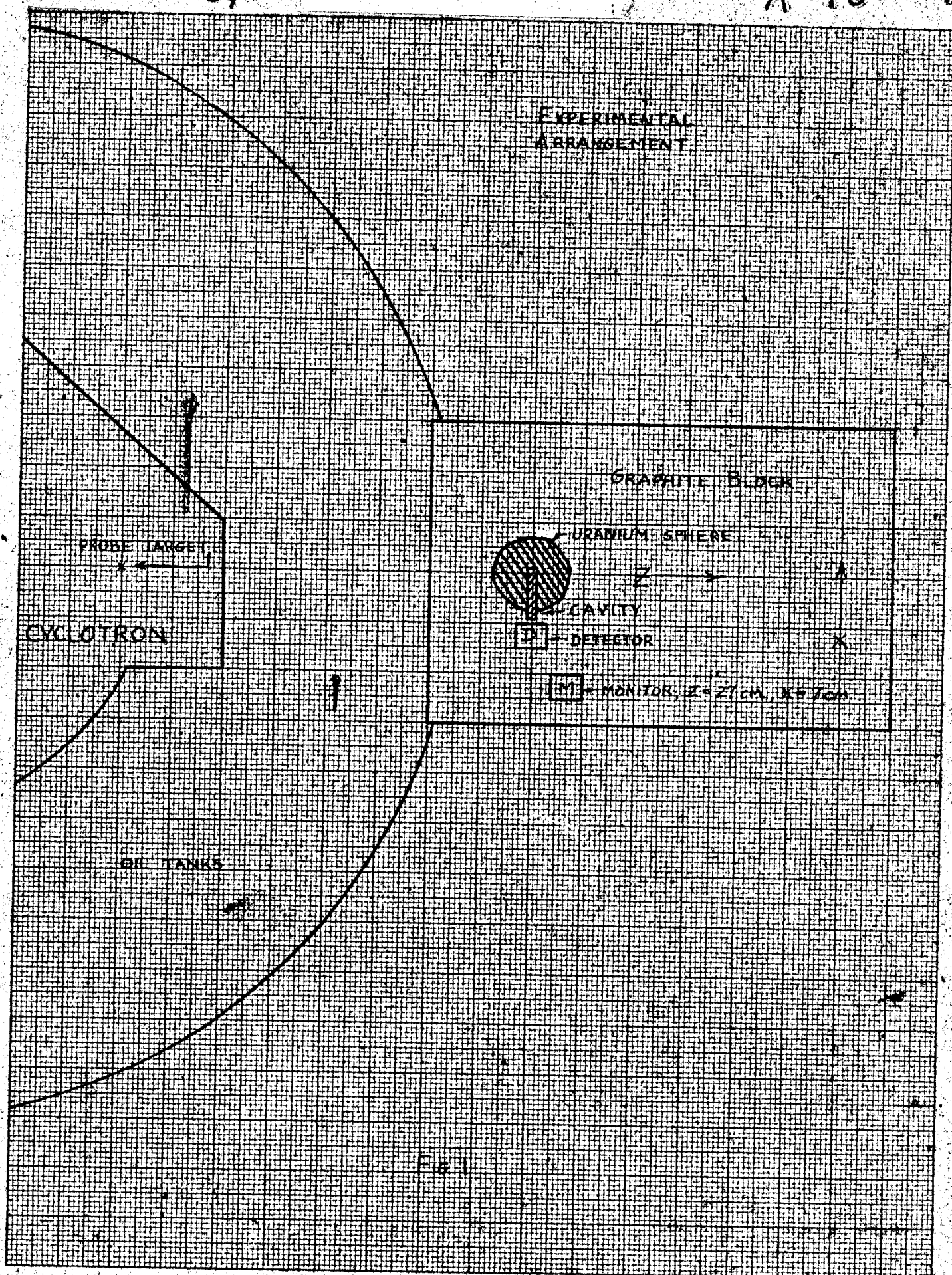


Fig.

