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ANALYSES OF STACK AND FIELD SAMPLES

DURING IET OPERATIONS

JUNE 1956

~~C.W. Doe 11-21-61~~

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ANALYSIS BRANCH

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This document consists of 36 pages.
No. 1 of 1 copies, Series A.

Report Written:
July, 1956

ANALYSES OF STACK AND FIELD SAMPLES

DURING IET OPERATIONS

JUNE 1956

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During recent operations at the IET, samples of the stack gases were taken for analysis in an attempt to learn something of the nature of the material being discharged to the atmosphere--in particular, whether the iodine fraction was in the gaseous or particulate phase. During the latter phases of the IET operations, field samples were also collected.

This report will give the analytical results obtained on the samples and will also describe the sampling device used in obtaining the samples. No attempt will be made to draw final conclusions from the results reported herein since this investigation will be considered exploratory only.

Sampling Device. The sampling train consisted of two 4-inch millipore filters and two scrubber traps connected in series to a high volume motoair pump. (See Figure 1.) The millipore filters with their back-up papers were mounted in series in a modified 4-inch Staplex high-volume air-sampling head. The scrubber trap consisted of a 4-liter bottle fitted with a rubber stopper containing a stainless steel inlet and outlet tube. A coil of perforated tygon tubing sealed at one end was attached to the lower end of the inlet tube to disperse the incoming gas. The scrubber traps were connected to the sampling head and to each other with short lengths of tygon tubing. The last scrubber in the train was connected to the motoair pump with ordinary garden hose.

The scrubbing solution was prepared by dissolving 4 g of Na_2CO_3, 4 g of $Na_2S_2O_3.5H_2O$, and 25 mg of NaI per liter of distilled water. Approximately 1500 ml of this solution was used in the first scrubber and 1000 ml in the second scrubber.

The first millipore filter was used to collect particulates, and the second millipore was used to check the collection efficiency of the first filter. The first scrubber was used to trap gaseous iodine and the second scrubber was used to check the collection efficiency of the first scrubber for iodine.

The results of the experiments indicate the efficiency of the first millipore filter for particulates and the first scrubber for gaseous iodine is better than 95%.

Sampling. Stack samples were taken by General Electric personnel and sent to the Analysis Branch of the Health and Safety Division. The scrubbing solution was transferred from the traps to pyrex bottles before being sent to the laboratory. The millipore filters and back-up papers were carefully transferred from the collecting head to covered glass petri dishes for shipment to the laboratory. All samples were received at the laboratory in good condition.

Sampling data was furnished the laboratory on part of the samples. Following is a list of the samples, the sampling data furnished with each, and a brief description of the appearance of the first millipore filter.

Experiment A. Sample taken 1930-2045 hours, 5/22/56. Sampling rate approximately 1.8 cubic feet per minute. First filter black in appearance. Engines operating partly on chemical fuel.

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Experiment B. (Sample not active enough for analysis when received.)

C. Sample taken 1920-2120 hour, 5/31/56. Reactor operating at approximately 85% full power. First filter white in appearance.

D. Sample taken 6/7/56. First filter brown in appearance.

E. Sample taken 6/12/56. Sampling was of short duration and smoke bombs were used during sampling period. First filter was black in appearance.

Field samples for analysis were taken by the Site Survey Branch of the Health and Safety Division and sent to the laboratory. Following is a list of the samples and the sampling data furnished with each:

Experiment S-A. Sample taken 1920-2335 hours, 6/5/56 at highway 28.

S-B. Sample taken 1320-2020 hours, 6/6/56 at the IET stock fence.

S-C. Sample taken 1328-2040 hours, 6/6/56 at highway 28.

S-D. Sample taken 1300-2015 hours, 6/7/56 at IET stock fence.

S-E. Sample taken 2130-2200 hours, 6/12/56 at highway 28.

S-F. Sample taken 1330-2215 hours, 6/12/56 at the IET stock fence.

S-G. Sample taken 1415-2056 hours, 6/13/56 at the IET stock fence.

S-H. Sample taken 2120-2210 hours, 6/13/56 at highway 28.

S-I. Sample taken 1500, 6/14/56, to 0350, 6/15/56 at the IET stock fence.

Analysis of Samples. In order to obtain as much information as possible each sample was divided into several parts and each part was treated separately. A 20.5-mm disc was cut from the center of the first millipore filter for beta decay of the raw sample, and the remainder of the filter was divided in half. One-half of the filter was placed in a plastic tube and counted in the welltype gamma scintillation counter to determine the gamma activity of the raw sample. A gamma decay curve and a gamma spectrum were run on this half. The second half of the filter paper was oxidized with a H_2SO_4-CrO_3 mixture and the iodine fraction separated by solvent extraction, using accepted radiochemical techniques. The iodine fraction was divided into two portions—(1) 1 ml for beta counting, and (2) 4 ml for gamma counting. A beta decay curve, a gamma decay curve, and a gamma spectrum were obtained on the iodine fraction.

The second millipore filter was treated in the same manner except that no gamma spectra were obtained. Also, when the activity on the second millipore filter was low the entire filter was used for iodine extraction.

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The first scrubber solution was treated with H_2O_2 and evaporated to 100-ml volume. The solution was then divided into 3 portions--(1) 5 ml for gamma counting, (2) 1 ml for raw beta counting, and (3) 94 ml for iodine separation. A beta decay curve, a gamma decay curve, and a gamma spectrum were obtained on the raw solution and on the separated iodine fraction.

The second scrubber solution was treated in the same manner except that no gamma spectra were obtained.

The back-up papers behind the first and second millipore filters were oxidized and the iodine fraction separated. Usually the entire paper was used for this purpose.

The above scheme, with the exception of the back-up paper treatment, is shown in outline form in Figure 2. In general, the outlined scheme was followed but on some of the earlier samples certain of the steps were omitted. This will be evident in the table of results.

The samples with activity too high to allow accurate counting on the size fractions indicated in the outline were divided into smaller portions. However, all results were calculated back to total sample size and are reported in counts per minute for total sample.

For each type of activity, determinations were carried out at essentially the same geometry so that the results on the iodine activities may be compared directly in counts per minute. A well-type scintillation counter using a thallium-activated sodium iodide crystal and an end-window G. M. tube were used for gamma and beta, respectively. Since the response of the thallium-activated sodium iodide crystal is energy dependent and the energy distribution of the raw sample on the first millipore filter differs from that of iodine, the raw gamma activity of the first millipore filter cannot be compared to that of the rest of the samples.

Results on Stack Samples. The distribution of gamma activity in the IET stack samples is given in Table I. In a given experiment all counts presented in the table were made at the same time $\pm$ 30 minutes and no correction for decay has been made. The date and hour given in the first column of each of the tables represents the date and hour that the count was made. The distribution of beta activity in the IET stack samples is given in Table II. The results presented in these two tables show that the collection efficiencies of the first millipore filter and the first scrubber solution are better than 95% for particulate and gaseous materials respectively.

Table III gives the distribution of iodine activity in the IET stack samples. Based upon the assumption that iodine found on the first millipore filter was particulate and that iodine found in the first scrubber solution was gaseous, the ratio given in the last column of Table III represents the ratio of gaseous to particulate iodine at the face of the filter. Note that the iodine found in the second millipore filter, both back-up papers, and the second scrubber solution are not included in this calculated ratio. Since the iodine data was incomplete on some of the above samples, this data was excluded in the calculated ratio on all experiments. Examination of Table I, experiment D, indicates that if the above iodine were included in the calculations it should probably be included as gaseous iodine, and only would change the ratio by approximately 10%. Note that in each sample following the first millipore filter the ratio of extracted iodine activity to raw activity is approximately

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the same as the ratio of extracted iodine activity to raw activity in the first scrubber solution, indicating that the material is probably very similar. It should be pointed out here that work in progress since this data was taken indicates the iodine recovery from the scrubber solution may have been slightly low due to incomplete extraction.

The ratio of iodine activity to total activity is given in Table IV for both beta and gamma activities. The gamma ratio is included here to show the excellent agreement with the beta ratio for these particular experiments. However, since the gamma count made with a thallium-activated sodium iodide crystal is energy dependent, the gamma ratio cannot be trusted to give an indication of the percentage of iodine activity in the total sample. In contrast the beta ratio does give a fair indication of the percentage of iodine activity in the sample.

Decay curves and gamma spectra on identical fractions of the four experiments showed no detectable differences. Therefore the curves obtained on the various fractions of experiment C have been chosen to represent all four experiments. Additional identification work was performed on portions of the #1 millipore filter from experiment D and the curves obtained in this work are also presented.

Figures 3, 4, 5a and 5b are gamma and beta decay curves and represent the raw particulate activity from the first millipore filter, the extracted iodine activity from the first millipore filter, the raw activity from the first scrubber solution, and the extracted iodine activity from the first scrubber solution, respectively. The samples for the gamma decay curves were kept in stoppered tubes so that the front end of the decay curves are influenced by the build-up of Xenon daughters. This probably accounts for the difference in the gamma and beta decay curves on the same material. The gamma decay curves of the iodine fractions have been resolved into the half-lives of I^{133} and I^{131}.

Gamma spectra of the raw millipore filter #1 taken at intervals after the sample was received are shown in Figures 6a, 6b, 6c, and 6d. Comparison of these four curves shows the decay of the I^{133} (half-life 21 hours) with time. The I^{132}(half life 2.4 hours) however, shows no decay over a period of three days. This suggests the presence of Tellurium ^{132}, the precursor of I^{132}, on the filter paper and this was later confirmed. The 0.33 Mev gamma peak, shown on these curves and which nearly covers up the I^{131}peak, was not identified.

Figure 7 shows the gamma spectrum of the chromic acid oxidation mixture of the millipore filter #1 of experiment D after the iodine fraction was extracted. Unfortunately, a similar curve on experiment C was not obtained. So although this curve is not directly comparable to the curves given in Figures 6a, 6b, 6c, and 6d, it is included here to indicate the gamma energies left after the iodine was removed from the millipore filter #1.

Figure 8 gives the gamma spectra of the iodine fraction extracted from the millipore filter #1. Three isotopes of iodine can be identified from this curve-- I^{131}, I^{132}, and I^{133}.

The gamma spectrum of the raw scrubber solution #1 is shown in Figure 9.

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Gamma spectra of the extracted iodine fraction from scrubber solution #1 taken at intervals after separation are given in Figures 10a, 10b, 10c, and 10d. The growth and decay of the Xenon daughters of iodine may be observed in these curves. The samples were in stoppered tubes so that the Xenon could not escape. The decay of the I^{133} isotope may also be observed in these curves. Note the absence of any indication of I^{132}. In the absence of the parent Te^{132}, the I^{132} originally trapped in the scrubber would be gone before the sample was received at the laboratory, since the half-life of I^{132} is only 2.4 hours.

Both the #1 millipore filter and the #1 scrubber solution were checked for the presence of tellurium. After the original iodine extraction the samples were set aside for approximately 20 hours to allow I^{132} to grow into equilibrium with Te^{132}. Then the iodine was again separated by extraction, and decay curves were obtained. The decay curves are given in Figure 14. From the curves it can readily be seen that there was tellurium on the first millipore filter but none in the first scrubber solution. The activity removed from the first scrubber solution during the second extraction is apparently some iodine left behind by the first extraction.

In order to establish the identity of the isotope giving rise to the 0.23 Mev gamma peak observed in the gamma spectrum of the raw #1 millipore filter, a portion of the #1 millipore filter from experiment D was wet ashed with nitric and sulfuric acids and a tellurium separation made on the residue. Figures 11a and 11b give the gamma spectra of the tellurium fraction at two different intervals after separation. The growth of I^{132} into equilibrium with the Te^{132} may be noted from these curves. Curve 11b was taken after equilibrium was reached. The difference in the amplitude of the iodine and tellurium peaks is due to the difference in the absorption coefficient of the NaI crystal for the different gamma energies. The energy dependence of the NaI crystal must be considered in attempting any quantitative interpretations from gamma spectra.

A gamma spectrum of the filtrate remaining after tellurium separation on millipore filter #1 is given in Figure 12. This curve indicates that one or more isotopes with gamma energies of approximately 0.14 and 0.32 Mev remain to be identified. An estimate of 50 to 80 hour half-life for the isotope with the 0.14 Mev gamma energy was made from the gamma spectra of the raw #1 millipore filter. This suggested that the 0.14 Mev gamma energy might be due to Tc^{99}, the daughter of 67-hour Mo^{99}. Accordingly another portion of the raw #1 millipore filter paper was wet-ashed, tellurium removed, and a molybdenum separation made on the residue. A beta absorption curve was run on the molybdenum fraction and is presented in Figure 15. Gamma absorption spectra of the filtrate from the molybdenum separation are given in Figures 13a and 13b. The half-life estimated from the change in amplitude of the 0.14 Mev peak is in the neighborhood of 6 hours, the half-life of Tc^{99}.

The 0.33 Mev peak shown on the curve was not identified because the sample was exhausted before identification could be completed.

Results on Field Samples:

The distribution of gamma activity in the field samples is given in Table V. The date and hour given in the first column of the table is the time at which the samples were counted.

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Table VI gives the distribution of iodine activity in the field samples. The ratio given in the last column of the table represents the ratio of gaseous to particulate iodine if the assumption is made that the iodine on the filter paper was particulate and that in the solution was gaseous at the face of the filter paper.

