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DETERMINATION OF THE NUMBER OF FISSIONS IN TWO IRRADIATED SAMPLES OF U^{235}

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DETERMINATION OF THE NUMBER OF FISSIONS IN TWO IRRADIATED SAMPLES OF U^{235}

During the second half of 1944 and the first months of 1945 several samples of U^{235} and Pu^{239} were irradiated in the Hanford and Clinton piles. These samples have been studied with regard to the various products of the intense neutron exposure, particularly the capture products U^{236} and Pu^{240}. Values of the exposure (nvt), i.e. the time integral of the flux (nv were given by the operators of the piles but there was reason to doubt the accuracy of these values. Therefore it was decided to determine the number of fissions which had occured in two of the uranium samples. This gives the exposure in terms of the fission cross section of U^{235}. The procedure was as follows.

A sample of uranium containing 200 mg of the 235 isotope was exposed to thermal neutrons in the graphite column of the "water boiler" at Los Alamos; the total exposure was about 1.6×10^{13} neutrons/cm^2 in about 5 hours of irradiation. This sample will be referred to as O(1). The exposure was monitored by a foil of normal uranium, referred to as EN31, containing 0.69 ug of U^{235}. The amounts of fissionable material in O(1) and EN(31) were compared directly by preparing deposits of known aliquots of O(1) and comparing their fission rates with that due to EN(31) in a double chamber. From the total number of counts in EN(31) during the monitored run, and this mass ratio, we obtain directly the total number of fissions in O(1). Simultaneously with the monitored exposure another sample of enriched uranium referred to as O(2) was irradiated in the center of the waterboiler. It received about 100 times as much exposure as O(1). Neither the exposure nor the mass of O(2) was accurately known. The use of this sample will be discussed below.

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One sample from Clinton, referred to as X, and one from Hanford, referred to as C(1), were investigated. From known aliquots of each of the samples X, C(1), O(1), O(2) barium was extracted. The gamma rays from the barium, with the lanthanum in equilibrium with it were counted by a G-M counter, using liquid cells for the samples. From these counting rates, the aliquots, the chemical yields, the proper correction for decay and the known number of fissions in O(1) the number of fissions in the other samples can be calculated. Similarly, cesium was extracted from other aliquots and the gamma rays were counted. In this case no correction for decay was needed. The activity of the cesium extracted from O(1) was too small and the determination had to be based on O(2) which in turn was determined by the other methods. Decay corrections did not enter the comparison of O(1) and O(2) since these samples were irradiated at the same time. Finally aliquots of all samples were counted without any chemical separation. The decay of the total gamma activity was followed on aliquot of O(2) in order to determine the proper corrections. Another aliquot was used to determine the half life of Ba^{140} with great accuracy. The result was 12.73 ± 0.03 days. In determining the decay corrections for X and C(1) the records of the power output of the piles were used. The relative day-to-day values of these records are probably quite reliable.

Having thus determined the number of fissions in the two samples X and C(1) and knowing the amounts of U^{235} contained in them we can calculate the fraction of the atoms that actually underwent fission. Using a conventional fission cross section of 5.4×10^{-22} cm^2 we also calculated the neutron exposures. By comparing the fraction of atoms that underwent fission with the amount of U^{236} formed one gets the ratio $\alpha = \sigma_r/\sigma_f$. All of the numerical results are summarised in table I.

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FISSION COUNTING

The ionization chamber and assiciated amplifier used for the monitored irradiation were the same as those used in the measurement of the fission cross section of U^{235} by Deutsch and Linenberger (LA - 100). The sample O(1) was placed in a platinum envelope back to back with the foil EN(31). Both samples had approximately equal areas (8 cm^2). The neutron flux was assumed to be sufficiently isotropic to make corrections due to absorption in O(1) negligible. The bias curve showed a rise of only 0.5 percent over a factor of six in discriminator bias. A correction of one percent was therefore applied to the observed number of counts. With this correction it was found that 1.45 x 10^7 fissions took place in EN(31) during the irradiation.

Four aliquots containing between one and five micrograms of U^{235} were taken from O(1) and evaporated to dryness on platinum foils, taking care to spread the deposit over approximately the same area as EN(31). The samples were compared with EN(31) in a double ionization chamber placed in the graphite column of the water boiler. Possible asymmetries in the chamber or the neutron flux were eliminated from the result by interchanging the position of the samples in the chamber. It was found that O(1) contains (3.05 ± 0.04) x 10^5 times as much fissile material as EN(31). Thus 4.42 x 10^{11} fissions took place in O(1) during the monitored irradiation.