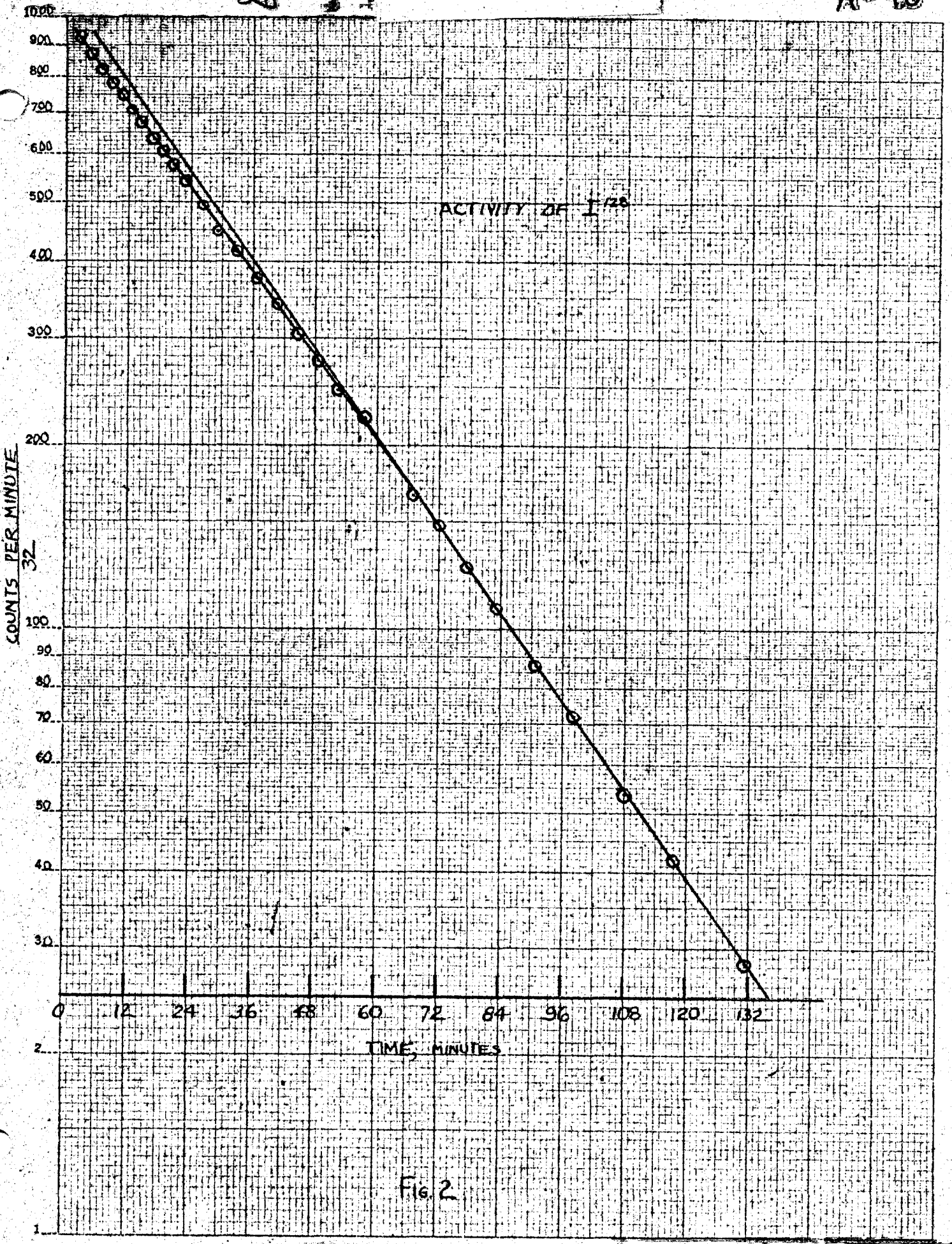


FIG. 2

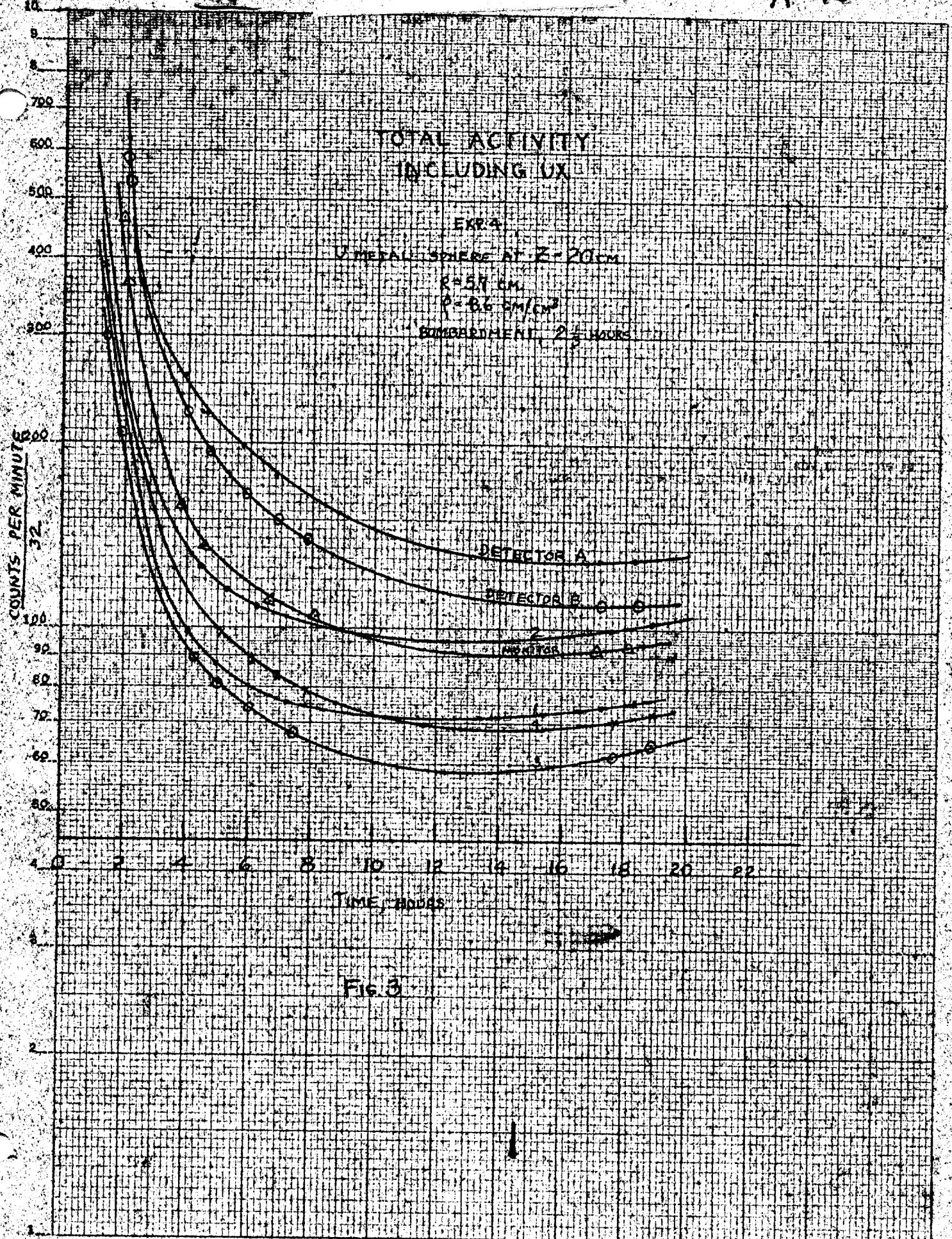

TOTAL ACTIVITY
INCLUDING UX
EKR 4
U METAL SPHERE AT Z = 20 CM
R = 5.1 CM
ρ = 8.6 GM/CM³
BOMBARDMENT 2½ HOURS
DETECTOR A
DETECTOR B
MONITOR
COUNTS PER MINUTE
32
TIME, HOURS

FIG. 3

A-40

EXP 4

GROWTH OF UX

UX METAL SPHERE AT $Z = 20$ cm.

$R = 5.7$ cm.

$\rho = 8.6$ gm/cm^3

BOMBARDMENT $2\frac{1}{3}$ HOURS

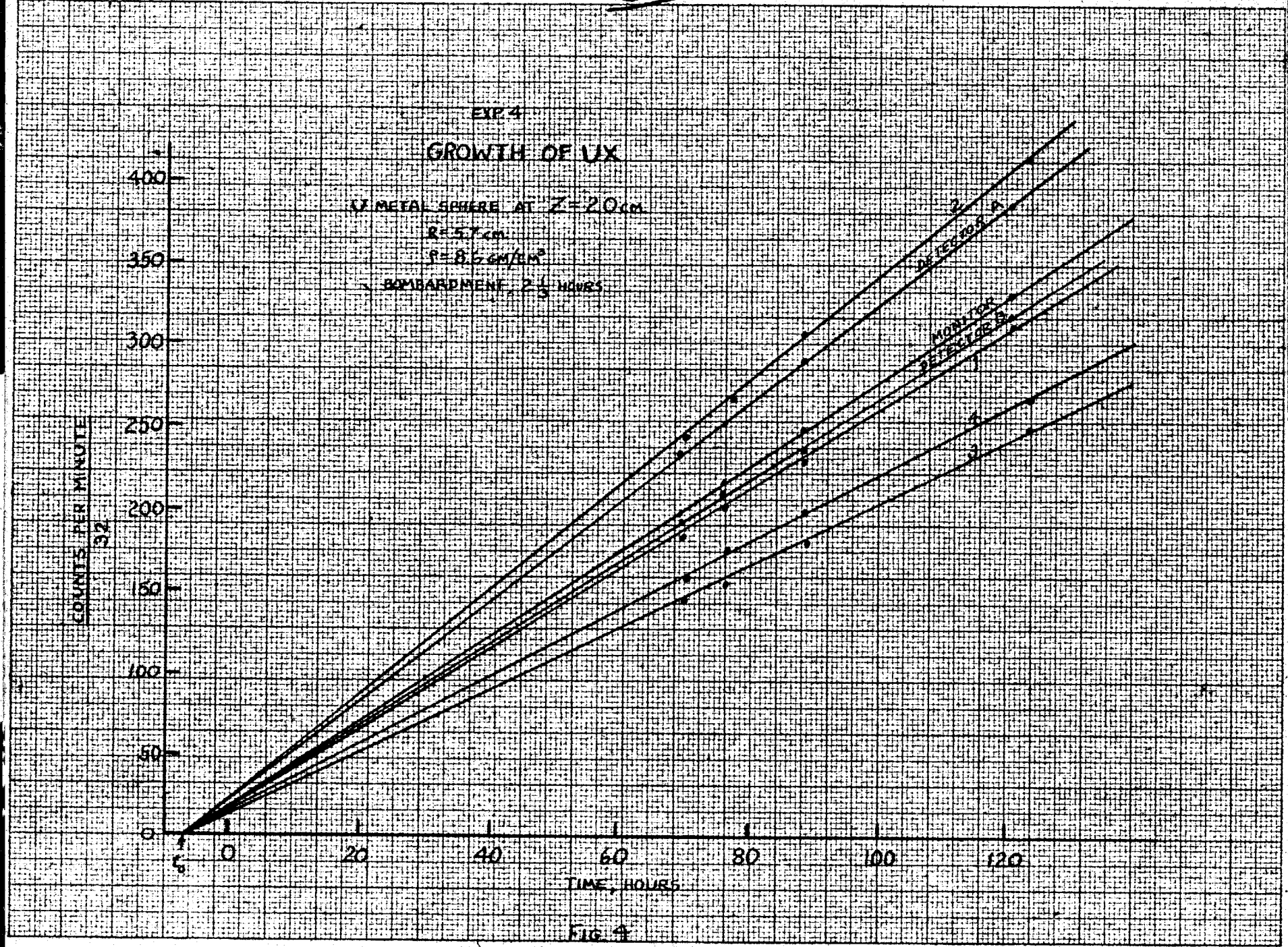
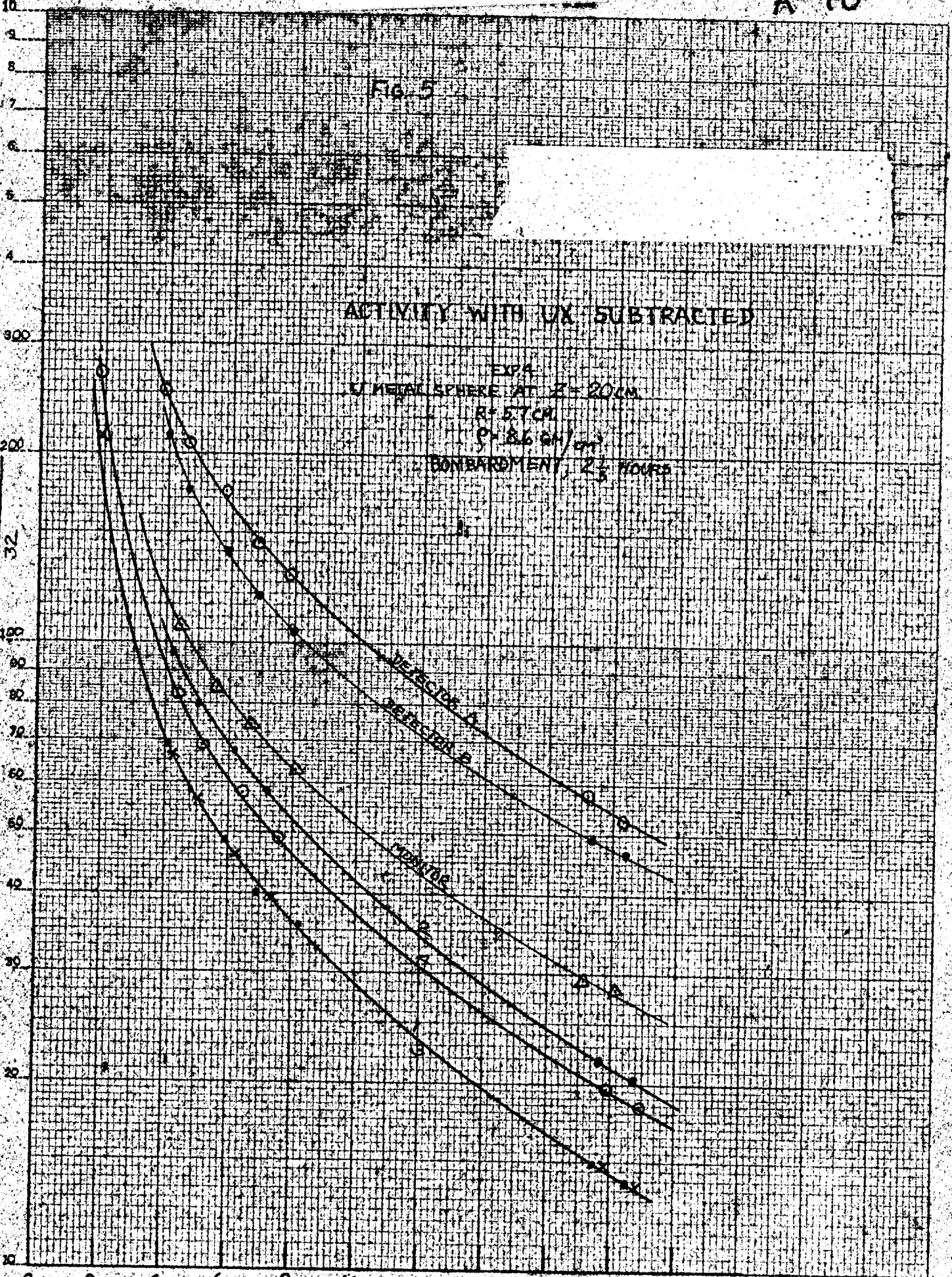