The ratio of iodine gamma activity to total gamma activity in the field samples is given in Table VII. This ratio is based only on the activity found on the first millipore filter and in the first scrubber solution.

No gamma spectra or decay curves were obtained on the field samples because of insufficient activity.

Summary of Results:

(1) The iodine activity in the IET stack appeared to be predominantly in the gaseous phase with the exception of experiment E in which smoke bombs were introduced into the stack during the sampling period. In fact, there appears to be some correlation between the iodine trapped as particulate on the first millipore filter and the amount of carbon deposited on the filter paper.

(2) The ratio of gaseous iodine to particulate iodine in the field samples is just reversed from that in the stack samples. This might be expected due to particulate fall-out and simultaneous diffusion of the gaseous cloud. Under certain conditions this could lead to essentially a sample of particulate fall-out while most of the gaseous cloud by-passed the sampling apparatus.

(3) Based strictly on the beta counting results, 30 to 50% of the activity in the IET stack samples during the operation was iodine. Although gamma counting results appear to verify the percentage figures for this set of experiments this seems to be fortuitous since the gamma counting technique used was energy dependent.

The only figures available for percentage of iodine activity in the field samples were based on gamma counting. These figures, given in Table VII, are questionable since the gamma counting was energy dependent.

TABLE I

DISTRIBUTION OF GAMMA ACTIVITY IN IET STACK SAMPLES

eriment	1st millipore paper		1st back-up paper		2nd millipore paper		2nd back-up paper		1st scrubber solution		2nd scrubber solution	
te and hour ounted	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m
xperiment A /24/56 .230		7,100 ± 89		1,130 ± 22		340 ± 18		340 ± 18		15,400 ± 128		385 ± 18
xperiment C /1/56 [redacted] 130	850,000 ± 1,350	77,000 ± 315		23,600 ± 155		13,500 ± 60		10,000 ± 48	1,220,000 ± 3,800	1,030,000 ± 1,670	40,000 ± 420	26,000 ± 129 [redacted]
xperiment D /12/56 1050	7,500,000 ± 8,700	352,000 ± 2,580	113,000 ± 470	100,000 ± 450	66,400 ± 360	48,400 ± 345	72,000 ± 380	67,600 ± 370	5,970,000 ± 24,400	4,440,000 ± 7,300	148,000 ± 1,760	115,000 ± 380
xperiment E /14/56 1530	6,220,000 ± 7,100	1,790,000 ± 3,800			6,760 ± 56	6,040 ± 54			320,000 ± 2,540	294,000[a] ± 660	7,000 ± 280	5,600 ± 170

a - corrected for partial spill

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TABLE II

DISTRIBUTION OF BETA ACTIVITY IN STACK SAMPLES FROM IET

Experiment Date and Hour Counted	1st millipore paper		1st back-up paper		2nd millipore paper		2nd back-up paper		1st scrubber solution		2nd scrubber solution	
	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m
Experiment C 1/56 130	332,000 ±940	34,500 ±420				5,300 ±115			500,000 ±6,350	425,000[a] ±1,370	16,500 ±900	10,000 ±50
Experiment D 1/12/56 1400	2,700,000 ±8,800	140,000 ±1,700			35,000 ±380				2,500,000 ±25,000	1,750,000 ±8,500	49,300 ±1,000	36,000 ±260

a -- corrected for partial sample spill

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TABLE III

DISTRIBUTION OF I_2 ACTIVITY IN STACK SAMPLES FROM IET [redacted]

Date and hour	Experiment No.	I_2 in 1st millipore paper c/m	I_2 in 1st scrubber solution c/m	I_2 in solution / I_2 on paper ratio
5/24/56 1230	A	7,100	15,400	2.16
6/1/56 2130	C	77,000	1,030,000	13.4
6/12/56 1050	D	352,000	4,440,000	12.6
6/14/56 1530	E	1,790,000	294,000	0.16

TABLE IV

RATIO OF I_2 ACTIVITY TO TOTAL ACTIVITY IN IET STACK SAMPLES

Date and hour	Experiment No.	Total activity Raw c/m	Total activity I_2 extract c/m	Total I_2 / Total activity
6/1/56 2130	C-gamma	2,070,000	1,107,000	0.53
	C-beta	832,000	459,500	0.55
6/12/56 1050	D-gamma	13,470,000	4,792,000	0.36
1400	D-beta	5,200,000	1,886,000	0.36
6/14/56 1530	E-gamma	6,540,000	2,084,000	0.32
		[redacted]		

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TABLE V

[redacted]

DISTRIBUTION OF GAMMA ACTIVITY IN FLUID SAMPLES

Experiment No. Date and hour	1st millipore paper		1st back-up paper		2nd millipore paper		1st scrubber solution	
	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m	Raw c/m	I_2 Extract c/m
S-A 6/6/56 1530		109 ±12						46 ±12
S-B 6/8/56 1200	1,010 ±30	340 ±26						90 ±12
S-C 6/9/56 1230	No significant activity							
S-D	No significant activity							
S-E 6/13/56 0930	270 ±22			No significant activity				
S-F 6/13/56 1550	4,400 ±48	1,220 ±32	625 ±28	380 ±26	140 ±24	110 ±24	830 ±120	530 ±18
S-G 6/15/56 1500	2,300 ±36	444 ±22						220 ±14
S-H 6/14/56 1300	280 ±22			No significant activity				
S-I 6/15/56 1500	3,660 ±44	1,310 ±30			220 ±24		700 ±120	520 ±18

[redacted]

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[redacted]

TABLE VI

DISTRIBUTION OF I_2 ACTIVITY IN FIELD SAMPLES

Date and hour	Experiment No.	I_2 in 1st millipore paper c/m	I_2 in 1st scrubber solution c/m	$\frac{I_2 \text{ in solution}}{I_2 \text{ on paper}}$ ratio
6/13/56 1550	S-F	1,220	530	0.43 ±0.03
6/15/56 1500	S-G	444	220	0.50 ±0.06
6/15/56 1500	S-I	1,310	520	0.40 ±0.03

TABLE VII

RATIO OF I_2 ACTIVITY TO TOTAL ACTIVITY IN FIELD SAMPLES

Date and hour	Experiment No.	Total Activity Raw c/m	Total Activity I_2 Extract c/m	$\frac{\text{Total } I_2}{\text{Total Activity}}$ Ratio
6/13/56 1550	S-F	5,230 ±130	1,750 ±37	0.34 ±0.03
6/15/56 1500	S-I	4,360 ±128	1,830 ±36	0.42 ±0.03

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[redacted]