The amount of U^{235} in EN(31) - a deposit of oxide of normal uranium - was determined both by weighing the deposit and by alpha count as 0.69 ± 0.02 microgram. The amount contained in O(1) was given by the chemistry division from whom it was obtained as 200 ± 4 milligram. Thus the ratio of the number of fissions in the two samples should be (2.90 ± 0.1) x 10^5, in fair agreement with the value found by the fission count. The value adopted for this ratio was a weighted

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average of the two, namely $(3.02 \pm 0.04) \times 10^5$.

The amount of U^{235} in sample X was determined by the same method. The fission count indicated that it contained $(3.63 \pm 0.04) \times 10^5$ times as much as EN(31). An independent determination of this amount was made as follows. Sample X was one of four samples irradiated simultaneously in the Clinton pile. The total amount of U^{235} in the four samples was given as 972 ± 20 mg by R. Dodson who prepared the samples. The gamma ray activity of the four samples was intercompared with an air filled ionization chamber with a vacuum tube electrometer, using various thicknesses of lead obsorber. This should give the fraction of the total number of fissions which occured in our sample X. A slight correction was then applied for the difference in neutron flux at the locations of the several samples, as indicated by the Clinton group. It was found that sample X contained 0.254 ± 0.001 of the total amount of U^{235} irradiated, or 247 ± 5 milligram. This gives a ratio to EN(31) of $(3.6 \pm 0.1) \times 10^5$, in excellent agreement with the value obtained by fission count.

PREPARATION OF SAMPLES

Samples O(1), O(2) and X consisted of uranium oxide in platinum envelopes. The envelopes were unfolded and the U_3O_8 dissolved in hot, 3N HNO_3. The platinum, with the fission products driven into it by recoil, was dissolved in aqua regia. The solution of uranium and platinum were combined before being treated further in the case of O(1) and O(2) but were treated separately in the case of sample X. The solutions were made up to 100 cc and appropriate aliquots taken volumetrically. All vessels were tested for activity when solutions were transferred and where possible, the volumetric aliquots were checked by their gamma ray activity. It was found to be very important to keep the solutions

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sufficiently acid to keep some of the activity (probably the Cb - Zr chain) from precipitating on the walls of the vessels. The platinum solution of sample X contained about 20 percent of the total activity but the fraction of the total barium activity and of the total cesium activity found in the platinum solution were only 12 and 15 percent respectively. The discrepancy may be due to the different ranges of the various products or it may be connected with the diffusion of gaseous members of the chains. It was checked by repeating the entire procedure for barium, described below, twice both for the uranium and the platinum solutions. Only moderate radiation protection was required for the procedures since the strongest sample - X - showed a gamma ray intensity equivalent to only 100 mg of radium, both radiations filtered through 1/4" of lead. Sample W(L) consisted of uranium metal pressed into an aluminum cylinder. Mr. Charles Rice of the chemistry division dissolved this sample, including a thin layer of aluminum facing the uranium. We used a sample containing 8 percent of this solution, given to us by Mr. Rice. No separate determination of the uranium by fission count was made on this sample. The entire sample W(1) contained 160.8 mg of U^{235}, according to Mr. Rice.

CHEMICAL PROCEDURES

A. Barium

To appropriate aliquots of the various samples was added a known amount of barium chloride (about 100 mg) and about 10 mg of strontium as chloride. The barium chloride was precipitated with 35 ml of concentrated HCl. The precipitate was centrifuged out, dissolved in about 4 ml of strontium chloride solution containing about 10 mg of strontium, and repricipitated with conc. HCl. This procedure was repeated five times. The final barium chloride precipitate was

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dissolved in 20 ml of water. About 10 mg of lanthanum was added and then precipitated with NH_4OH. The supernatant containing the barium was transferred to another vessel, acidified, lanthanum added and precipitated again. This procedure was repeated five times. To the final supernatant was added about 40 mg of lanthanum and enough 12N HCl to make the solution about 1N in hydrogen ions. It was then diluted to about 80 ml. Three volumetric flasks (25 ml each) were filled from this solution. The content of one of these flasks was immediately analyzed for barium. Aliquots were taken from the other two flasks for counting samples. Whenever the amount of barium in a counting sample was great enough, it too was determined analytically at the end of the experiment. Barium was determined by first precipitating barium chromate, reducing the dichromate ion with iodide and then titrating the free iodine with thiosulphate. The yields in the purification procedure varied between 75 and 85 percent. The barium was apparently very pure Ba^{140} as shown by the pure exponential decay of the material extracted from O(1) and the much older sample X. Also lanthanum extracted from both barium samples decayed as pure La^{140} and represented the same fraction (88 percent) of the activity of the Ba - La equilibrium mixture.