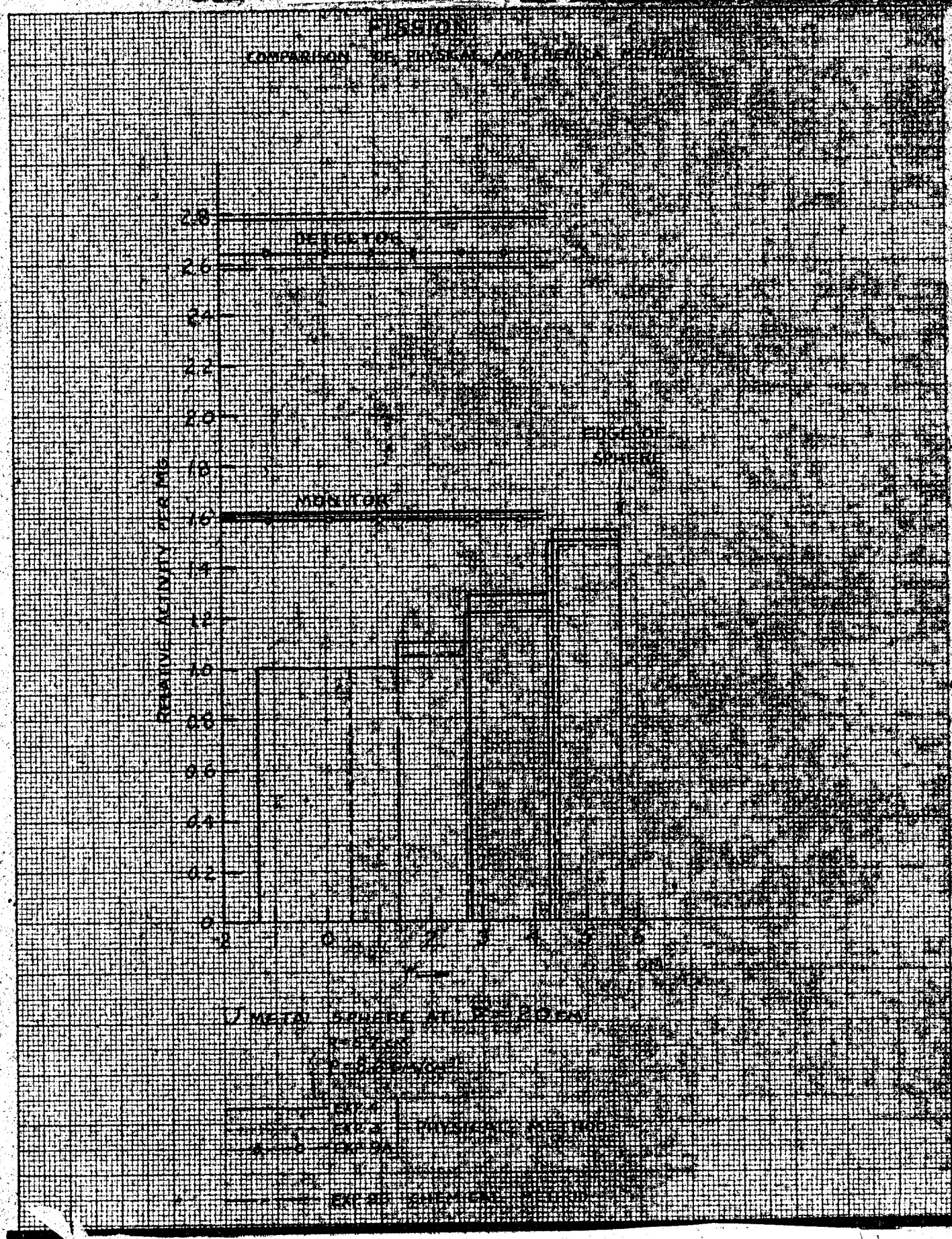
FIG 4

A-140 7

Fig. 5



838-34



KEUFFEL & ESSER CO., N. Y. NO. 358-14
Millimeters, 10th lines heavy.
MADE IN U. S. A.

Exp. 9A consisted in measuring by the Physical Method, the activity of an extra detector and monitor exposed simultaneously with samples of Exp. 9B. & 38-35