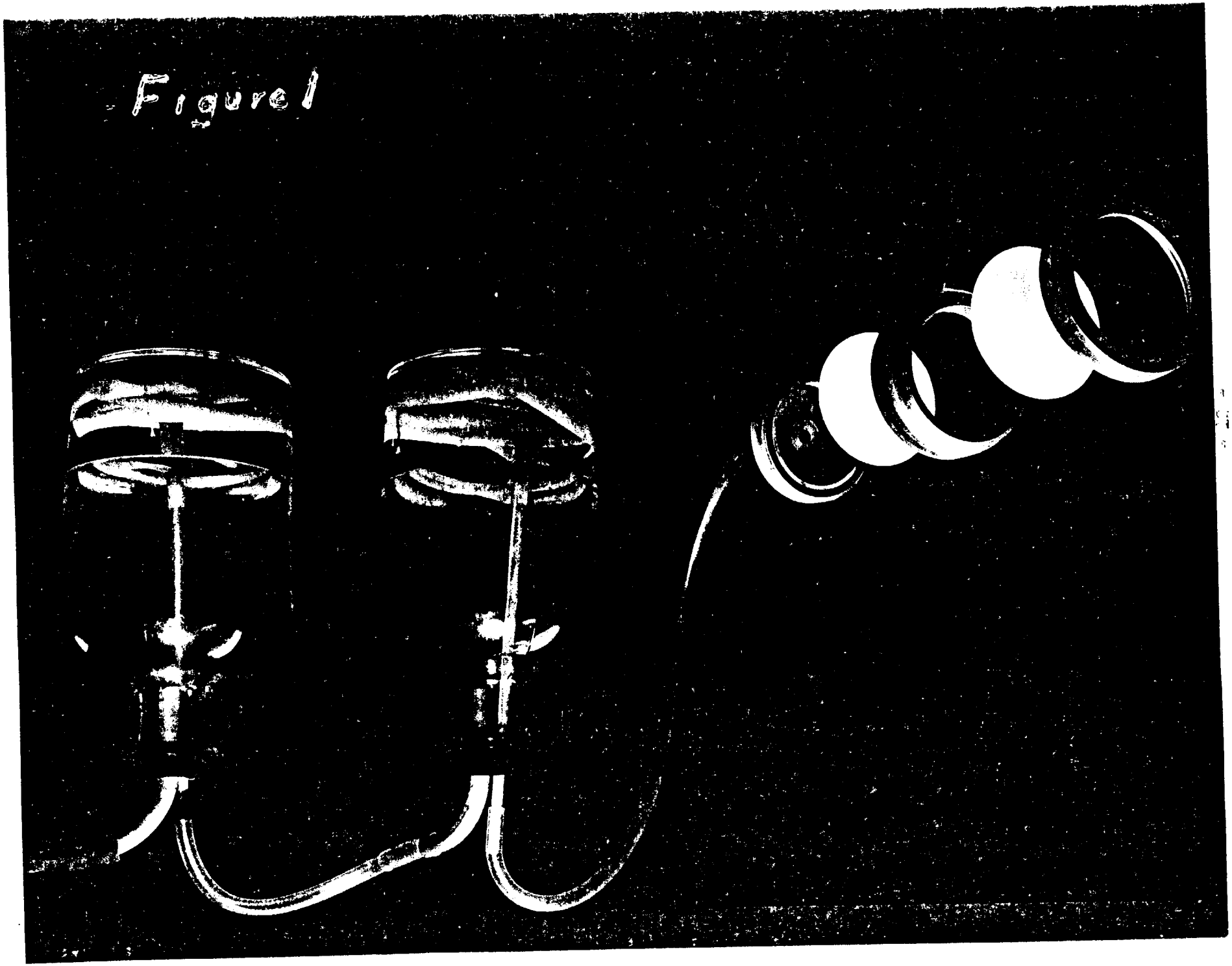

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FIGURE 2

ANALYTICAL FLOW SHEET OF ANP STACK EXPERIMENTS

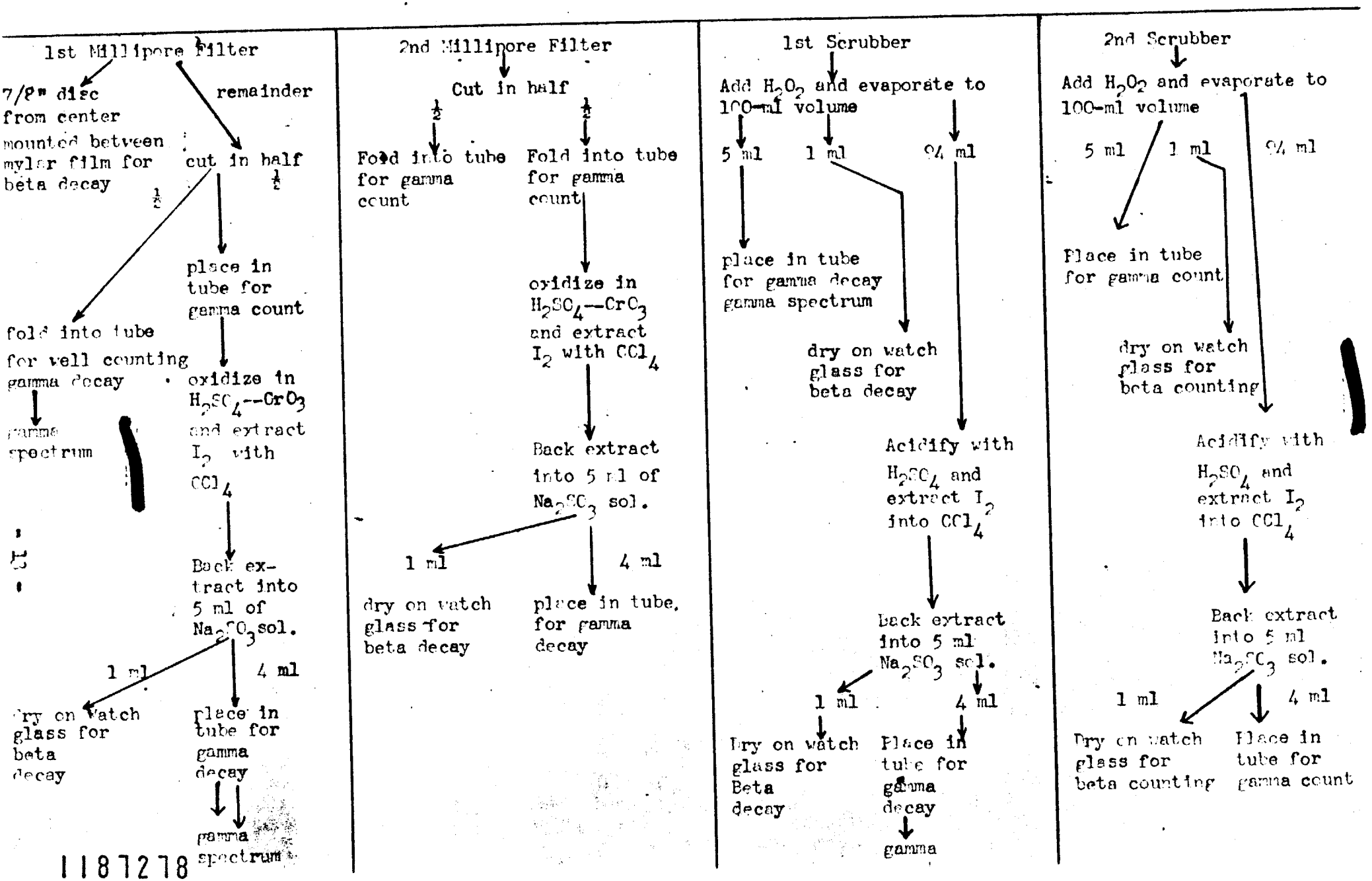

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Figure 3

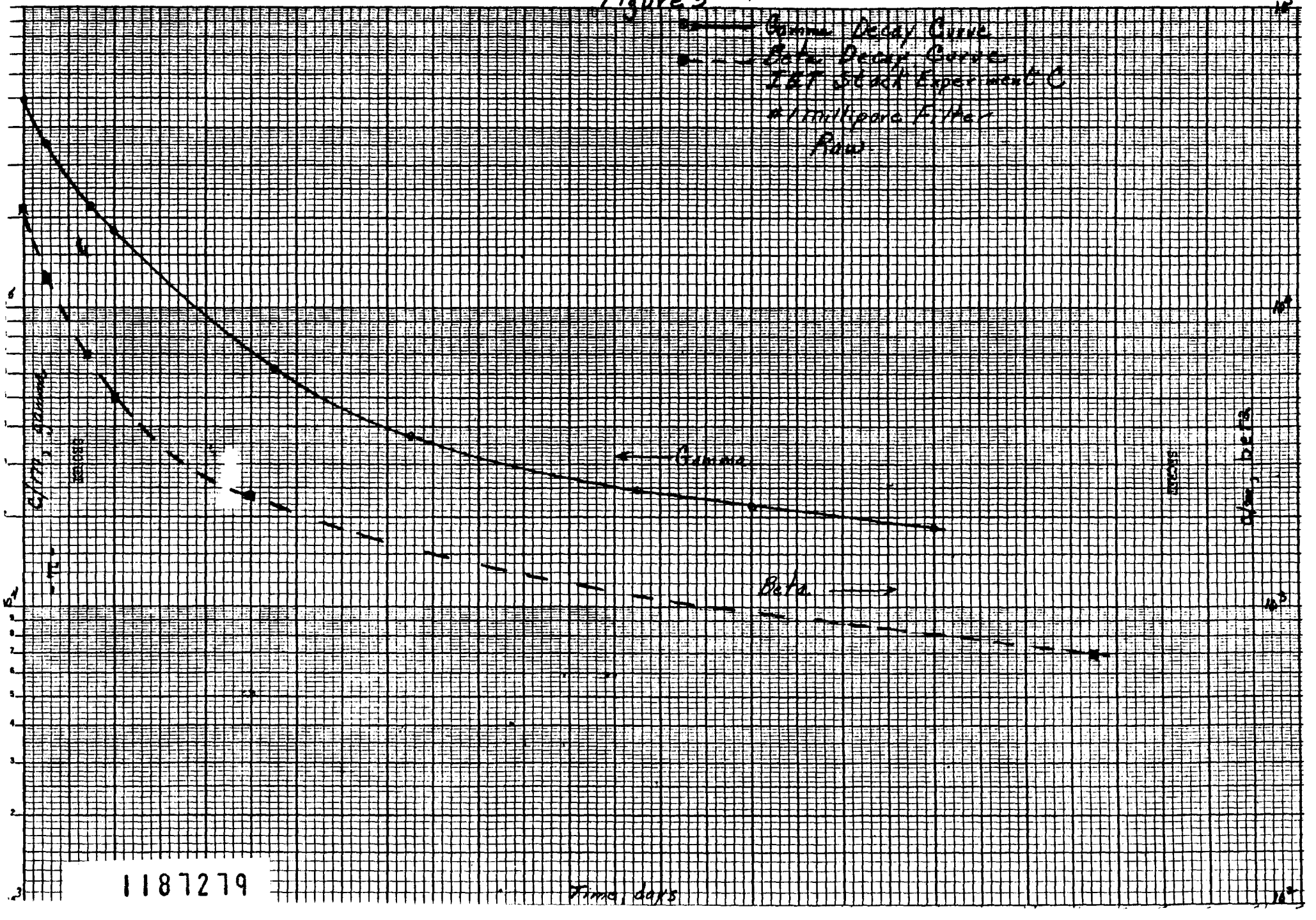

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Figure 4

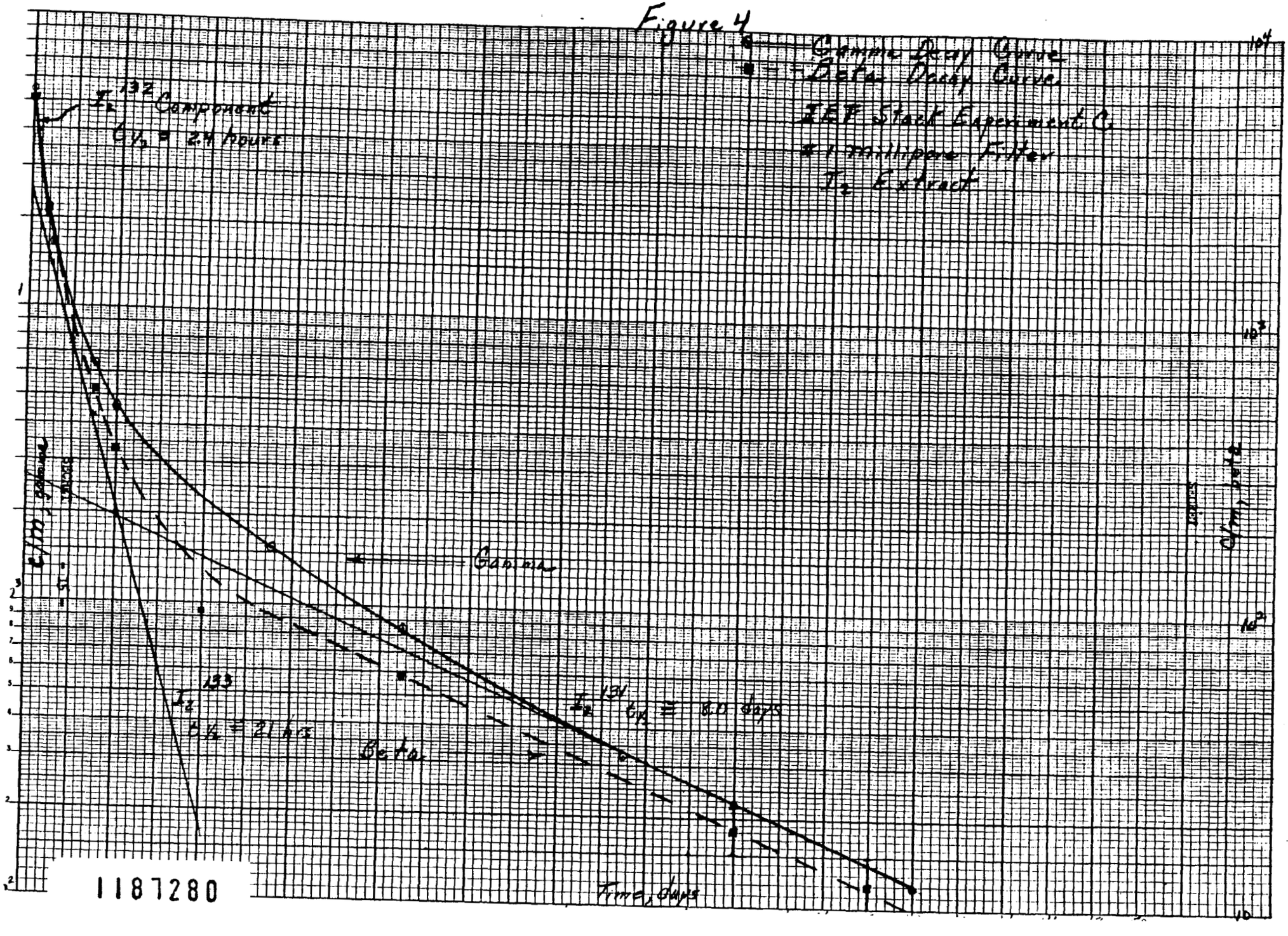

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Figure 5a