B. Cesium

About 100 mg of cesium was added to appropriate aliquots of the samples. In those cases where there was platinum present in the solution, i.e., O(2) and the platinum fraction of X, cesium chloroplatinate precipitated immediately. This precipitate was dissolved in hydroxylammine and put through a number of group separations. Silver carrier was added and precipitated as chloride, antimony as sulphide, barium as carbonate and lanthanum as hydroxide. Each step was repeated until the precipitate showed no further activity. The cesium which had

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remained in the supernatant was then precipitated as perchlorate. In the case of the uranium fraction of sample X and of sample W(1) the cesium carrier remained in the original aliquot solution and the above purification procedures were carried out with this solution. The cesium perchlorate precipitates were dried on sintered glass gooch crucibles at 135°C to constant weight. After the final weighing they were dissolved and gamma rays were counted in the manner described below. Then the perchlorate precipitation was repeated, weighing and counting the precipitates until the specific activity was constant. The radioactive purity of the cesium was tested in one case by precipitating it as chloroplatinate, dissolving the precipitate and reprecipitating as perchlorate. The specific activity remained unchanged. The samples showed no observable decay over periods of the order of a month. Except that sample O(2) showed an initial decay indicating a feeble activity of half-life 10 to 30 days, which could not be separated from cesium by our procedure. No mention of short lived fission cesium was found in the literature but apparently the activity observed by us was also found by the Chicago group and established to be cesium (A. Turkevitch, private communication).

The chemical yields of our procedure were in the neighbourhood of 50 percent.

GAMMA RAY COUNTING

The experimental arrangement used to measure the gamma ray activities of the samples is shown in Fig. 1. Two similar brass-wall Trost counters were used in these experiments. The counters were surrounded by 1/32" lead cylinders in order to exclude very soft gamma rays or hard beta rays since such radiations would be strongly absorbed in the samples themselves. This might have caused a

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dependence of counting rate on the amount of dissolved material. The samples were contained, as solutions, in glass cells as shown in Fig. 1. Care was taken to fill the cells to a standard height (almost full) and to mix the contents thoroughly before counting. The acidity of the solutions was kept high to prevent precipitation of active material on the wall. When the solutions were removed from the cells the latter never showed any appreciable activity. Most samples were counted on both counters and the results were perfectly consistent. The ratio of the efficiencies of the two counters was 1.066 and independent of energy. The efficiencies for counting a 1.2-Mev gamma ray in this arrangement were $(2.08 \pm 0.1) \times 10^{-3}$ and $(2.22 \pm 0.1) \times 10^{-3}$ respectively, as found with the use of a standard solution of Co^{60}, calibrated by coincidence counting methods. The absolute efficiency does not enter these experiments, however.

The counters were frequently checked with a radium "standard" consisting of about 2 microgram of radium and its decay products in solution in one of the glass counting vessels. The day-to-day variations of the counting rate due to this standard indicated an intrinsic random probable error of about one percent which could not be reduced by prolonged counting. The source of this error is obscure. Since most of the significant counts were obtained with a statistical error of less than one percent, the overall probable error of a single count was generally about 1.5 percent. This is consistent with the observed deviations of repeated counts, decay curves etc. All counts were repeated as many times as was practical and the final errors in the gamma ray activities of individual samples were always less than 1 percent. The counting loss correction of the counters and scale of 64 used was determined by observing the decay of a sample of Mn^{56} and measuring the deviation of the early points from the straight exponential decay. The result indicated a "dead-time" of about 3×10^{-4} seconds and the

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correction never exceeded three percent for any significant samples. The counting rates of most samples were initially 3000 to 4000 c.p.m. except in the case of cesium from O(2) which gave only about 100 c.p.m. The background count was usually between 30 and 40 c.p.m.

CORRECTION FOR DECAY

Because of the long duration of the irradiation of samples X and W(1) it was necessary to know the half life of Ba^{140} with considerable accuracy. The activity of two samples was followed on the gamma ray counters, the decay extending over factors of fourteen and forty respectively. The half life was determined by the method of least squares. The two samples indicated values of 12.73 days and 12.77 days respectively. We adopted the value 12.75 ± 0.03 days. This value was supported by the fact that two independent determinations of the barium activity of sample X, separated by weeks gave the same result when this value of the barium half life was used.