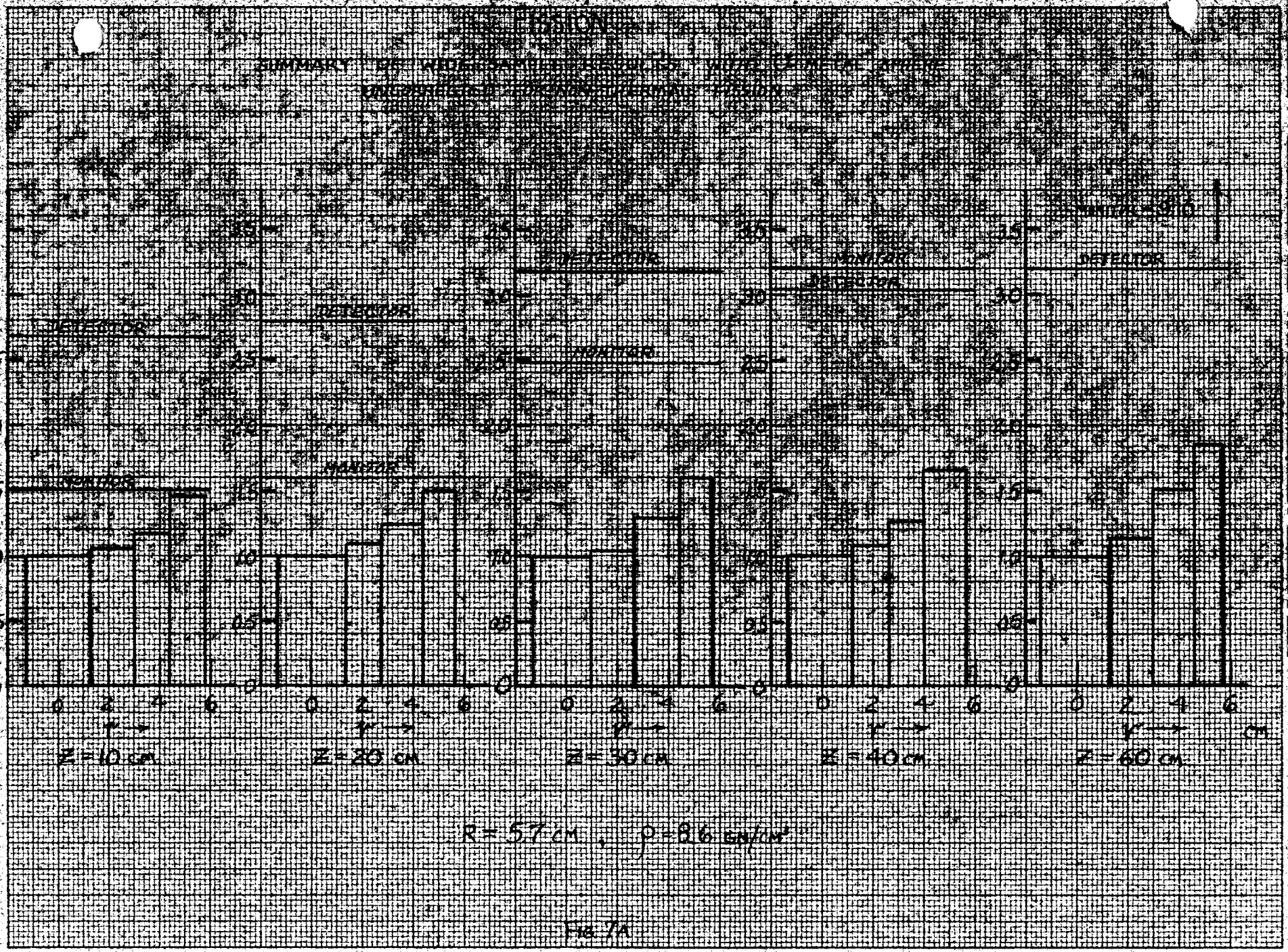


FIG. 7A

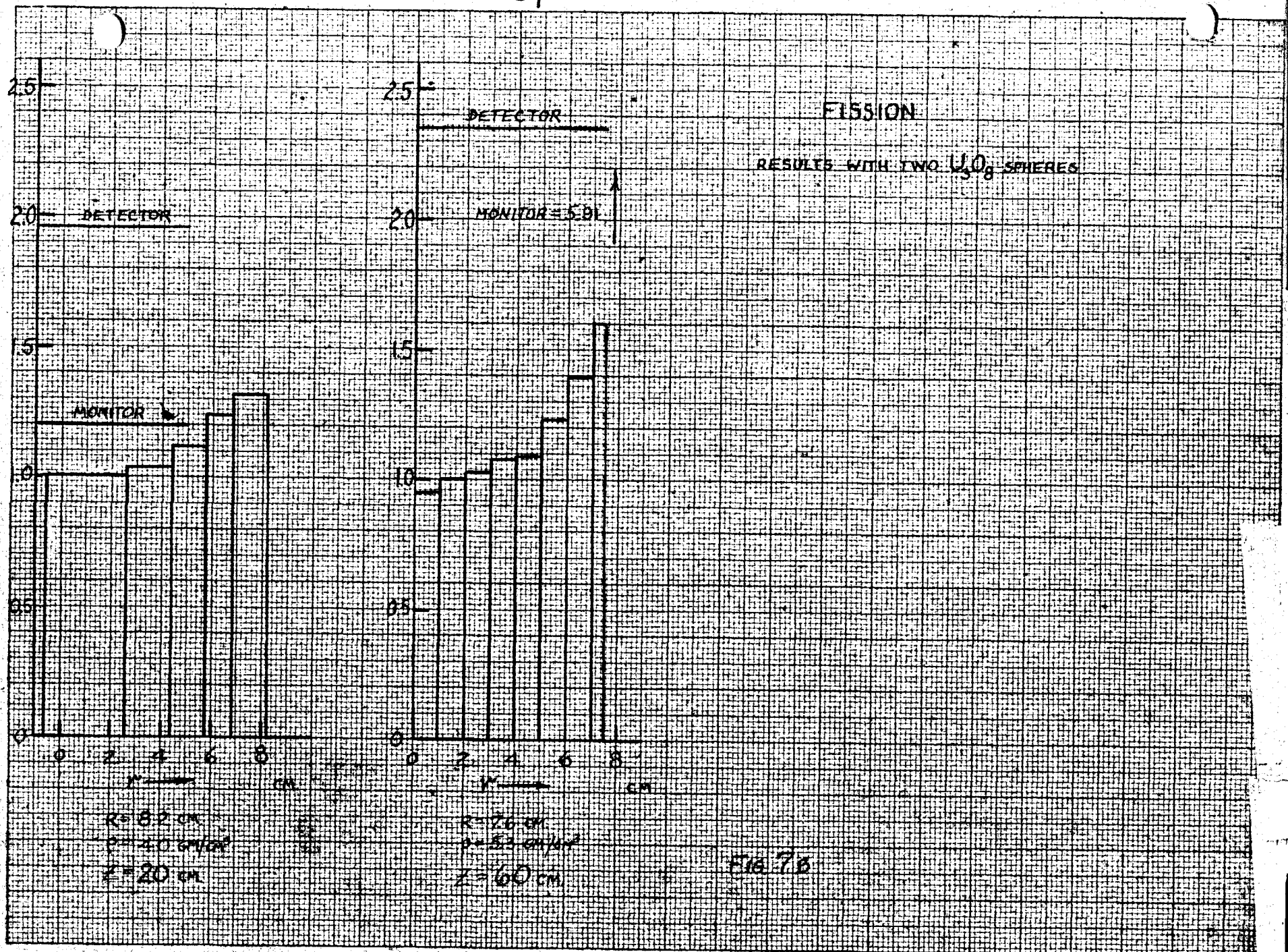

FISSION
RESULTS WITH TWO U_3O_8 SPHERES
DETECTOR
MONITOR
MONITOR = 5.01
2.5
2.0
1.5
1.0
.5
0
0 2 4 6 8
cm
R = 8.2 cm
ρ = 4.0 gm/cm²
Σ = 20 cm
R = 7.6 cm
ρ = 5.3 gm/cm²
Σ = 60 cm