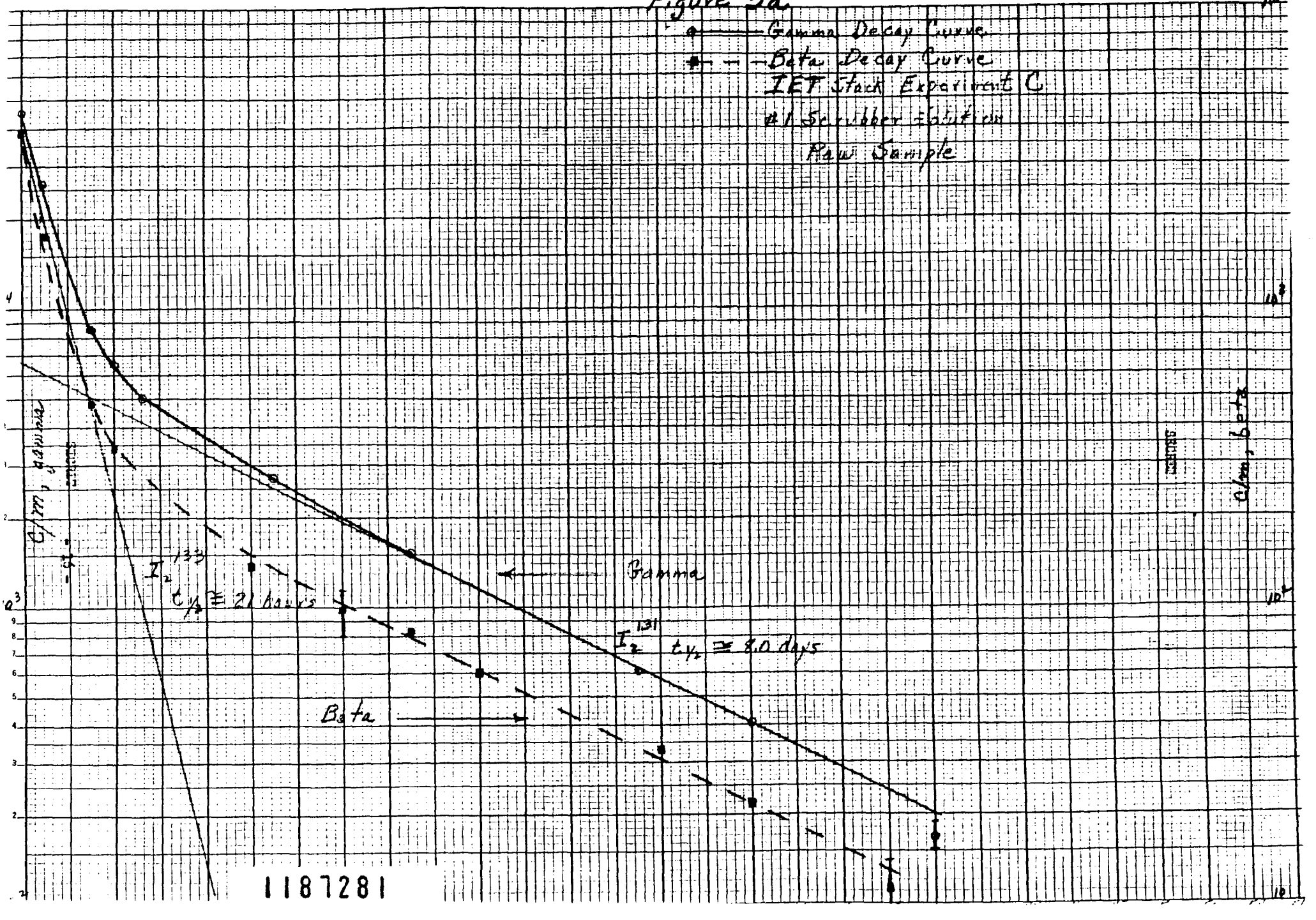

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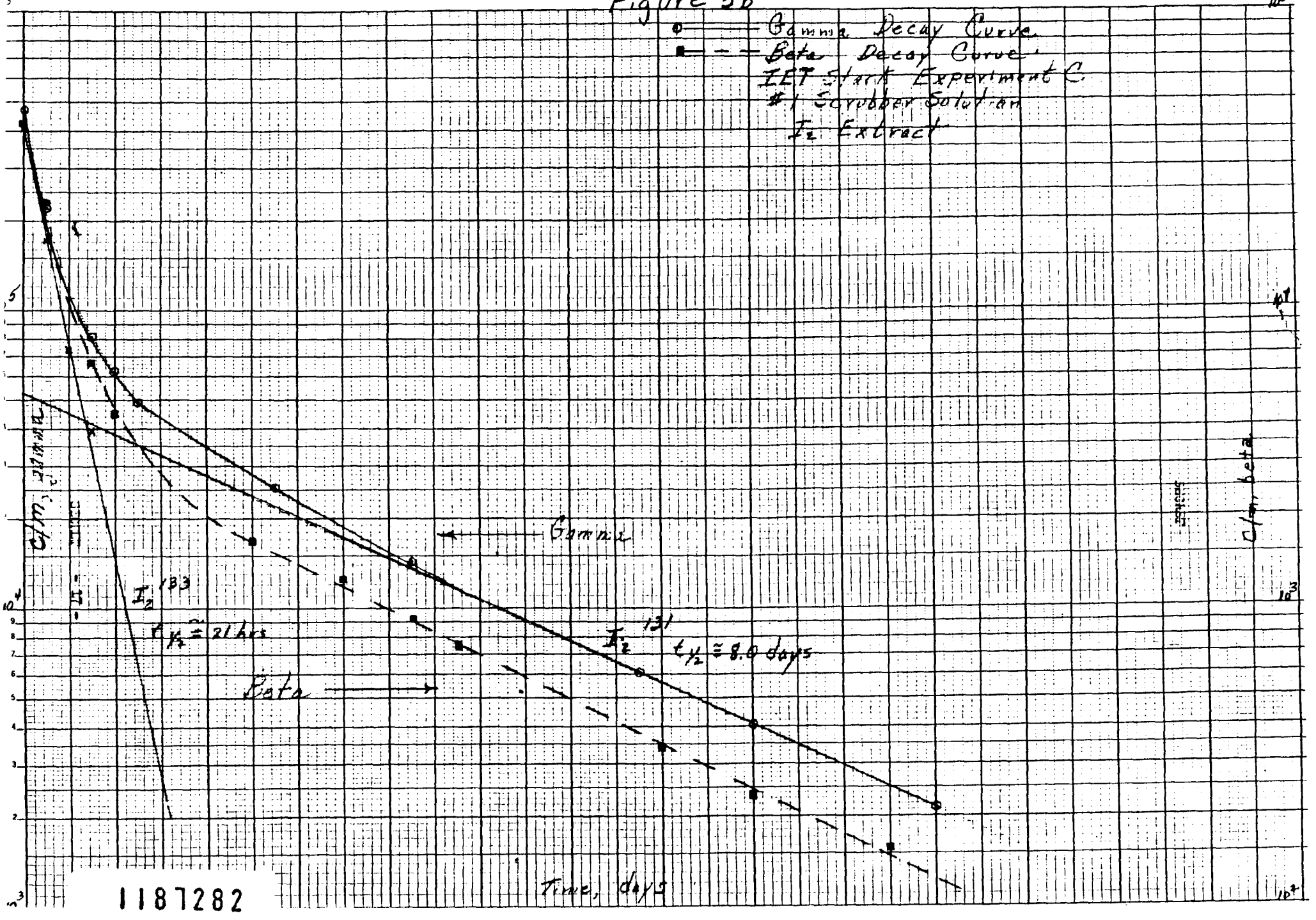
Figure 5b



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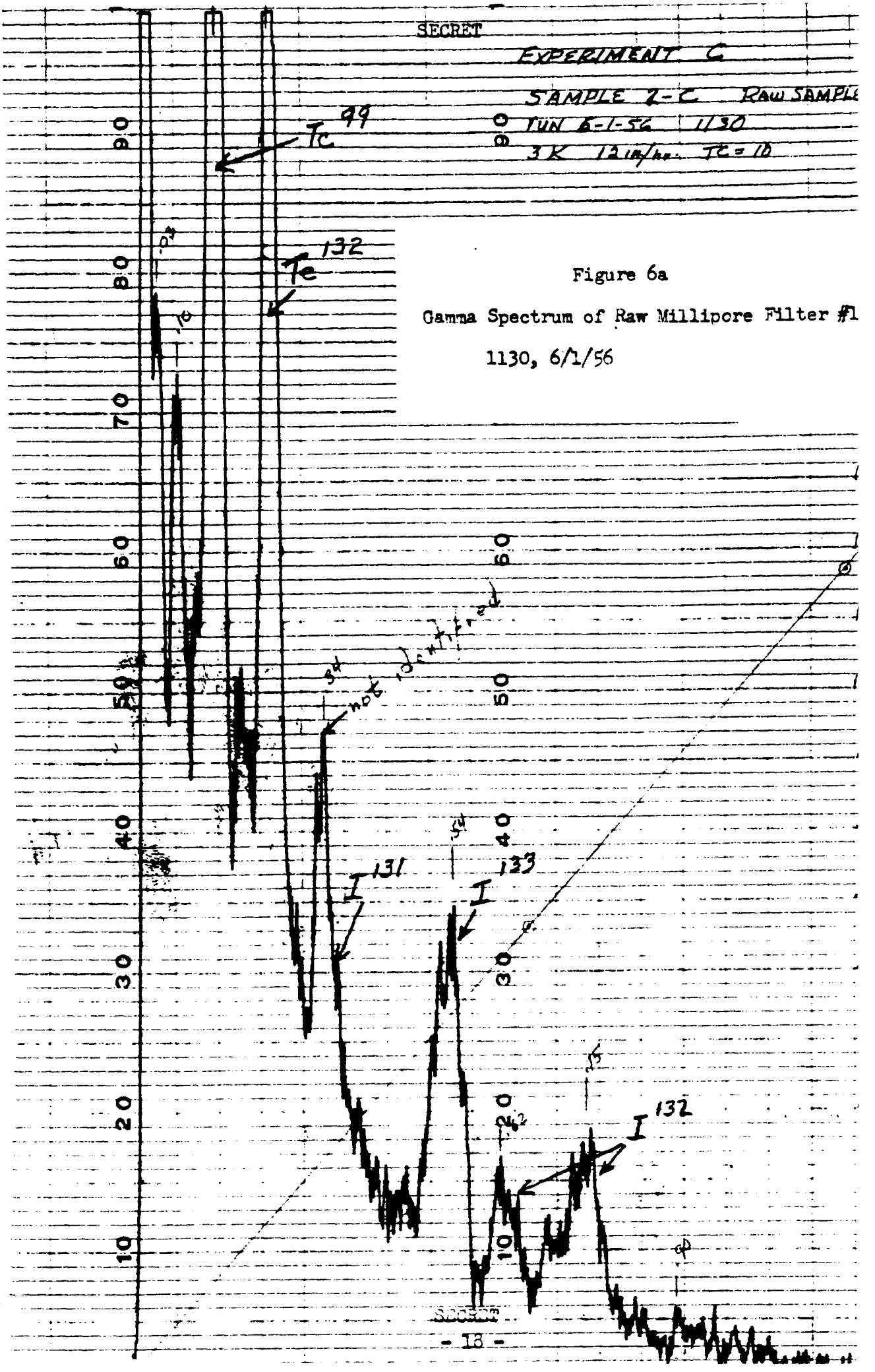

Figure 6a

Gamma Spectrum of Raw Millipore Filter #1

1130, 6/1/56

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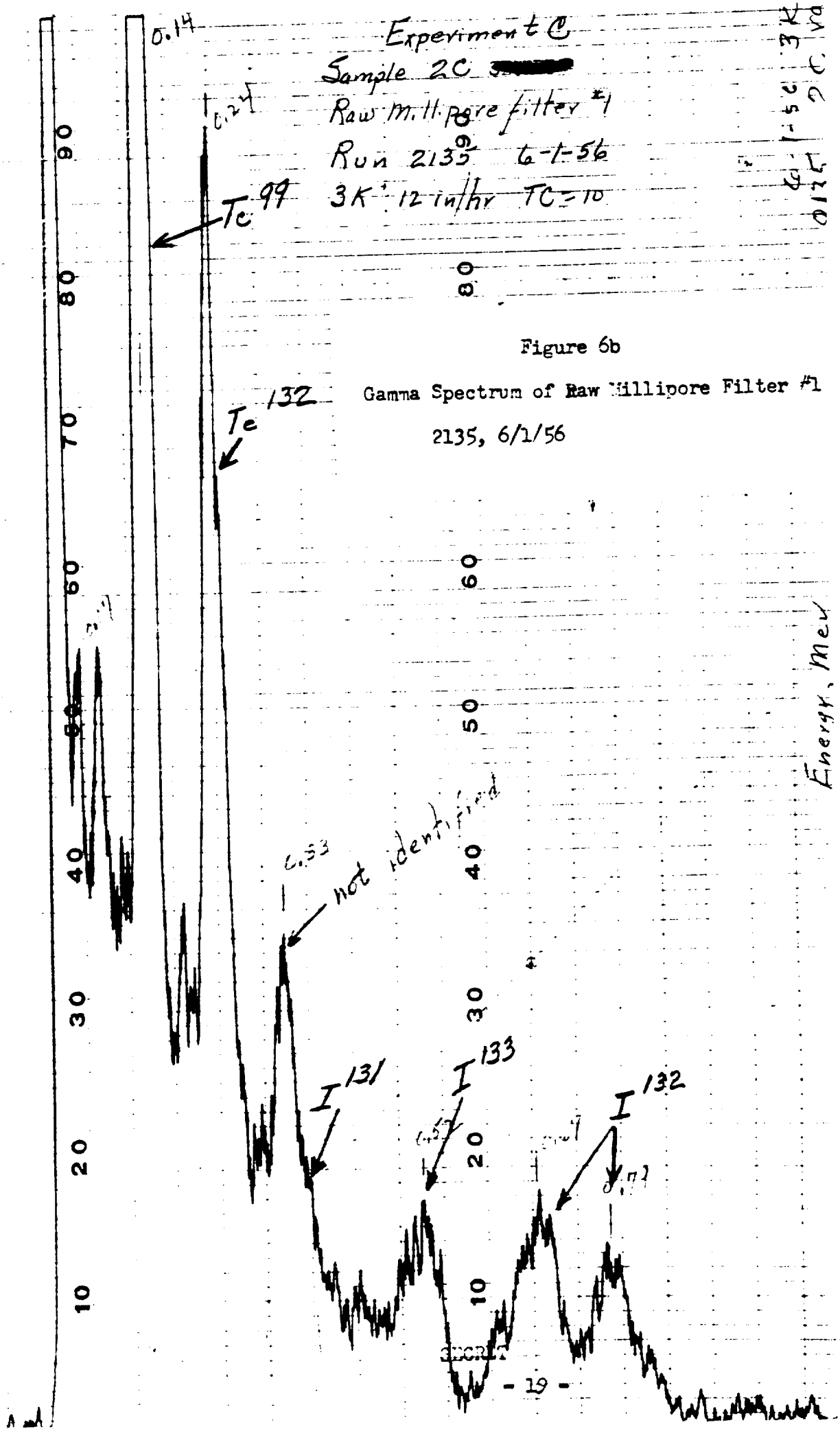