In correcting for the decay of the pile-irradiated samples, the daily power dissipation of the piles during the irradiation was taken as a measure of the neutron intensity. Taking as M_t the energy dissipated (in megawatt - days) on a day t days before our experiment, the correction for barium decay is

$$F_{Ba} = (\Sigma M_t 2^{t/12.75}) / \Sigma M_t$$

The summation extends over the duration of the irradiation. X plots of M for the two irradiations are shown in Figs. 2 and 3. The correction for decay in the case of samples O(1) and O(2) can be taken as extending simply from the (logarithmic mean time of the exposure in the water boiler, since the irradiation lasted only a few hours and was very nearly constant.

The decay of the untreated samples of fission products follows a much

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more complicated curve. It was observed on a sample of 0(2) for about 150 days. Denoting by f(t) the relative activity t days after a short irradiation, the correction for the pile-irradiated samples becomes

$$F_\gamma = (\Sigma M_t f(t)/\Sigma M_t$$

In Fig. 4 we show f(t) multiplied by t, in order to compress the scale.

RESULTS

The results of the entire experiment, together with the steps leading to them and the probable errors of each are summarized in the table of results. One observation which may be of some interest and which is not shown in the table are the actual counting rates per fission. We found for the long lived cesium 1.30×10^{-12} c.p.m. per fission. For the 12.75-day barium in equilibrium, we found $8.0 \times 10^{-9} e^{-\lambda t}$ c.p.m. per fission, where t is the time elapsed since the fission. The counting rate due to the combined gamma rays can be read directly from Fig. 4. These values might be used for approximate determinations of the number of fissions if counters similar to ours are used with 1/32" lead shielding and are calibrated with a standard source of Co^{60} to compare with the efficiency of 2.08×10^{-3} for a 1.2 Mev gamma ray to which the above values refer.

The fraction of U^{235} converted by radiative capture was determined by Williams and Yuster with a mass spectrograph and is reported in a companion report (LA - 510). It is shown in the starred items of our table of results for completeness. The value of 0.186 for the ratio of the capture and fission cross sections is in good agreement with the value obtained by Williams and Yuster in a comparison of the production of U^{236} and the depletion of U^{235} in one of the irradiated samples. In comparing this value with those previously obtained by comparing σ_f with $(\sigma_f + \sigma_r)$ $\left\{\begin{matrix}(LA - 90)\\(LA - 100)\end{matrix}\right.$ (CP 153) it must be remembered that our present results

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refer to pile neutrons in which a small amount of fast fission occurs. The value for thermal neutrons should therefore be somewhat higher, probably between 0.19 and 0.20. This is just about the average of the older determinations mentioned.

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TABLE OF RESULTS

No. of fissions in EN(31)		$(1.45 \pm 0.015) \times 10^{7}$
Ratio of amounts of U^{235} O(1) .EN(31)		$(3.05 \pm 0.04) \times 10^{5}$
No. of fissions in O(1)		$(4.42 \pm 0.08) \times 10^{12}$
Ratio of no. of fissions O(2): O(1)	Ba^{140} count	43.6 ± 1.2
	Total γ count	45.6 ± 1
	Average	45.0 ± 0.8
Ratio of no. of fissions X : O(2)	Cs count	$(4.53 \pm 0.2) \times 10^{3}$
Ratio of no. of fission X : O(1)	Ba^{140} count	$(2.09 \pm 0.1) \times 10^{5}$
	Total γ count	$(1.96 \pm 0.1) \times 10^{5}$
	Cs count	$(1.96 \pm 0.1) \times 10^{5}$
	Average	$(2.01 \pm 0.06) \times 10^{5}$
No. of fissions in X		$\boxed{(8.9 \pm 0.3) \times 10^{17}}$
Amount of U^{235} in X mg		243 ± 3
Fraction of U^{235} in X which underwent fission		$\boxed{(1.40 \pm 0.07) \times 10^{-3}}$
Fraction of U^{235} in X converted into U^{236} *		$(2.63 \pm 0.03) \times 10^{-4}$
Ratio of capture to fission cross sections $\alpha = \sigma_r/\sigma_f$		$(0.188 \pm 0.01$
Exposure of X, assuming $\sigma_f = 5.40 \times 10^{-22} cm^2$. Neutrons/$cm^2$		$(2.59 \pm 0.13) \times 10^{18}$
Ratio of no. of fissions W(1)/O(2)	Cs count	$(2.4 \pm 0.2) \times 10^{4}$
Ratio of no. of fissions W(1)/O(1)	Ba^{140} count	$(1.06 \pm 0.05) \times 10^{6}$
	Total γ count	$(1.13 \pm 0.07) \times 10^{6}$
	Cs count	$(1.07 \pm 0.1) \times 10^{6}$
	Average	$(1.08 \pm 0.04) \times 10^{6}$
No. of fissions in W(1)		$\boxed{(4.8 \pm 0.25) \times 10^{18}}$
Amount of U^{235} in W(1) mg		161 ± 7
Fraction of atoms in W(1) which underwent fission		$\boxed{(1.17 \pm 0.08) \times 10^{-2}}$
Fraction of U^{235} in W(1) converted into U^{236} *		$(2.16 \pm 0.04) \times 10^{-3}$
$\alpha = \sigma_r/\sigma_f$		0.184 ± 0.015
Exposure of W(1) neutrons/cm^2		$(2.15 \pm 0.15) \times 10^{19}$
α, average for X and W(1)		$\boxed{0.186 \pm 0.008}$