FIG. 7b

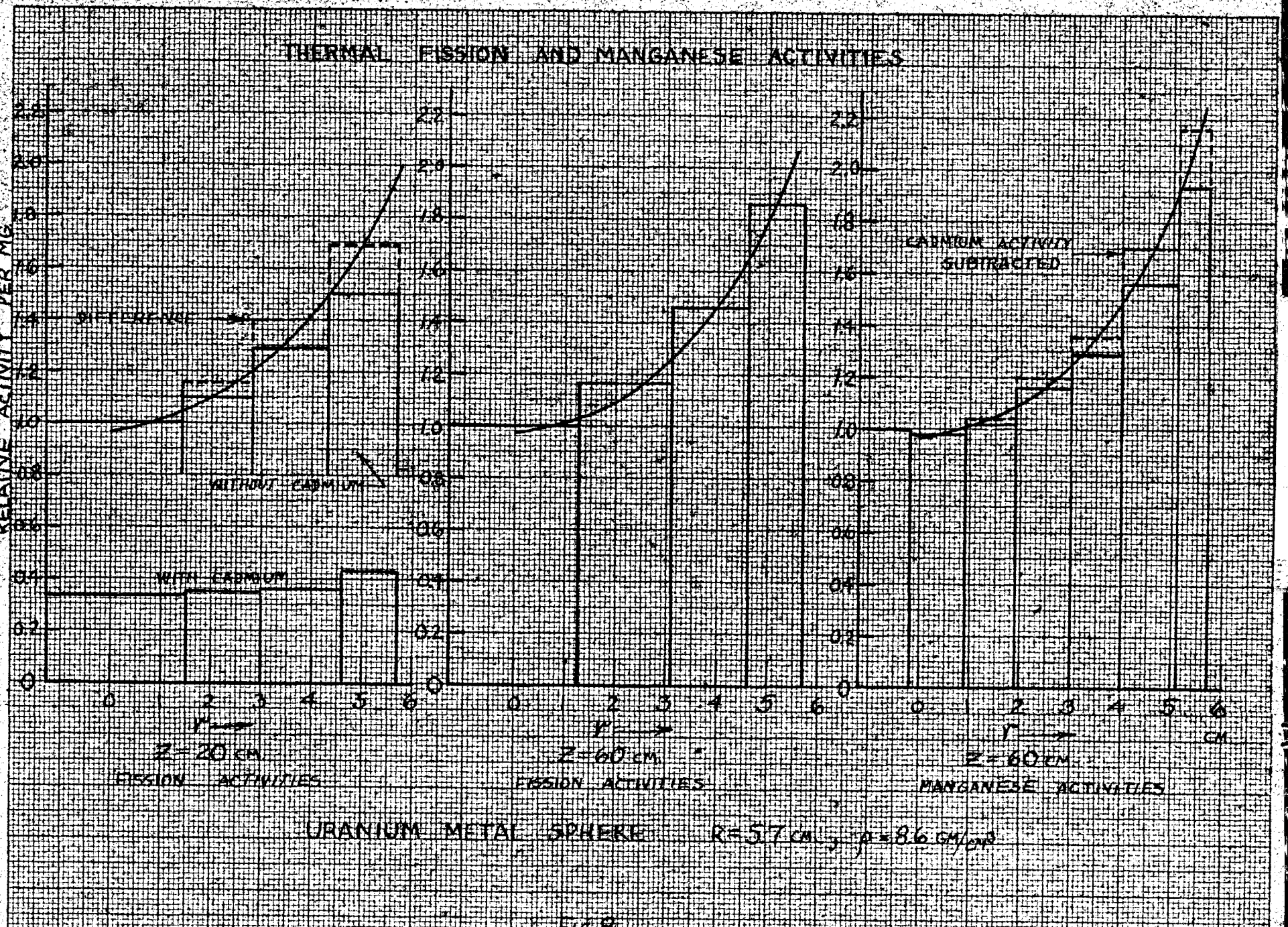


Fig 8

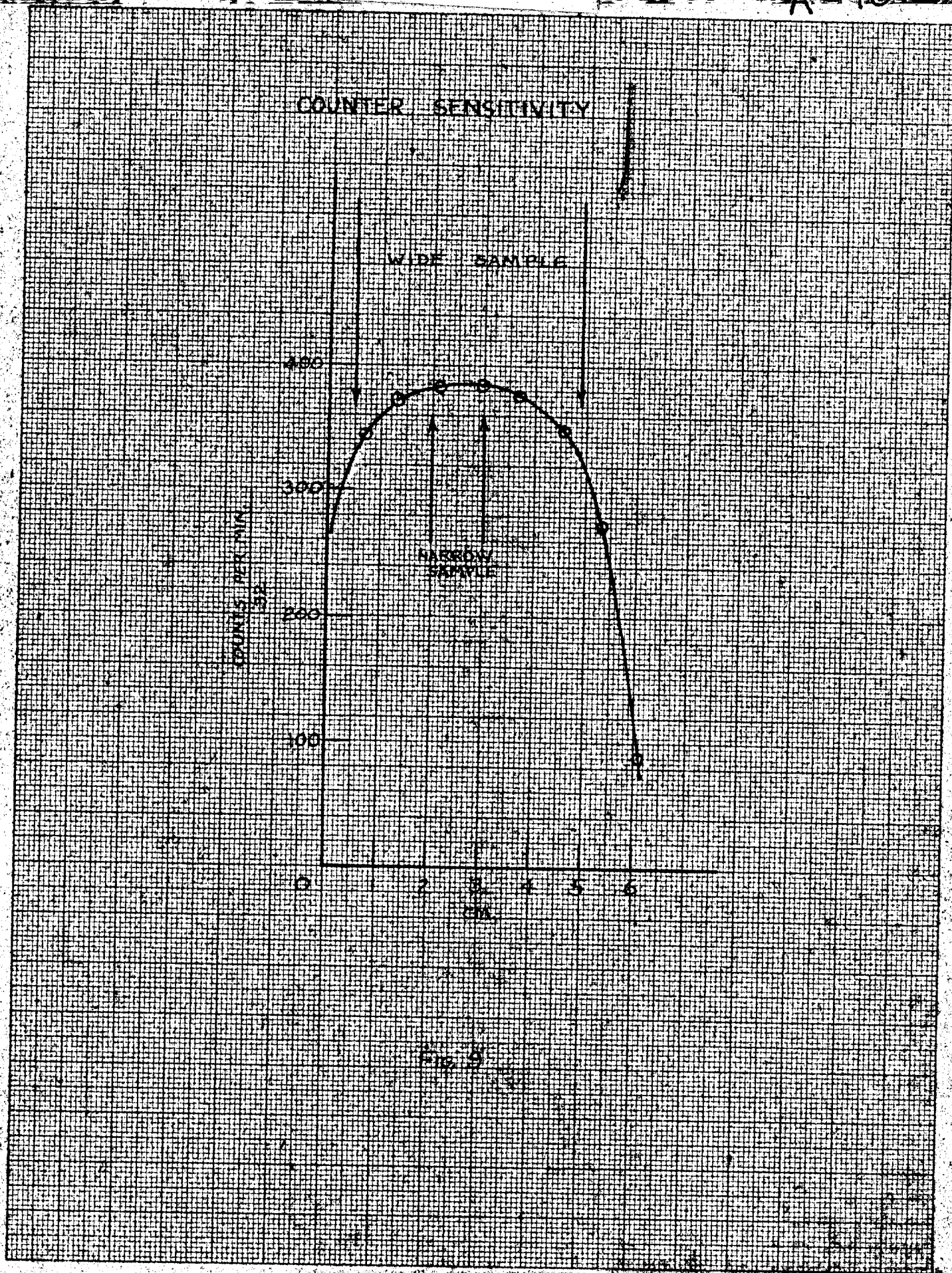


Fig. 9

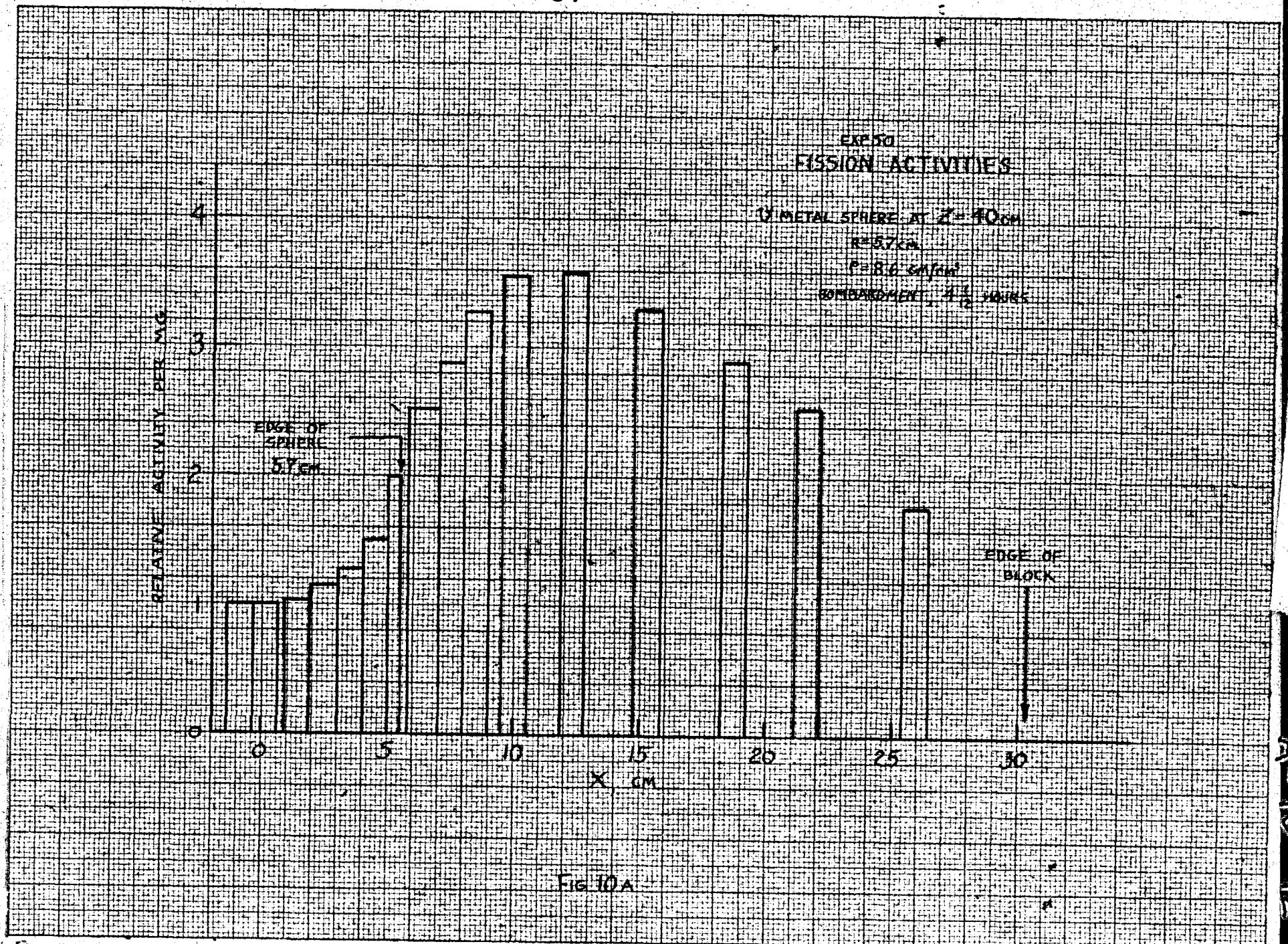


FIG 10A