Figure 6b

Gamma Spectrum of Raw Millipore Filter #1

2135, 6/1/56

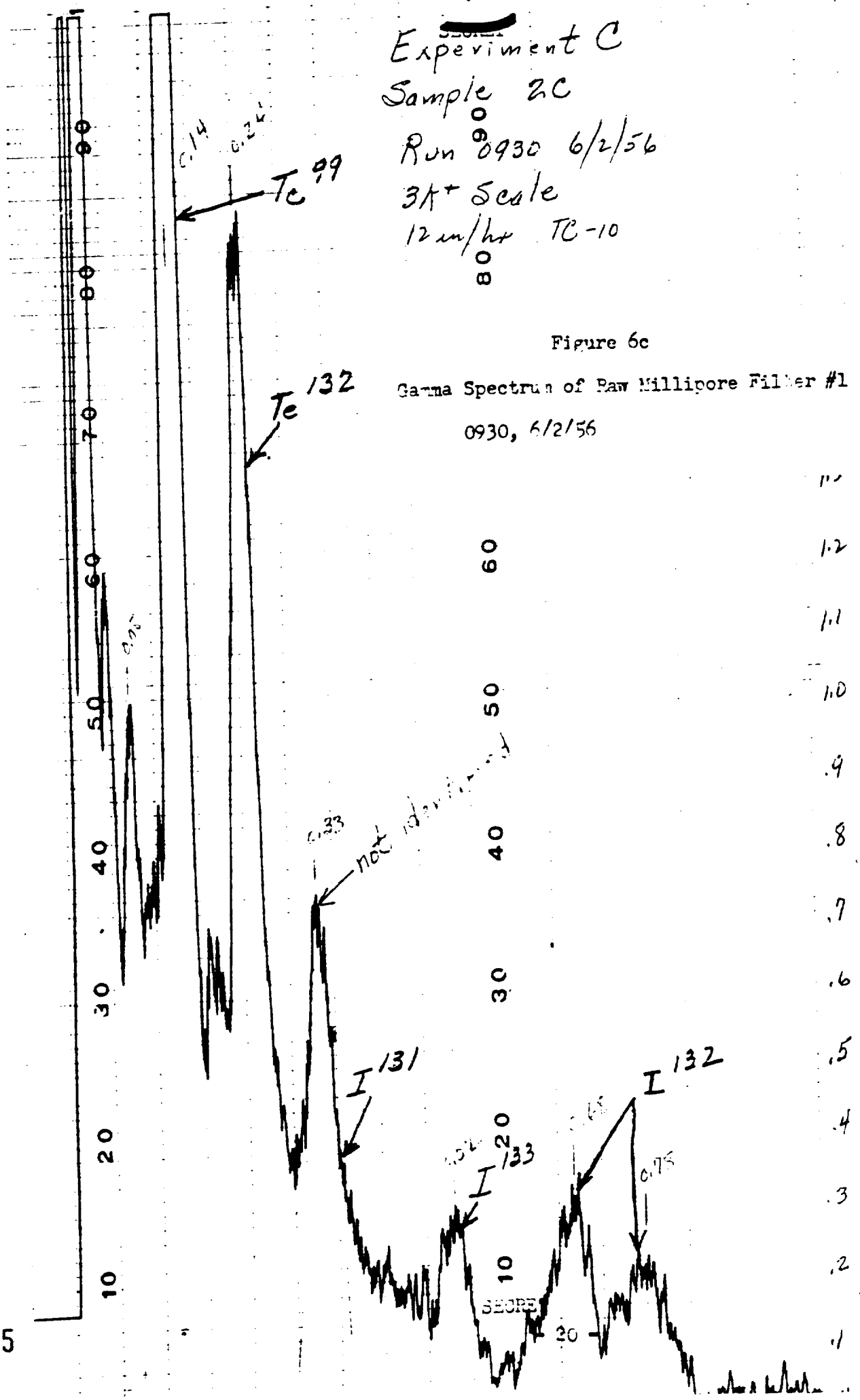

Figure 6c

Gamma Spectrum of Raw Millipore Filter #1

0930, 6/2/56

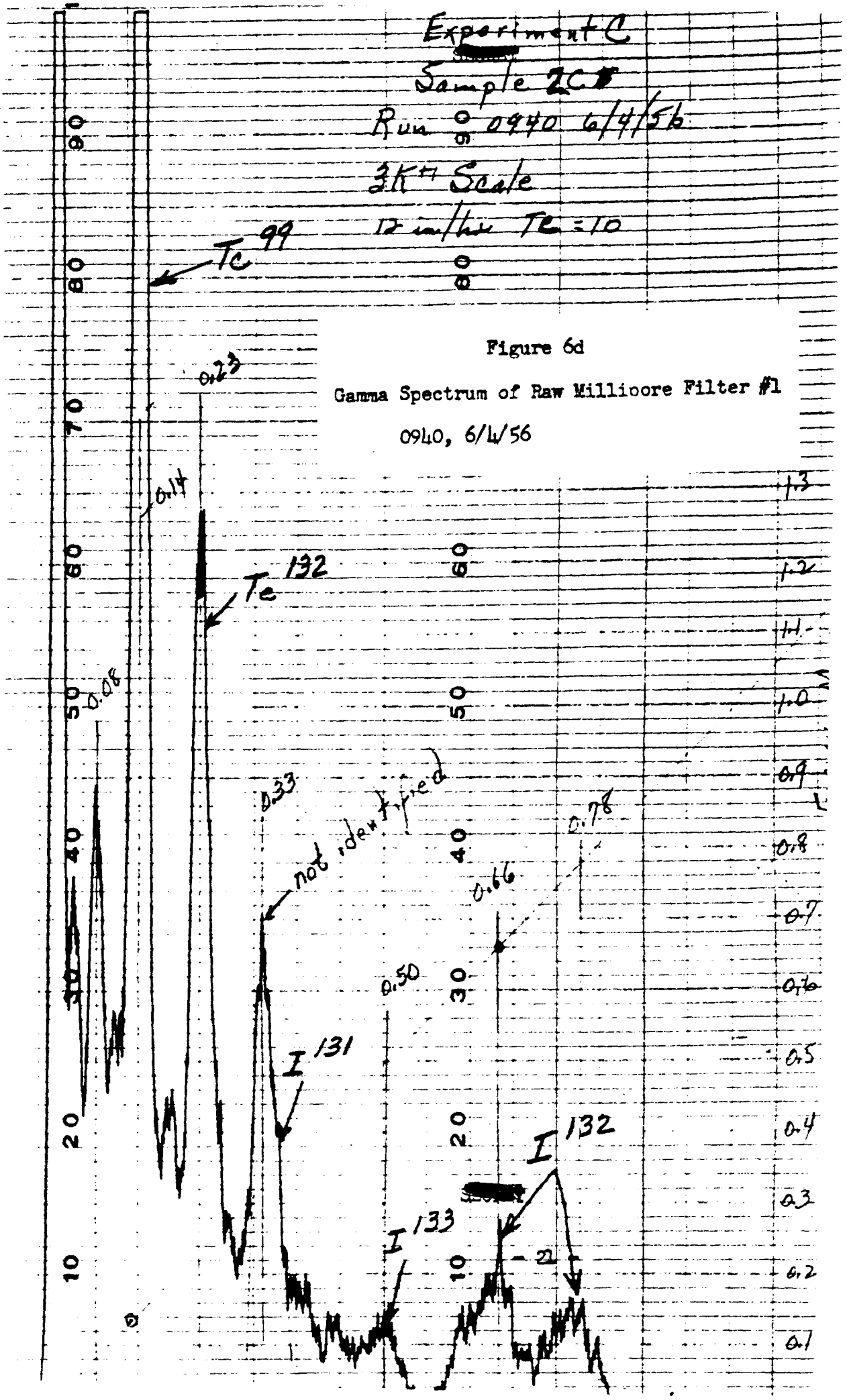

Figure 6d

Gamma Spectrum of Raw Millipore Filter #1

0940, 6/4/56

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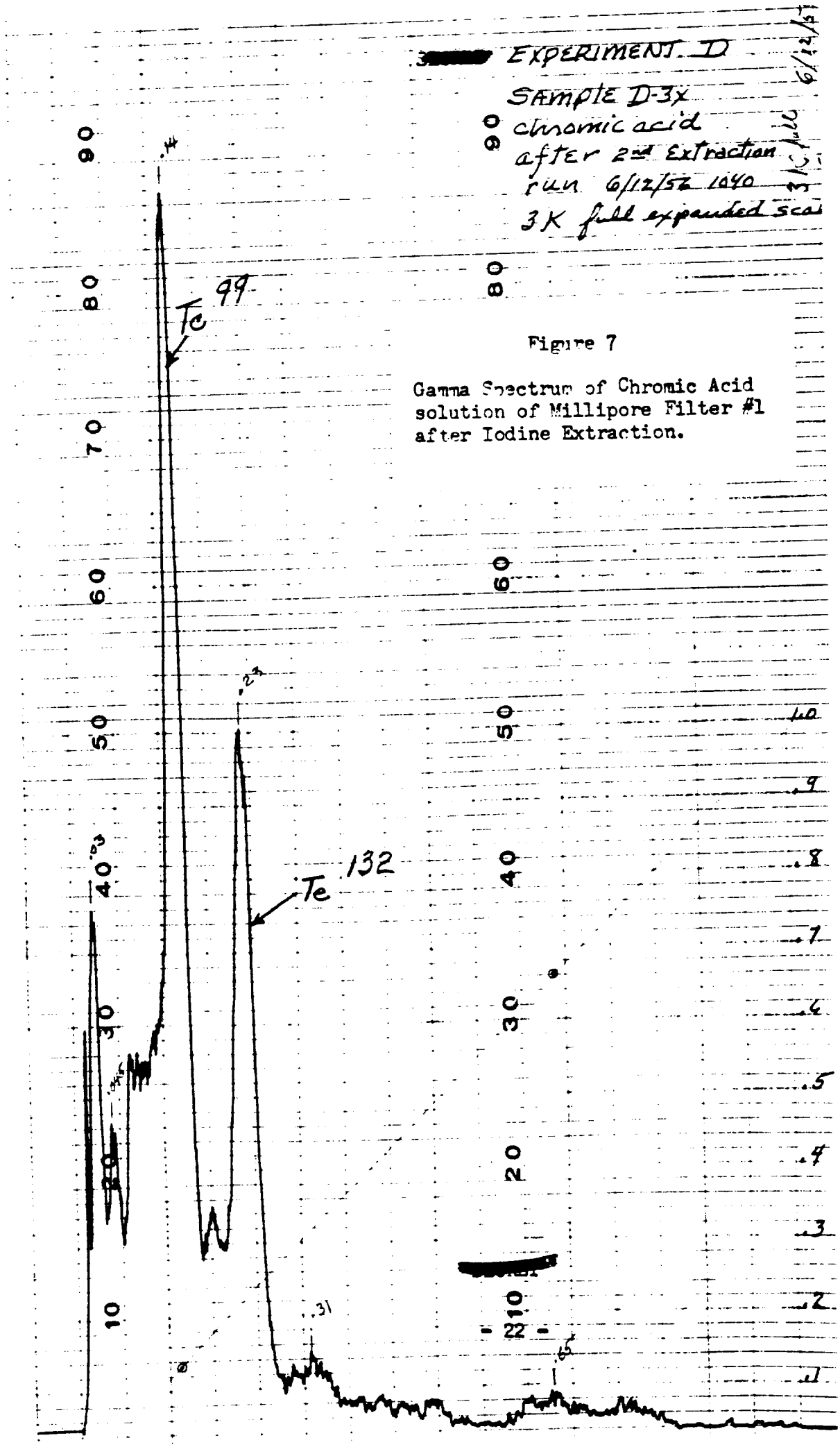

Figure 7

Gamma Spectrum of Chromic Acid solution of Millipore Filter #1 after Iodine Extraction.

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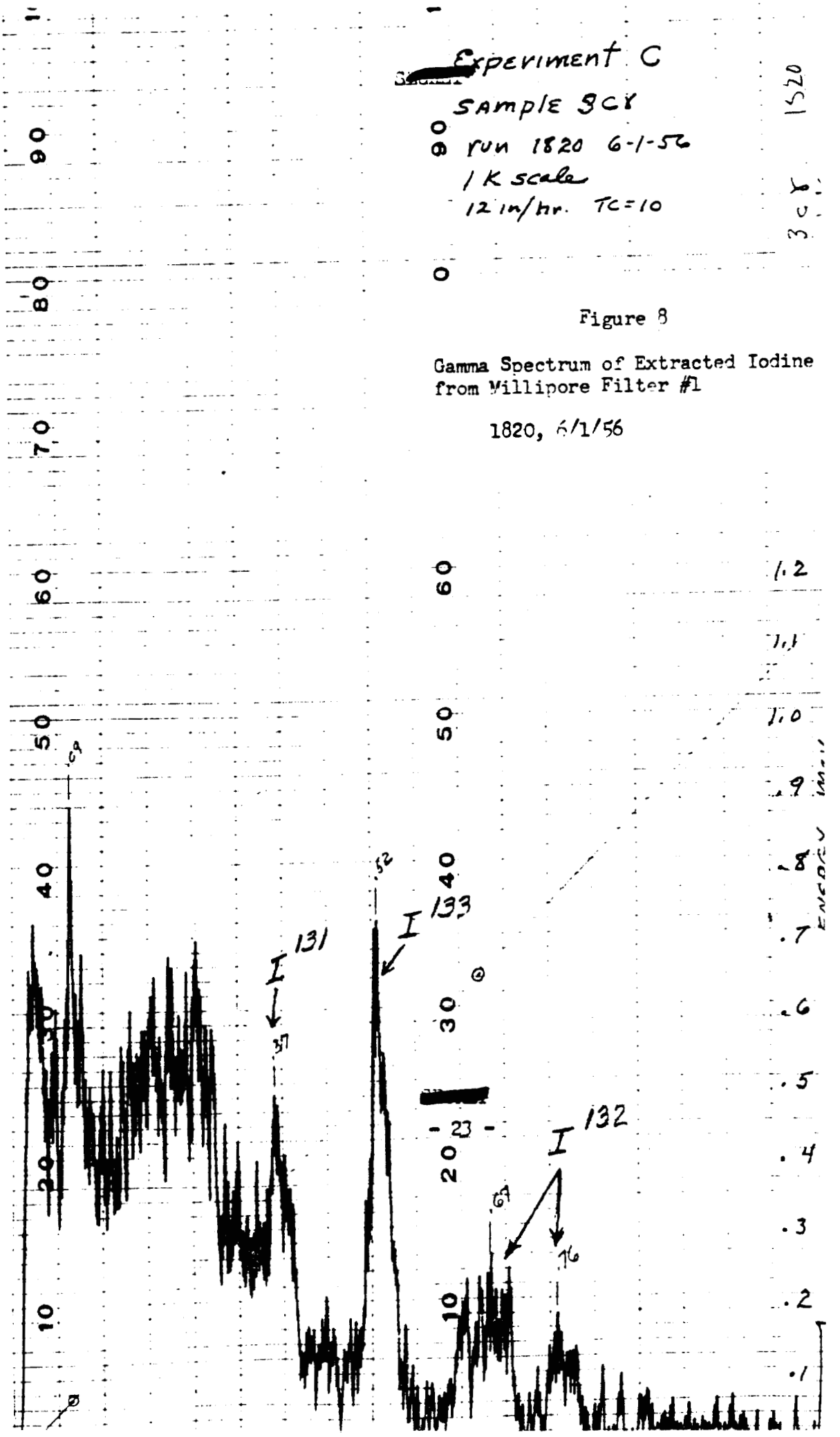