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* Communication by Dudley Williams

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SUMMARY

The gamma-ray activity of two samples irradiated in the Clinton and Hanford piles and of the barium and cesium extracted from them were compared with the activities extracted from a sample in which the number of fissions was determined by monitoring during the neutron exposure. The results obtained show good internal consistency and indicate that $(8.9 \pm 0.3) \times 10^{17}$ and $(4.8 \pm 0.25) \times 10^{18}$ fissions respectively had occured in the two samples. These results are compared with mass spectrometer data obtained by Williams and Yuster on the same samples to find the ratio of the capture and fission cross sections of U^{235}.

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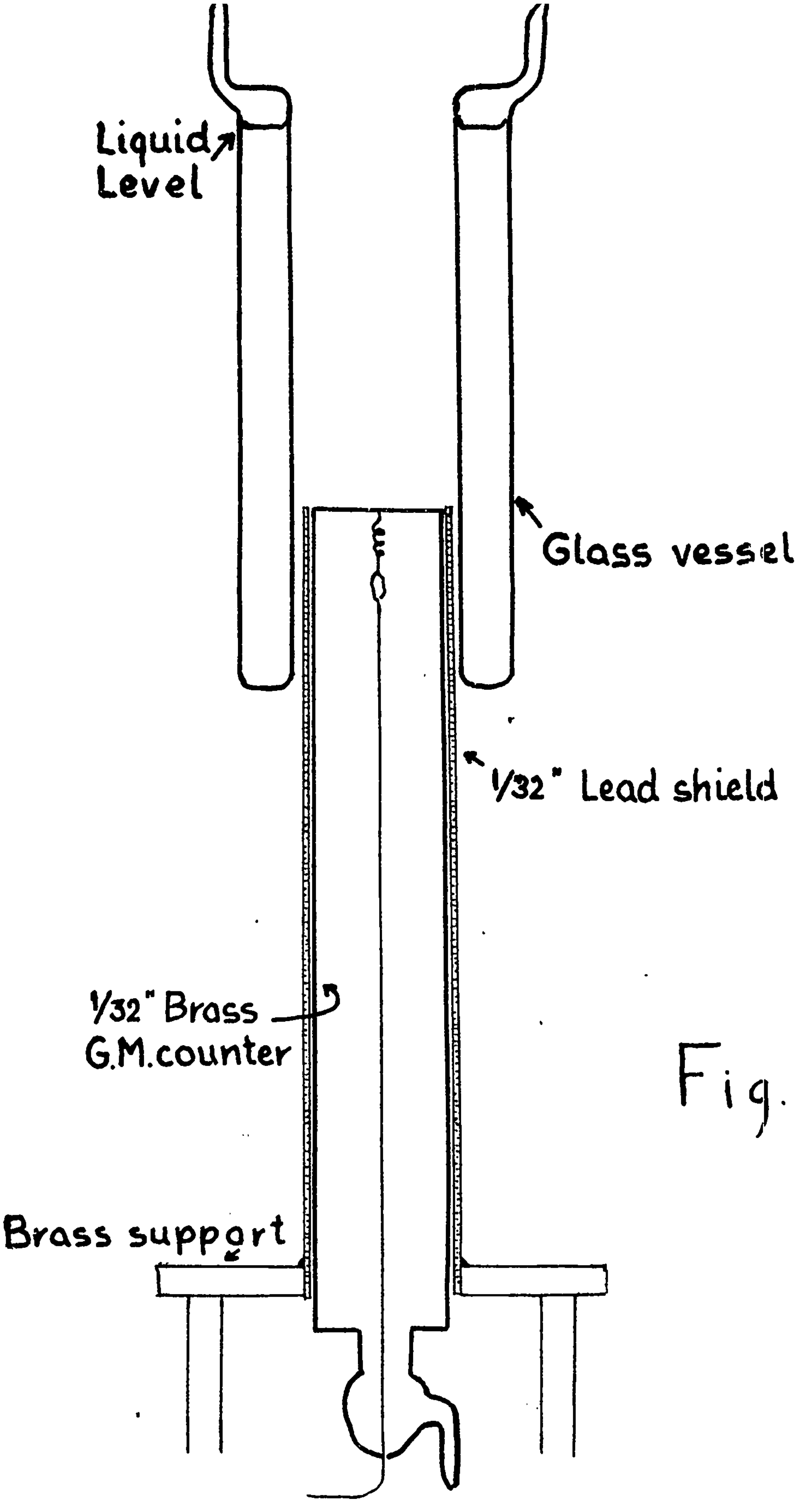
Liquid Level
Glass vessel
1/32" Lead shield
1/32" Brass G.M. counter
Brass support