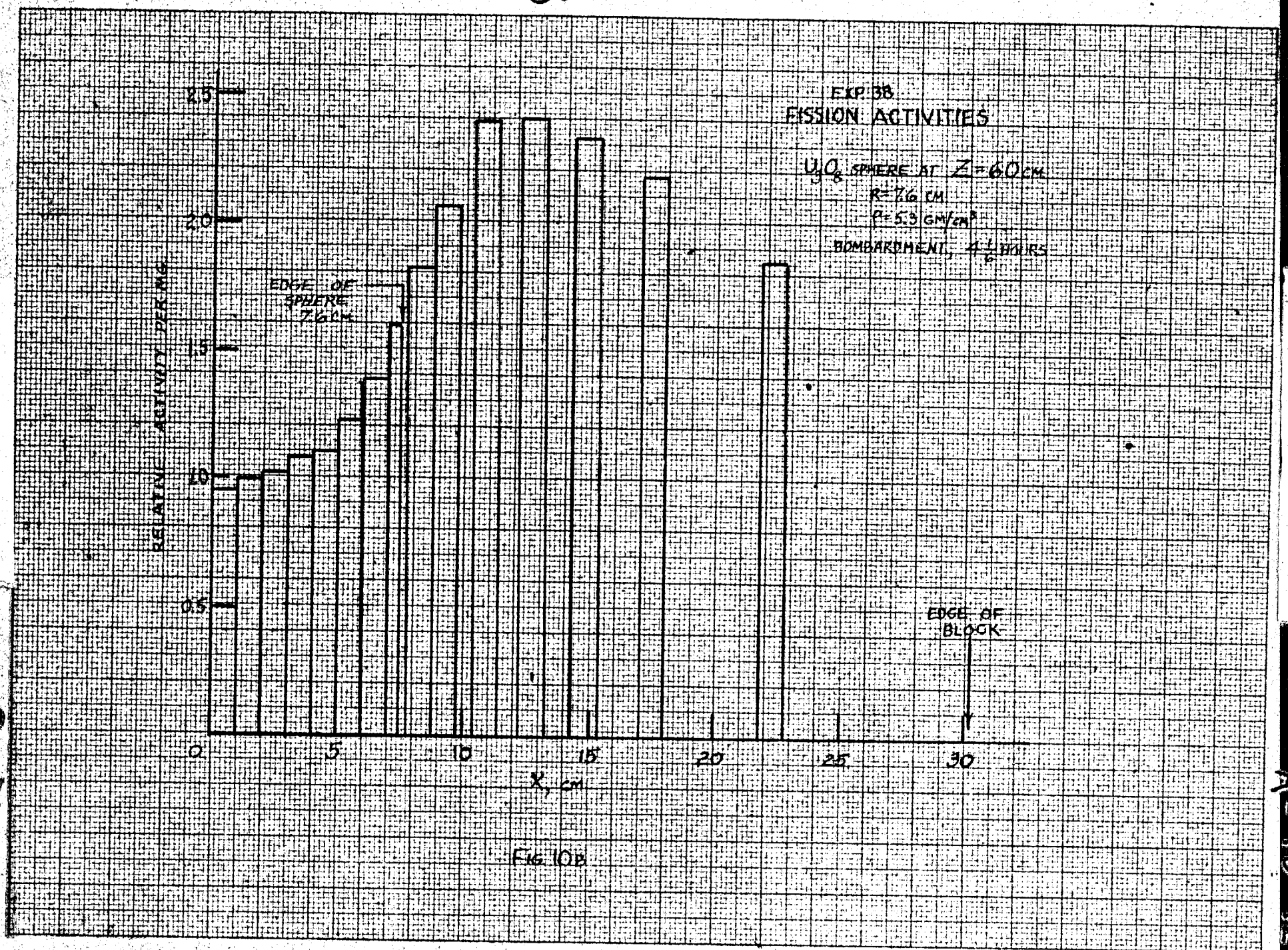
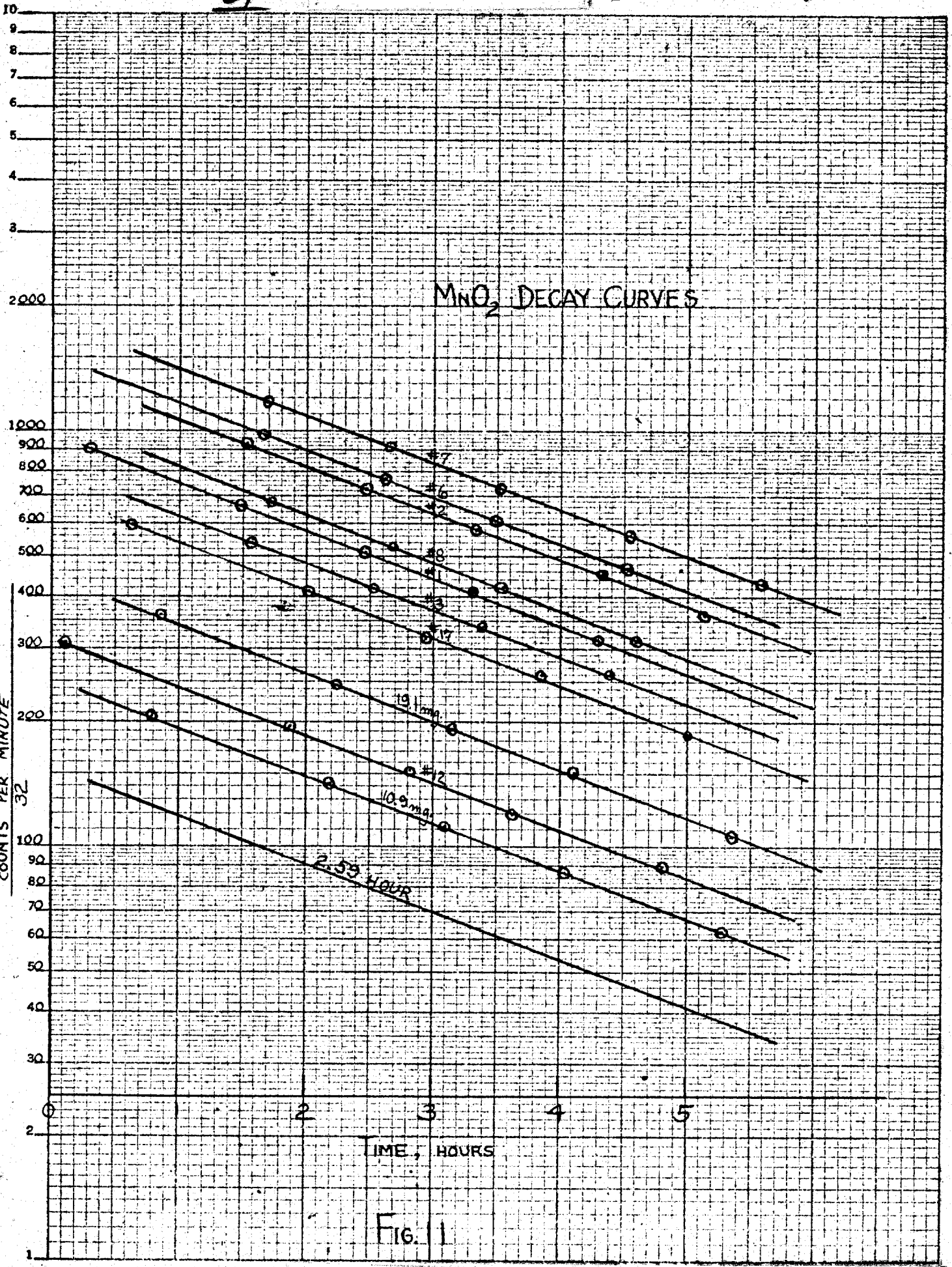


FIG. 10B



KEUFFEL & ESSER CO., N. Y. NO. 359-71
Semi-Logarithmic, 3 Cycles X 10 to the Inch.
MADE IN U. S. A.