Figure 8

Gamma Spectrum of Extracted Iodine from Millipore Filter #1

1820, 6/1/56

1187288

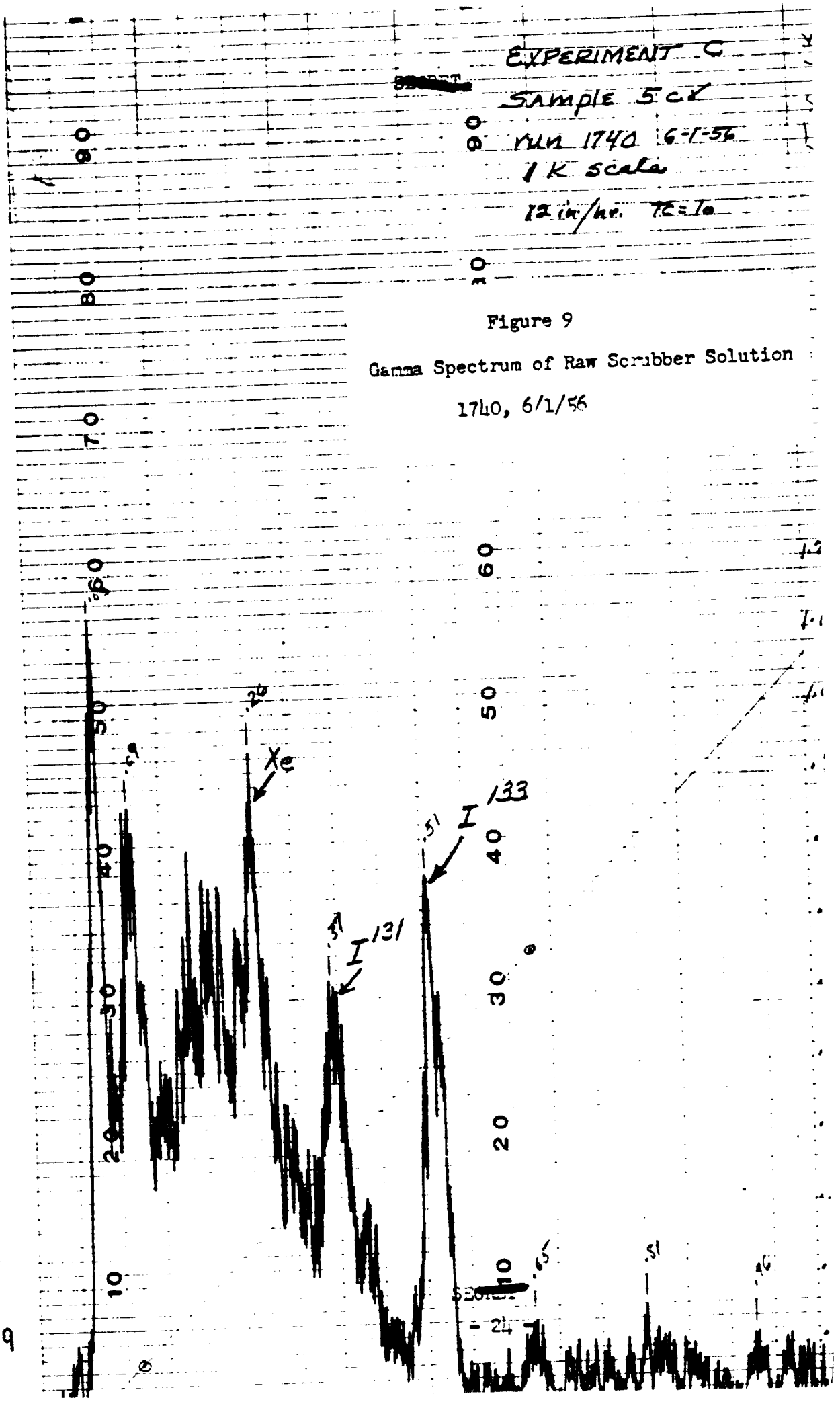


Figure 9

Gamma Spectrum of Raw Scrubber Solution

1740, 6/1/56

1187289

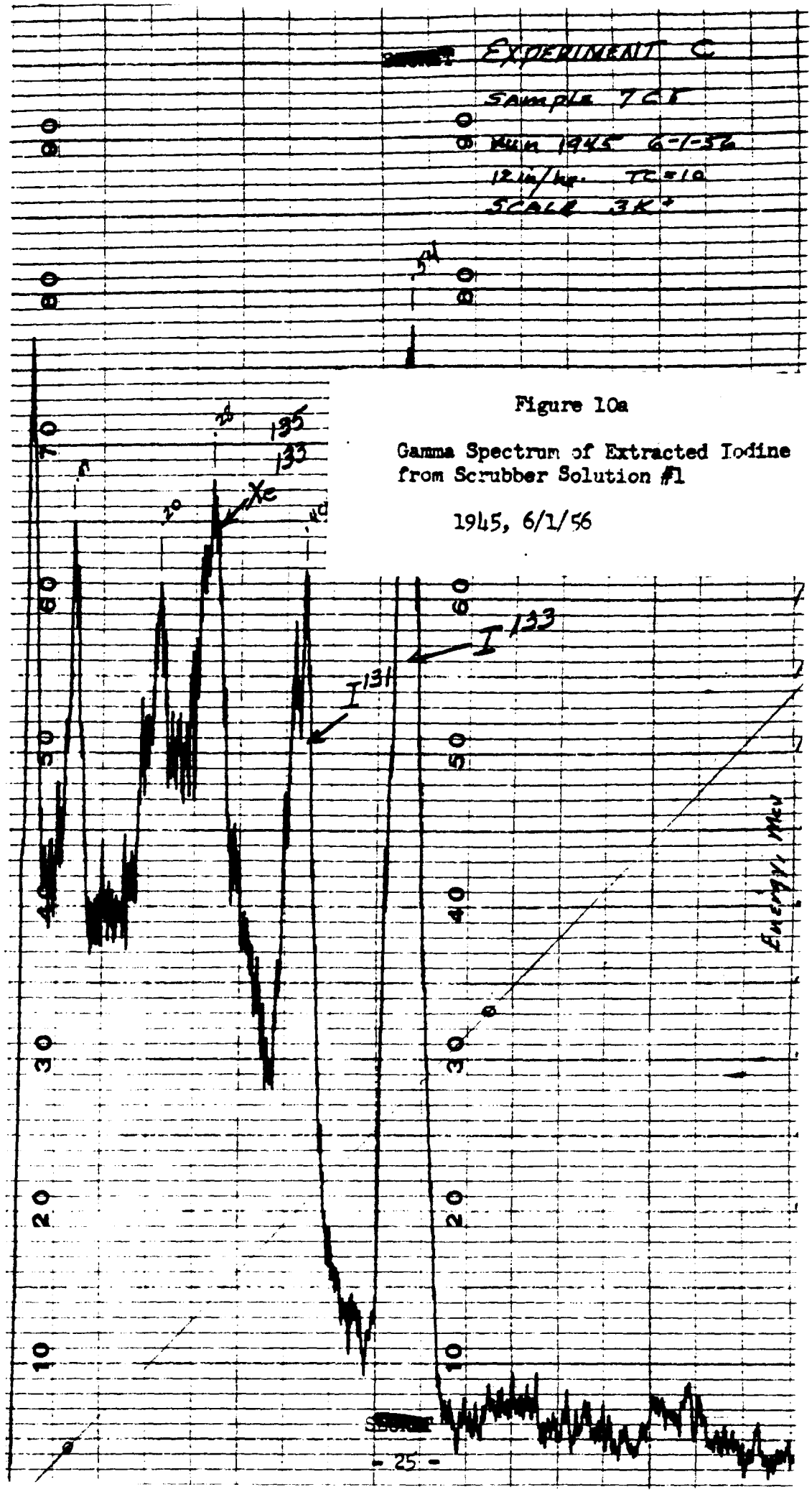


Figure 10a

Gamma Spectrum of Extracted Iodine from Scrubber Solution #1

1945, 6/1/56

1187290

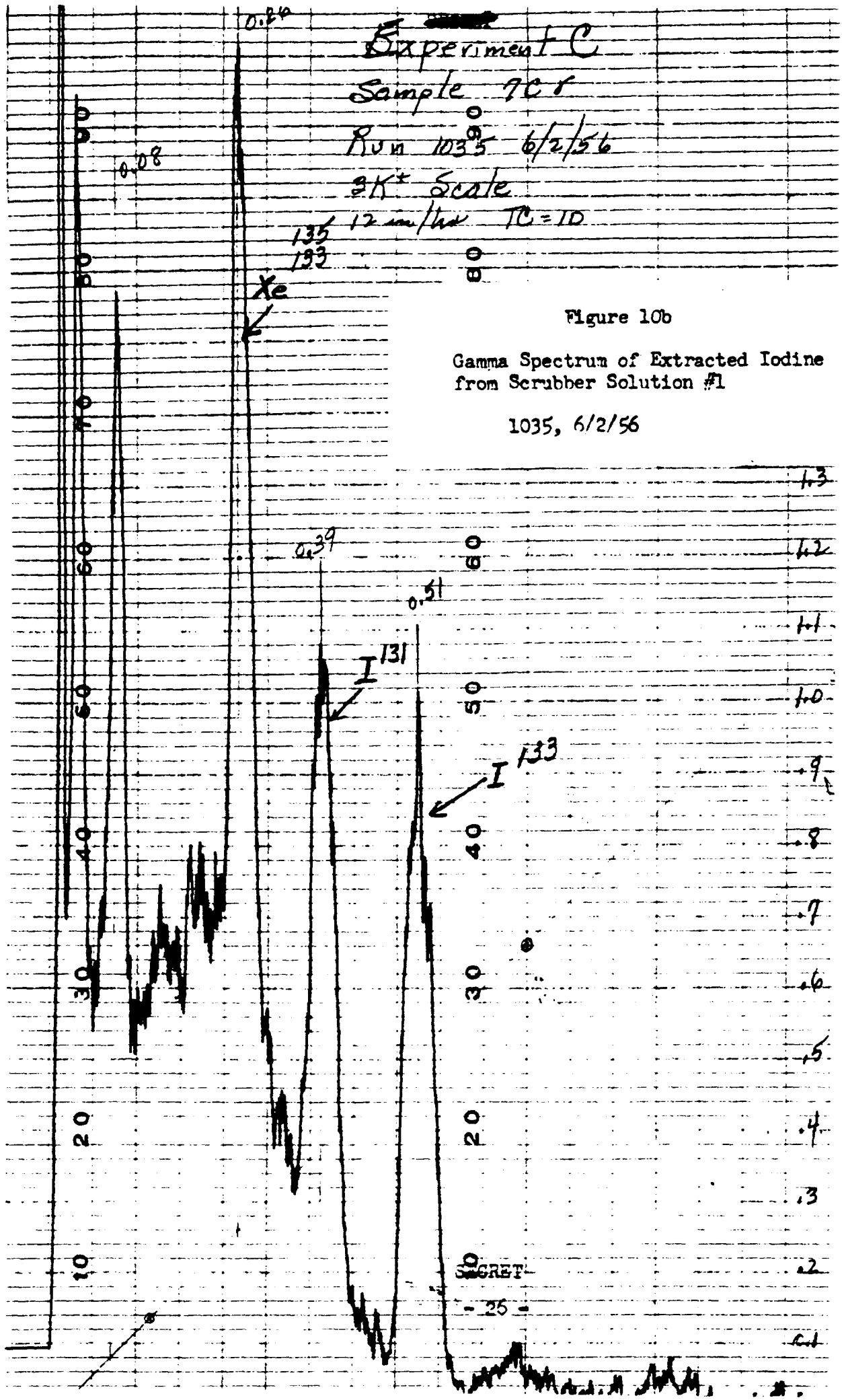


Figure 10b

Gamma Spectrum of Extracted Iodine from Scrubber Solution #1

1035, 6/2/56

1187291