Fig. 1

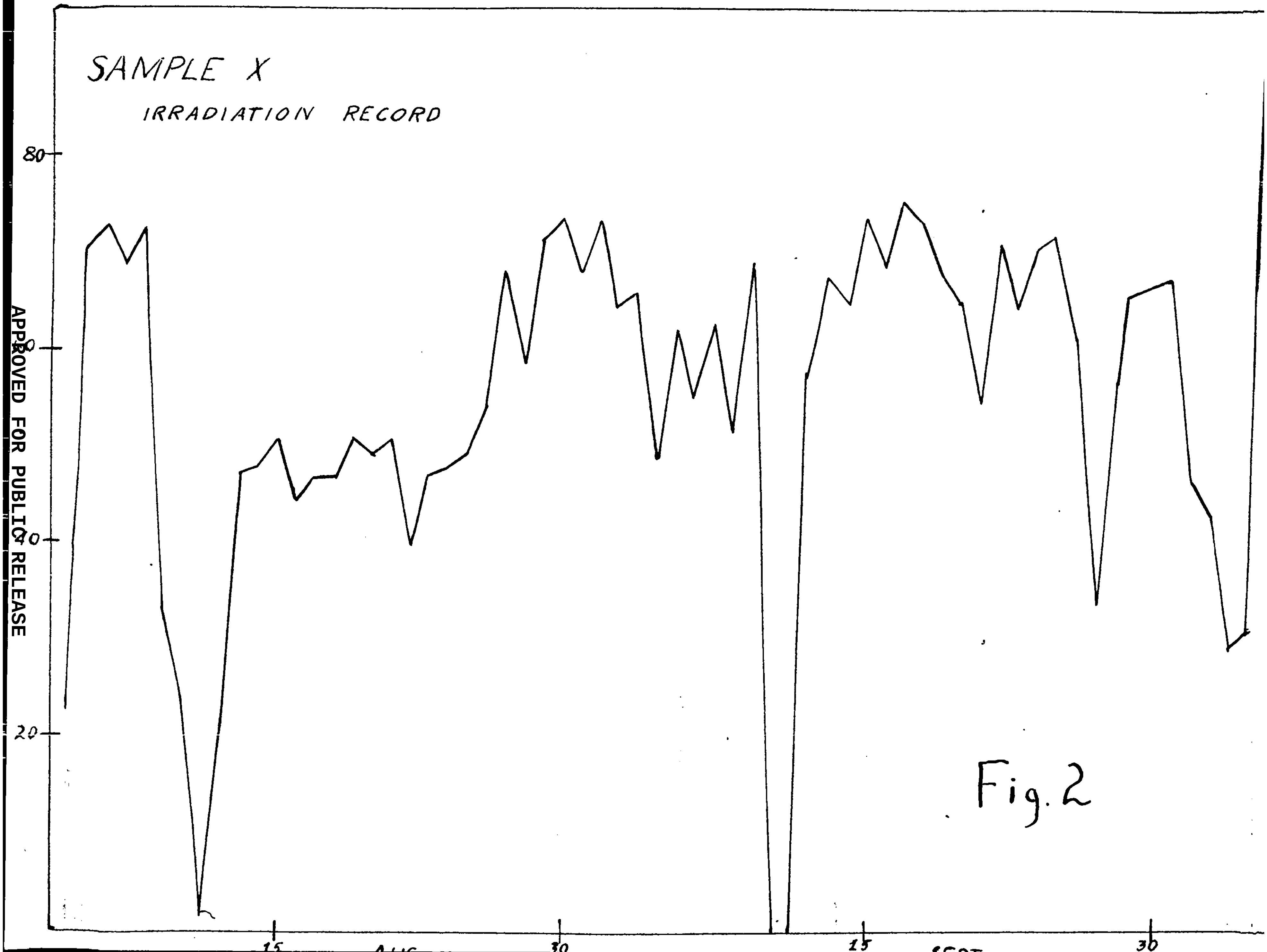

Fig. 2

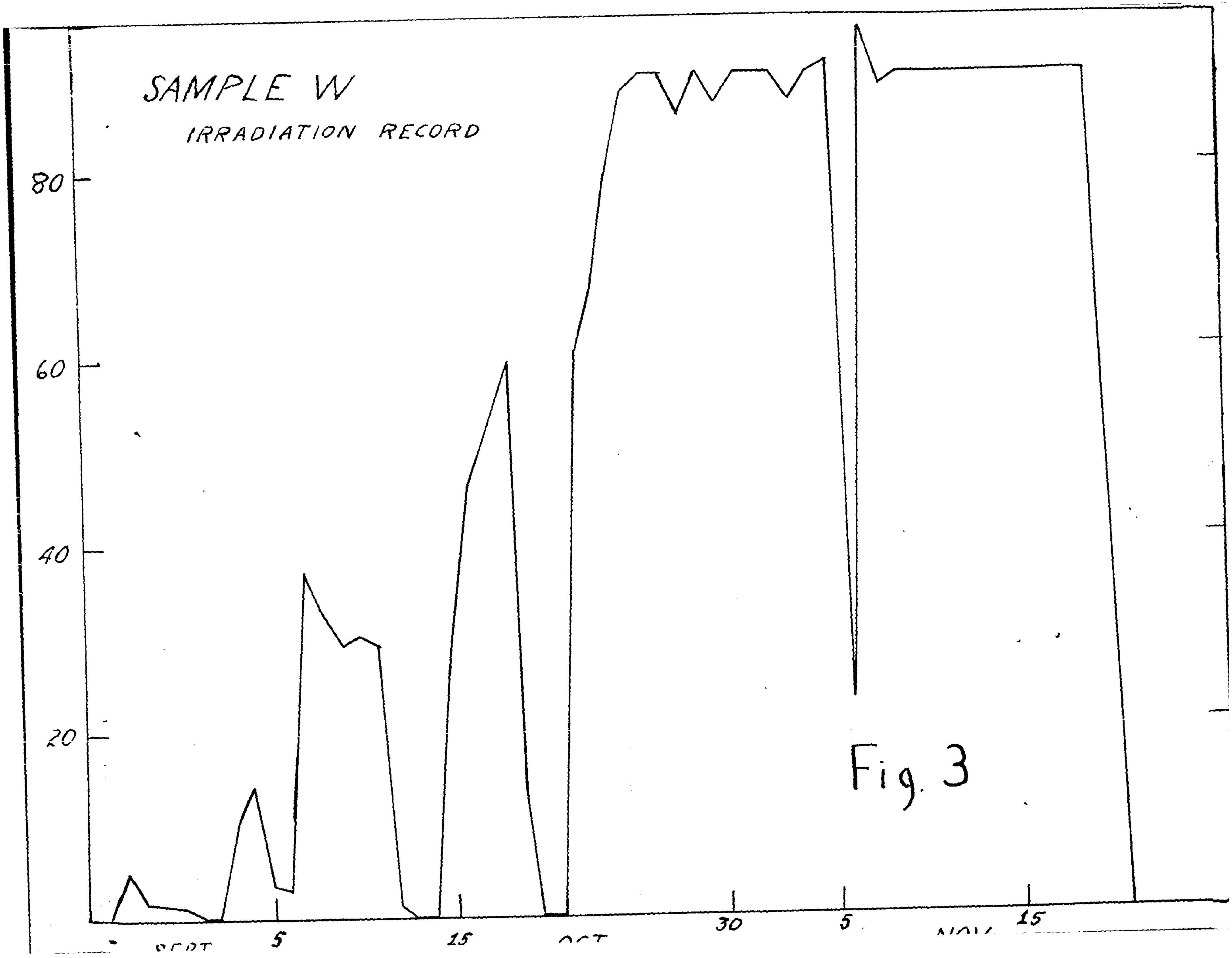

Fig. 3