FIG. 11

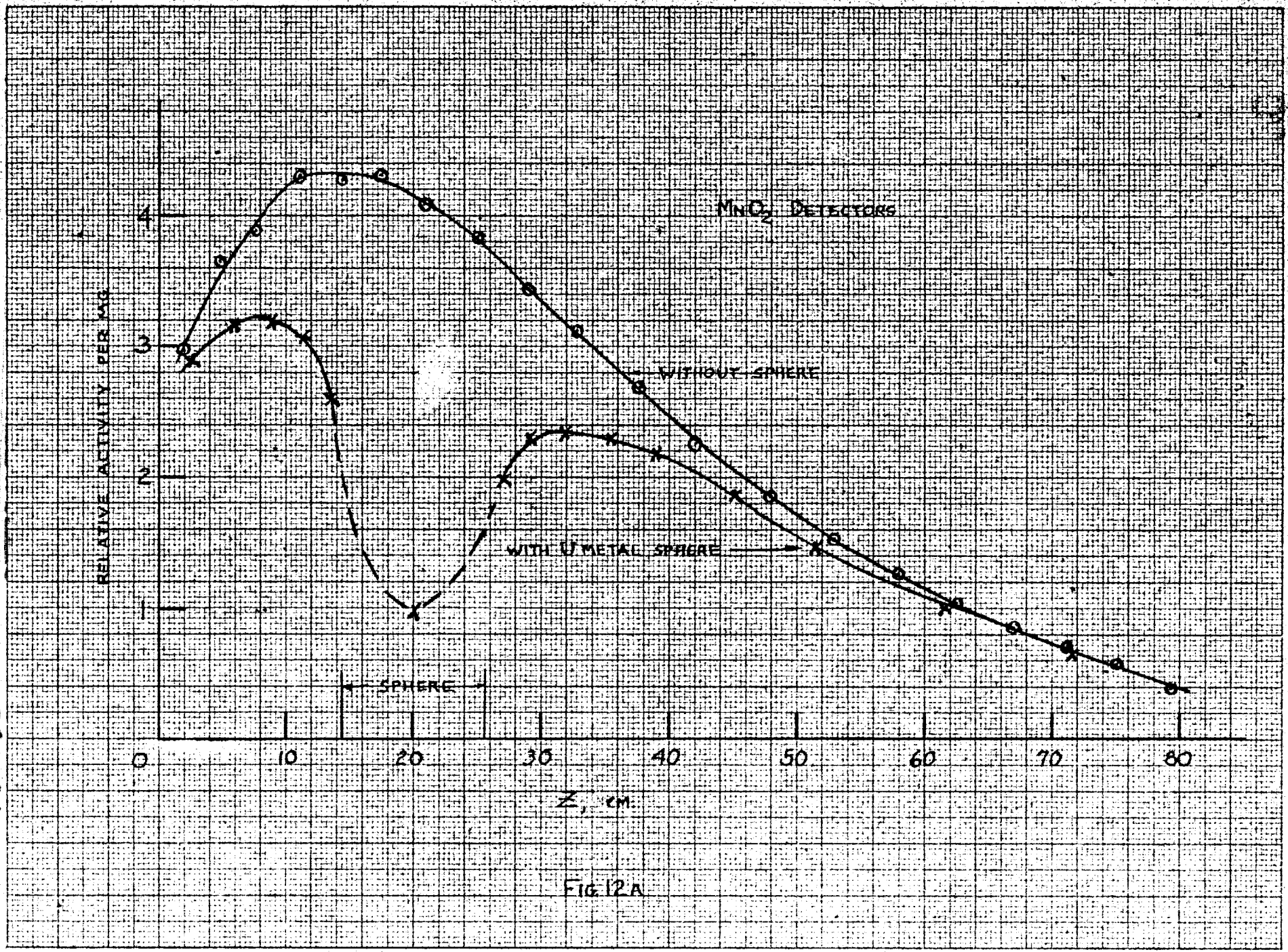


FIG 12A

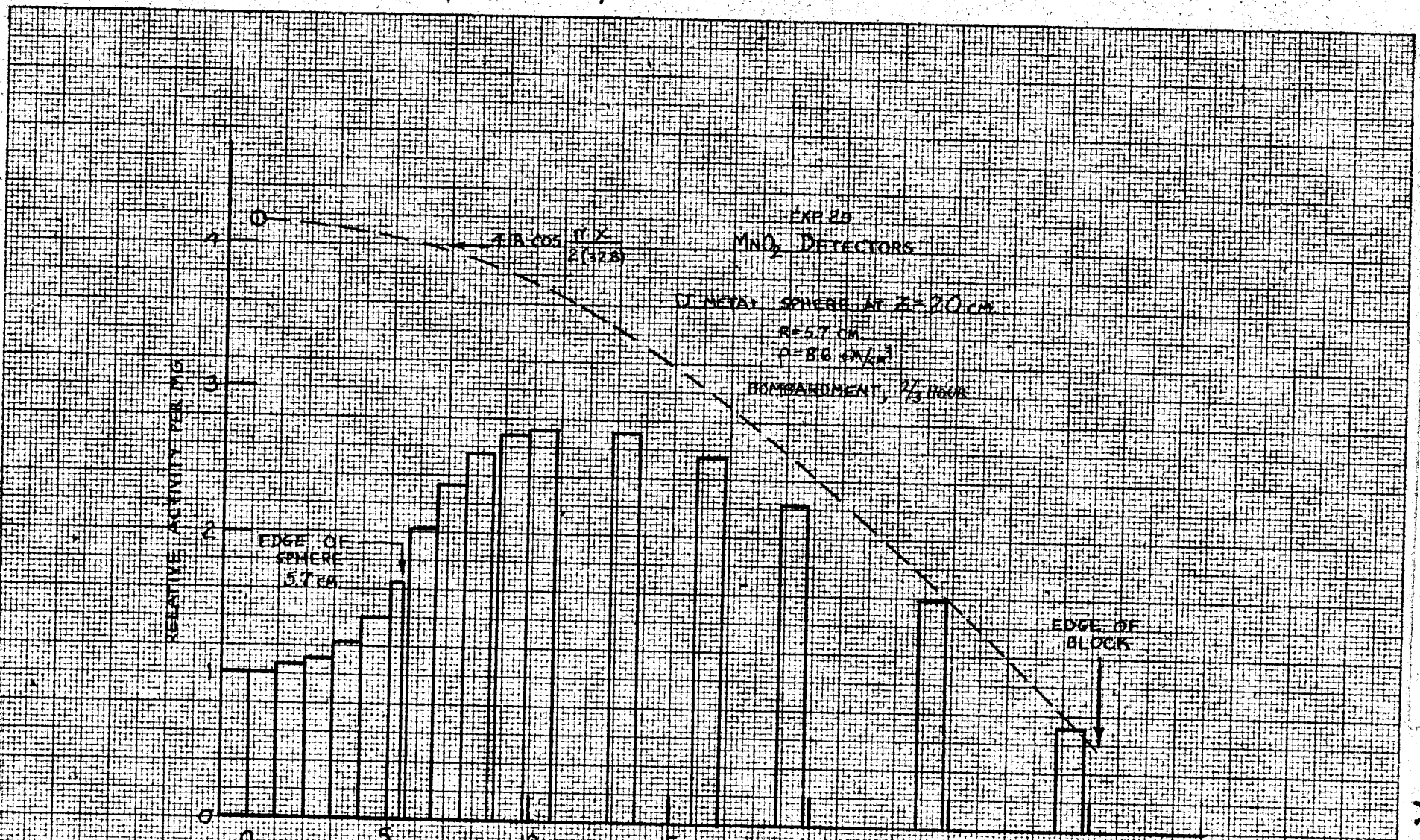


FIG. 2B

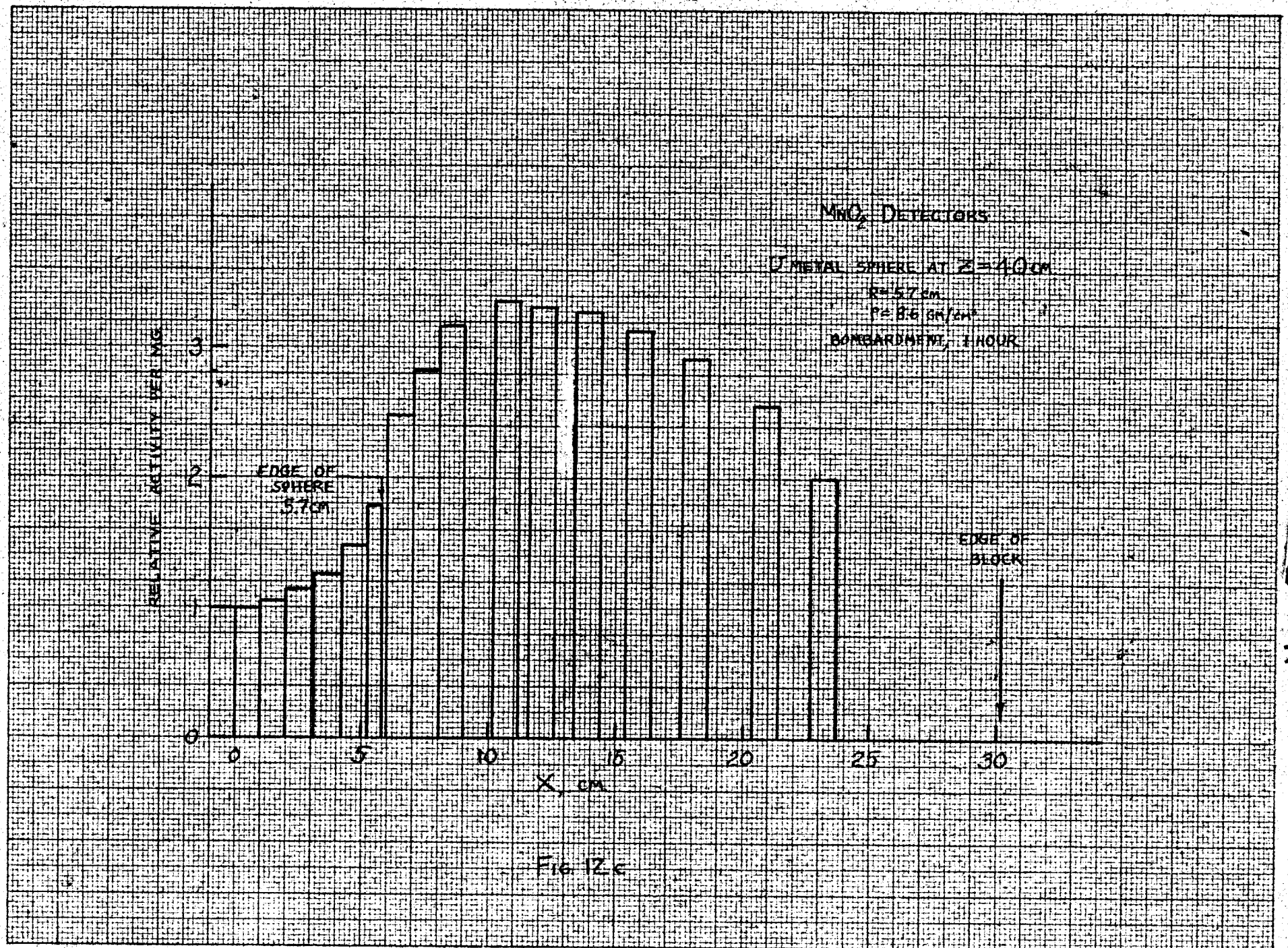


FIG. 12-C

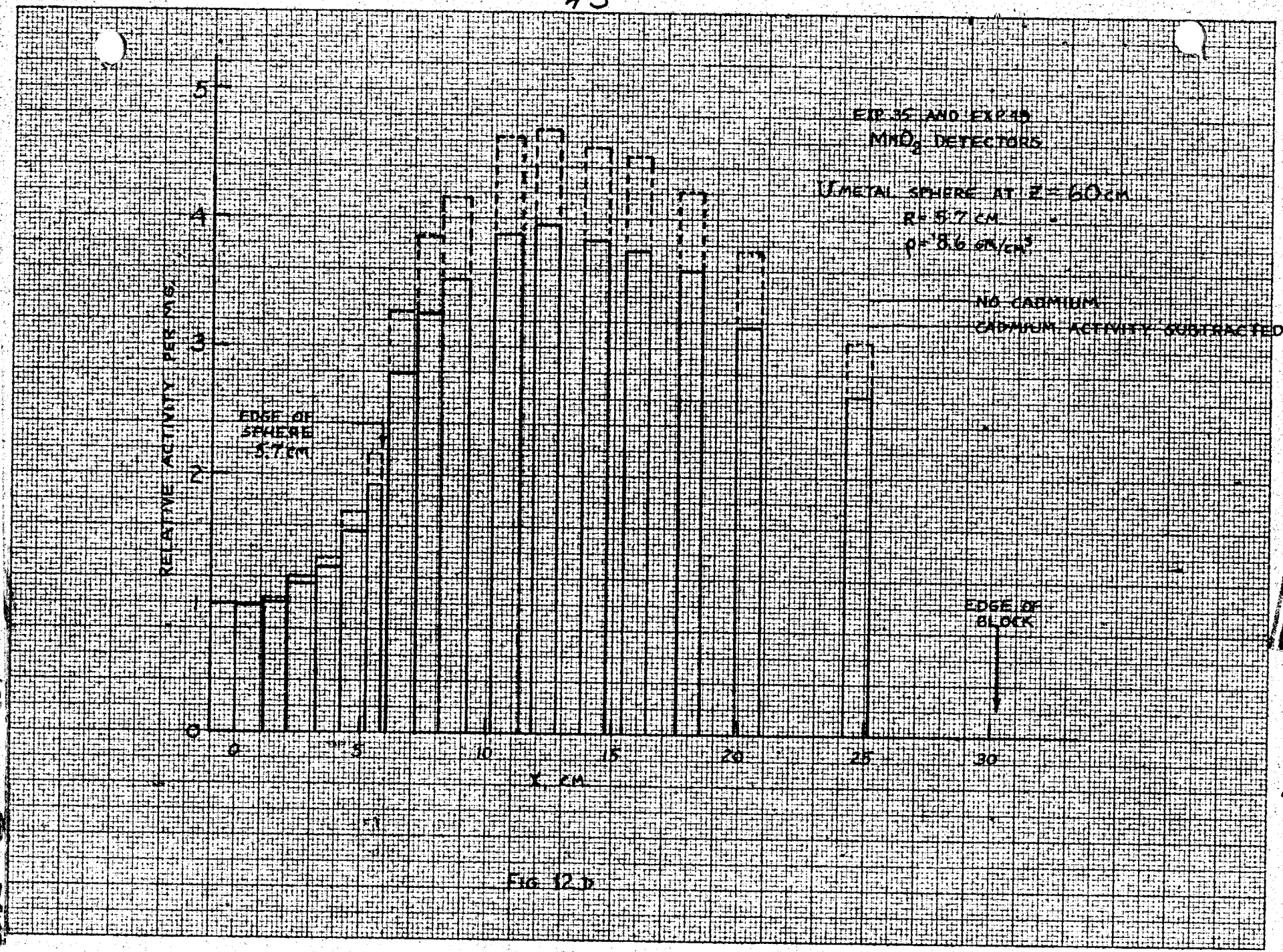


FIG 12 b

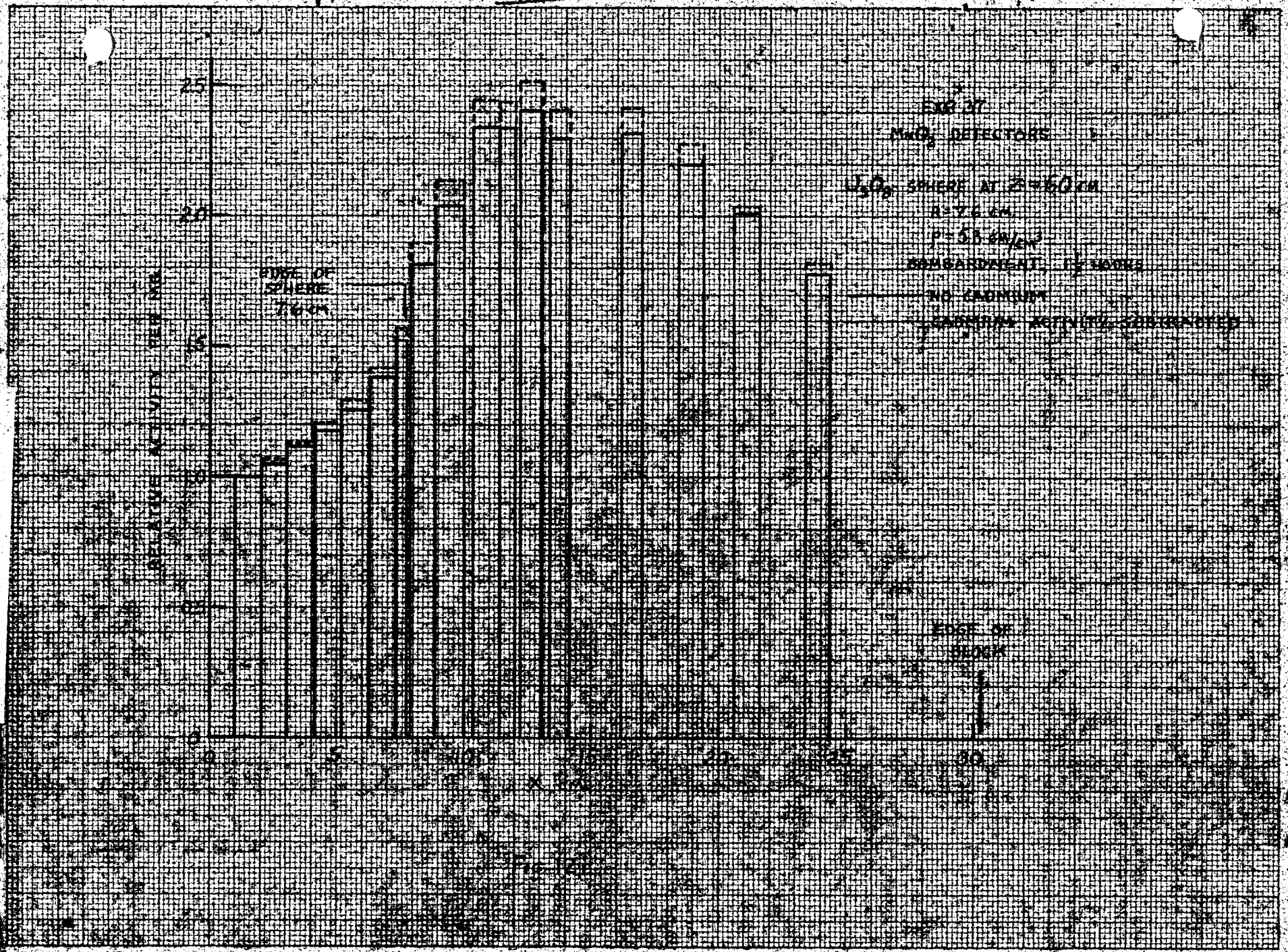

EXP 37
MnO_2 DETECTORS
U_3O_8 SPHERE AT Z=60 cm
R=7.6 cm
ρ=5.5 gm/cm³
BOMBARDMENT, 1½ HOURS
NO CADMIUM
CADMIUM ACTIVITY SUBTRACTED
EDGE OF SPHERE 7.6 cm
EDGE OF BLOCK
RELATIVE ACTIVITY per gm
2.5
2.0
1.5
1.0
0.5
0
5
10
15
20
25
30
X cm

Fig. 24