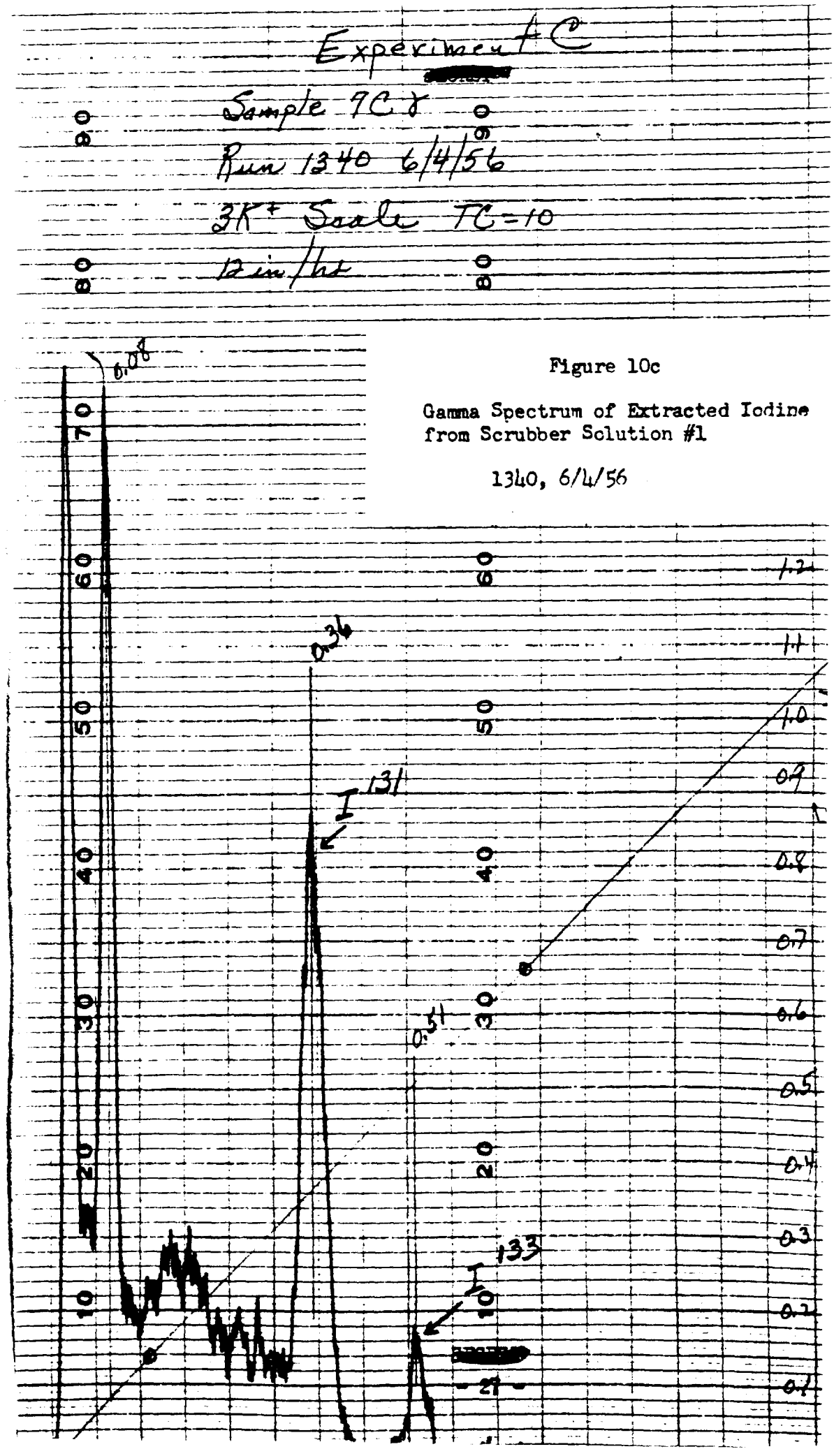


Figure 10c

Gamma Spectrum of Extracted Iodine from Scrubber Solution #1

1340, 6/4/56

1187292

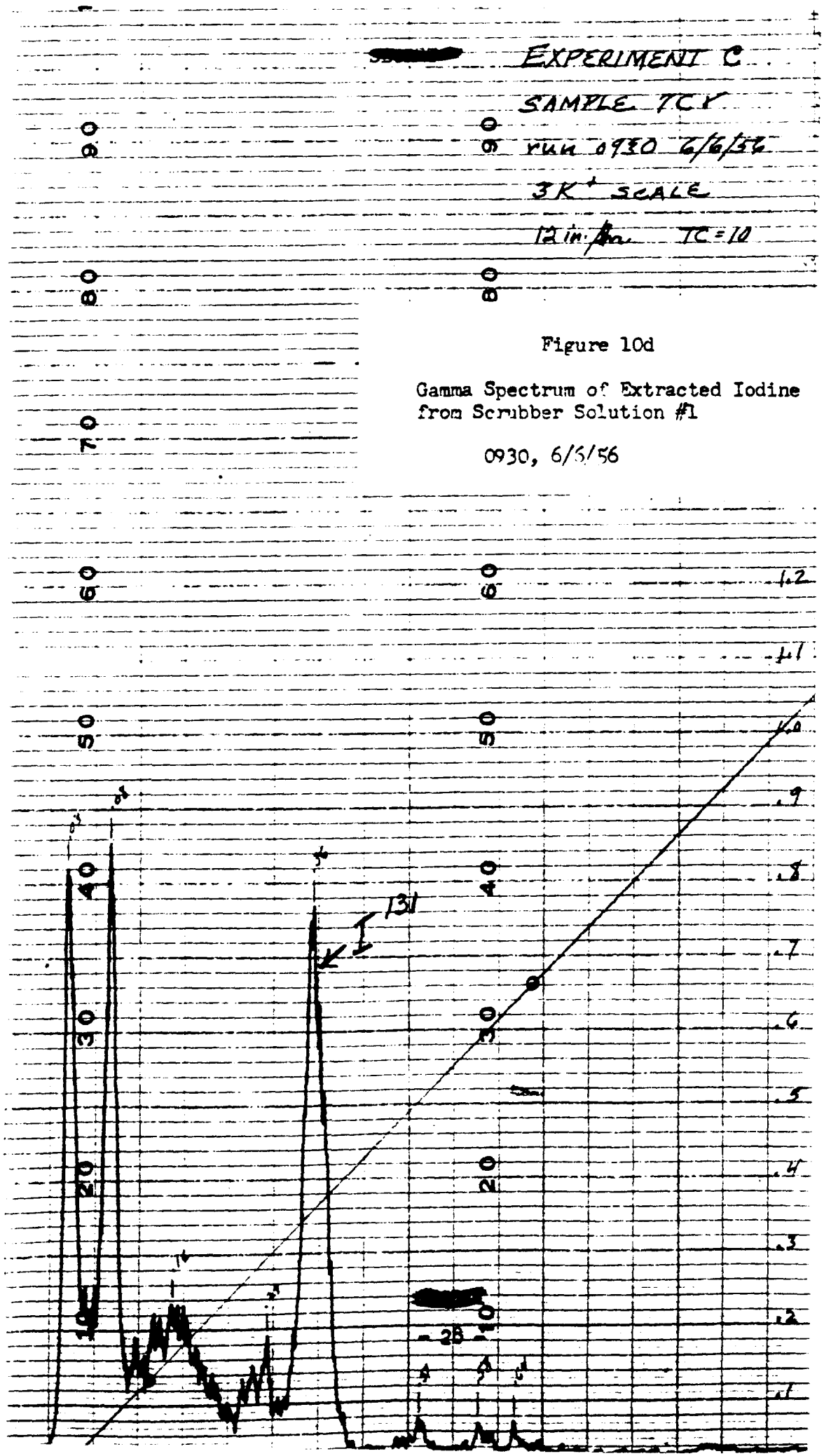


Figure 10d

Gamma Spectrum of Extracted Iodine from Scrubber Solution #1

0930, 6/6/56

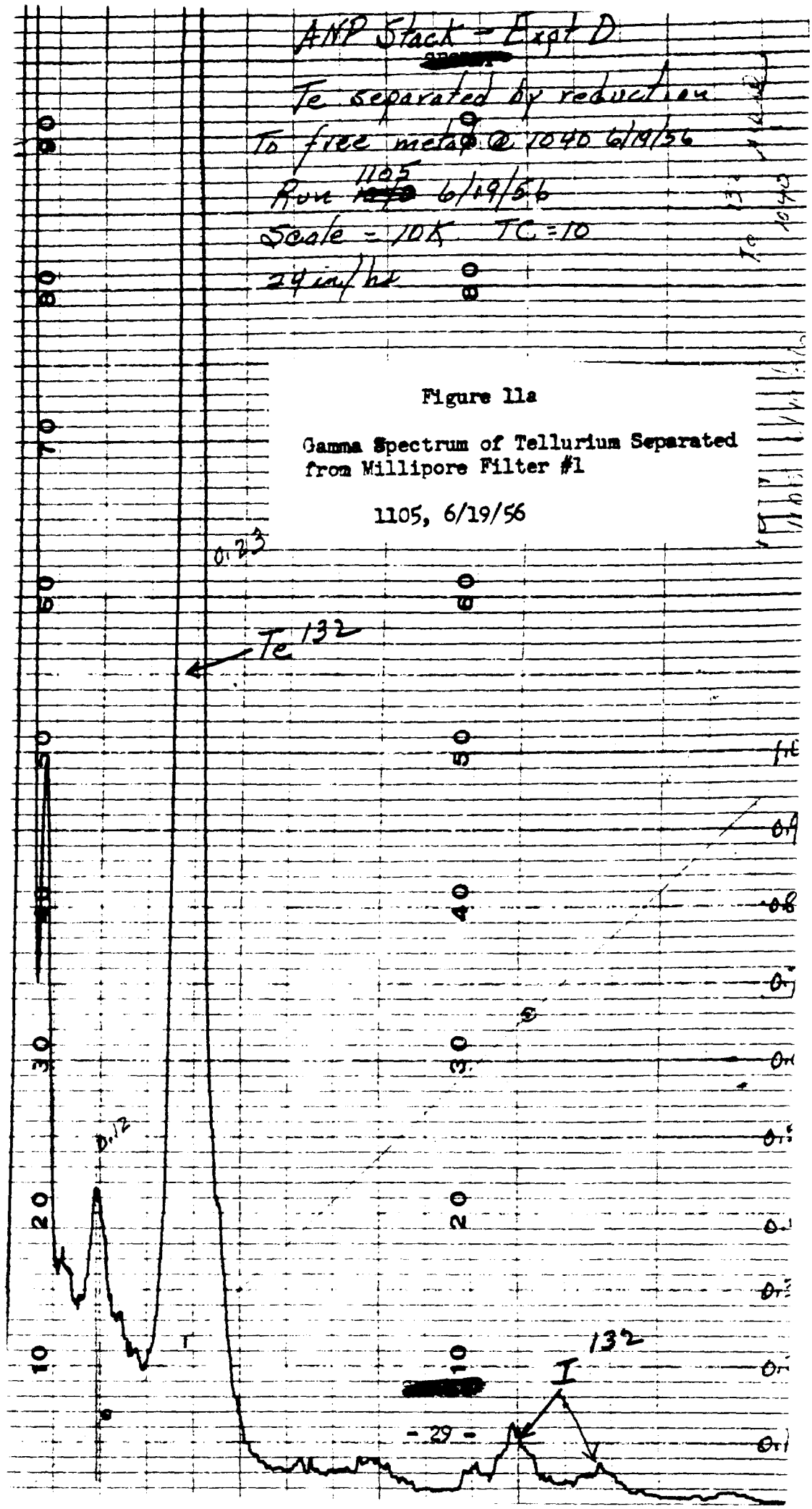


Figure 11a

Gamma Spectrum of Tellurium Separated from Millipore Filter #1

1105, 6/19/56

1187294

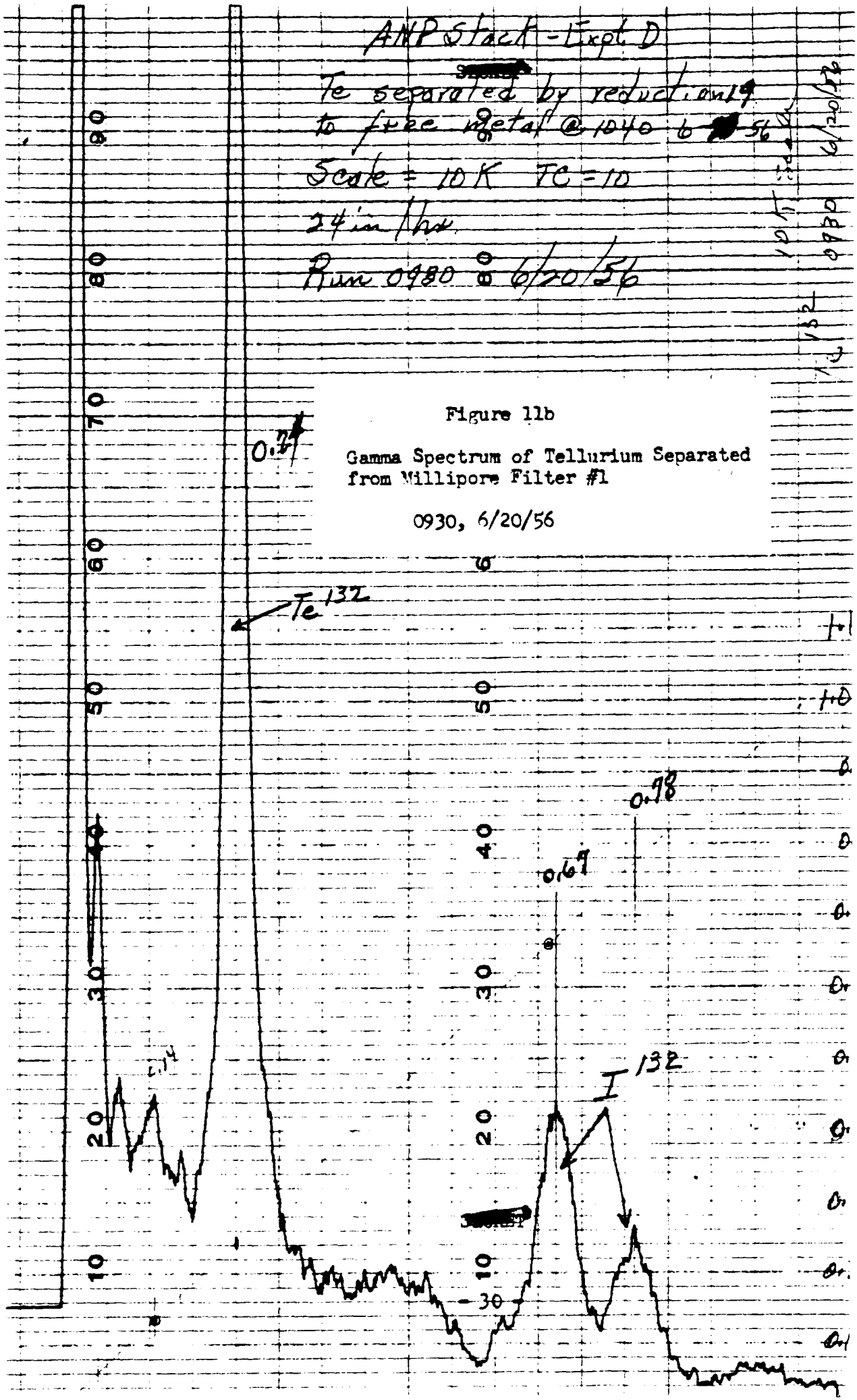


Figure 11b

Gamma Spectrum of Tellurium Separated from Millipore Filter #1