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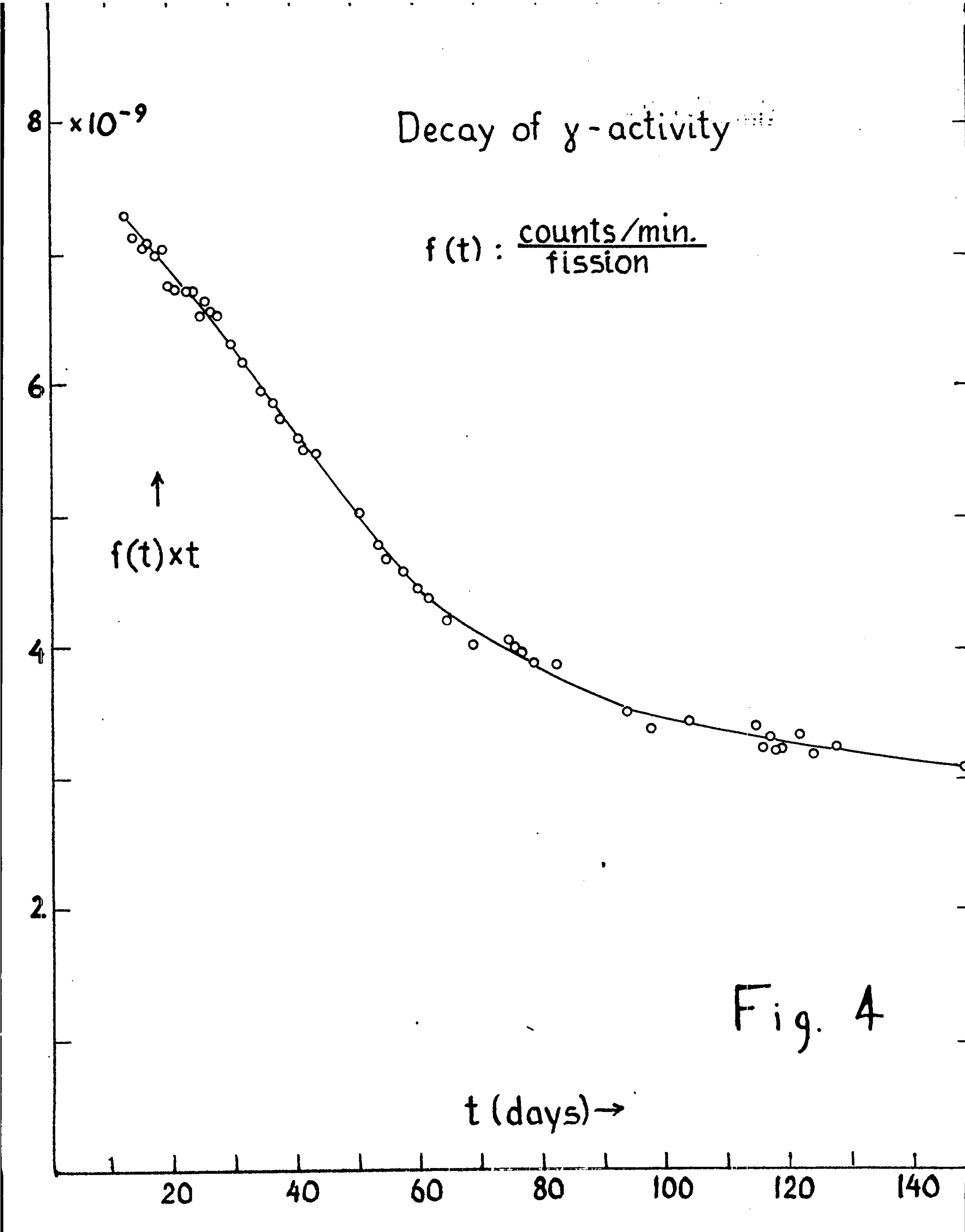
8 ×10⁻⁹
6
4
2
Decay of γ-activity
f(t) : counts/min. / fission
↑ f(t)xt
t (days)→
20
40
60
80
100
120
140

Fig. 4