0930, 6/20/56

1187295

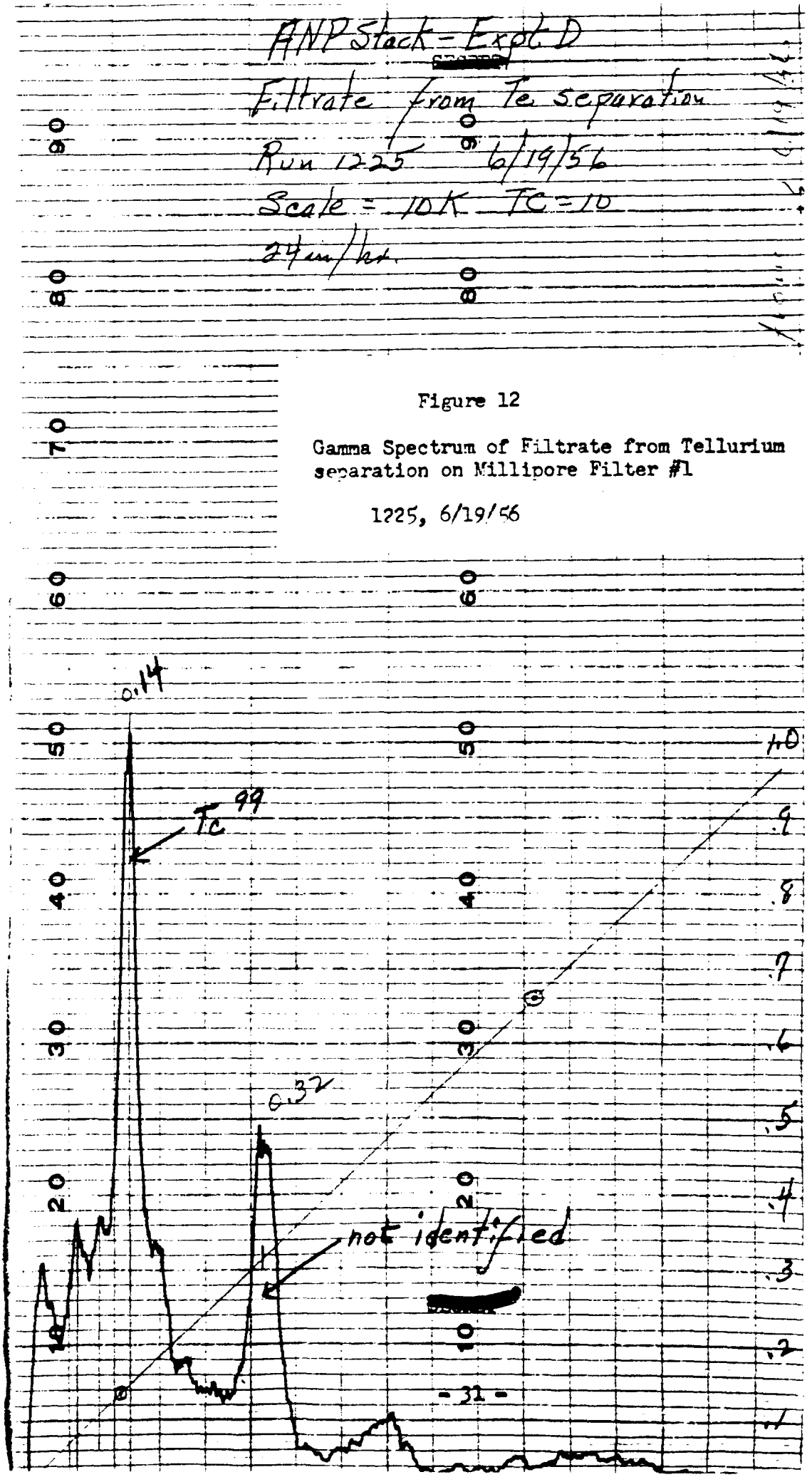


Figure 12

Gamma Spectrum of Filtrate from Tellurium separation on Millipore Filter #1

1225, 6/19/56

1187296

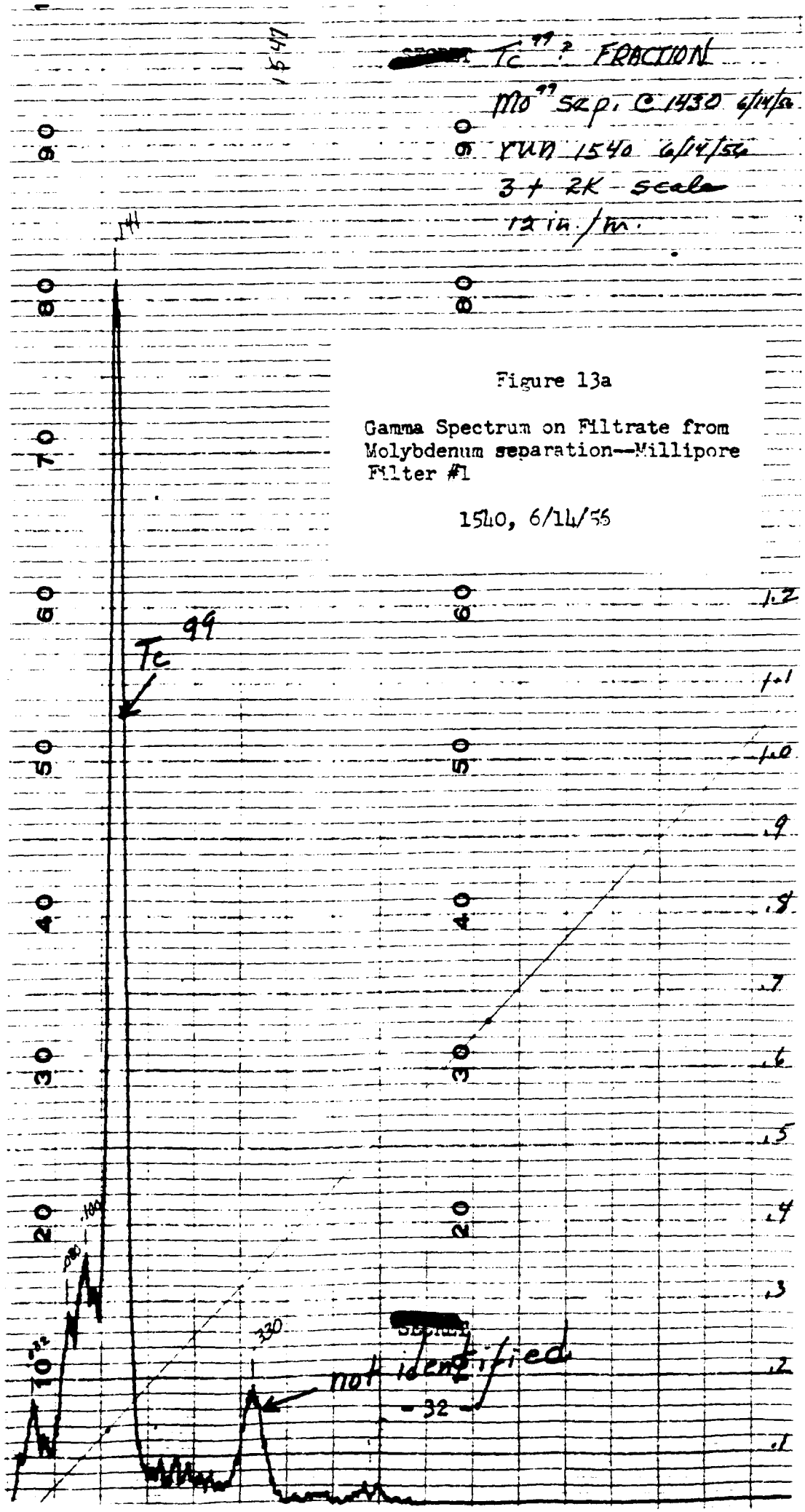


Figure 13a

Gamma Spectrum on Filtrate from Molybdenum separation—Millipore Filter #1

1540, 6/14/56

1187297

[redacted]

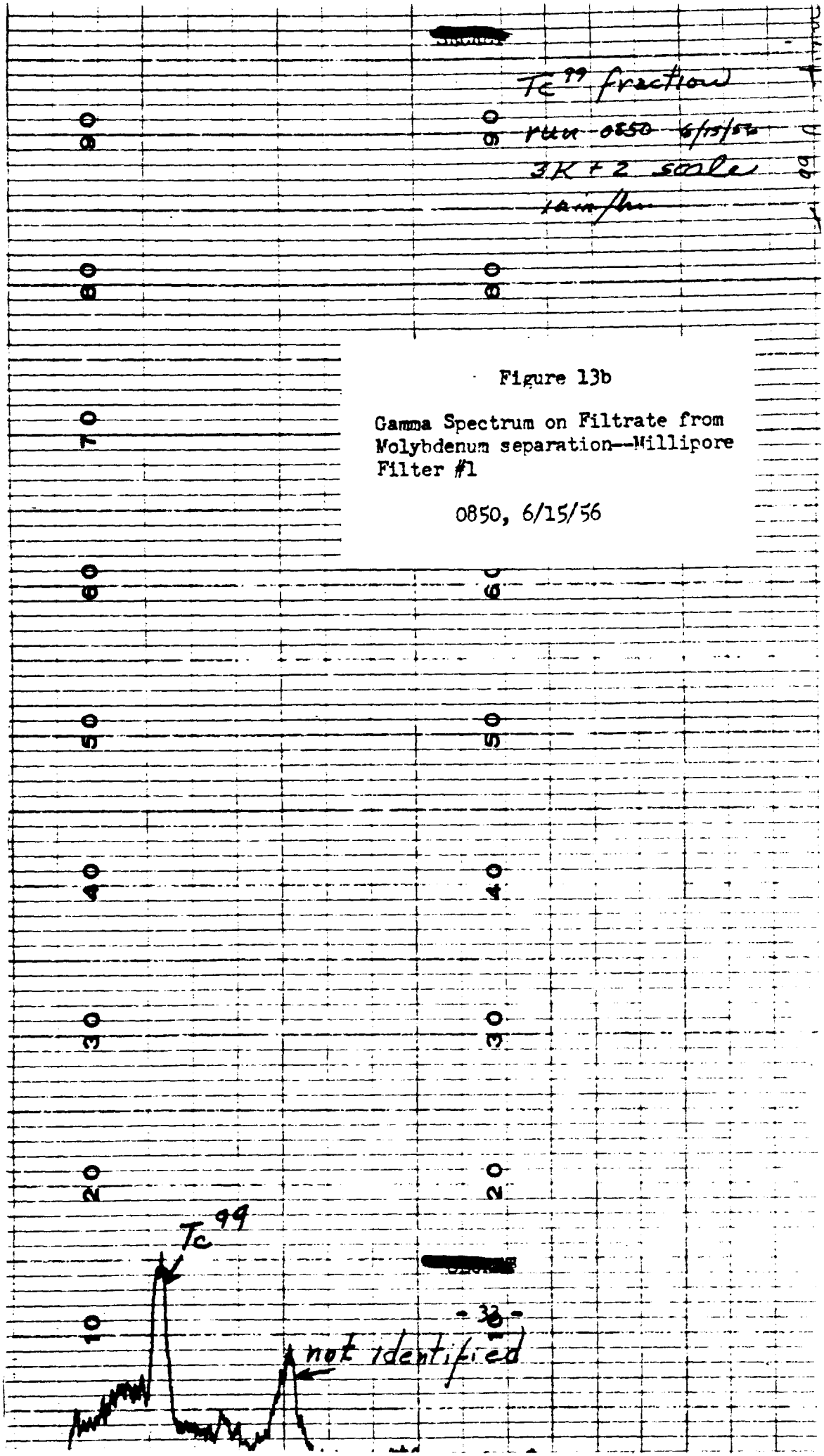


Figure 13b

Gamma Spectrum on Filtrate from Molybdenum separation--Millipore Filter #1

0850, 6/15/56

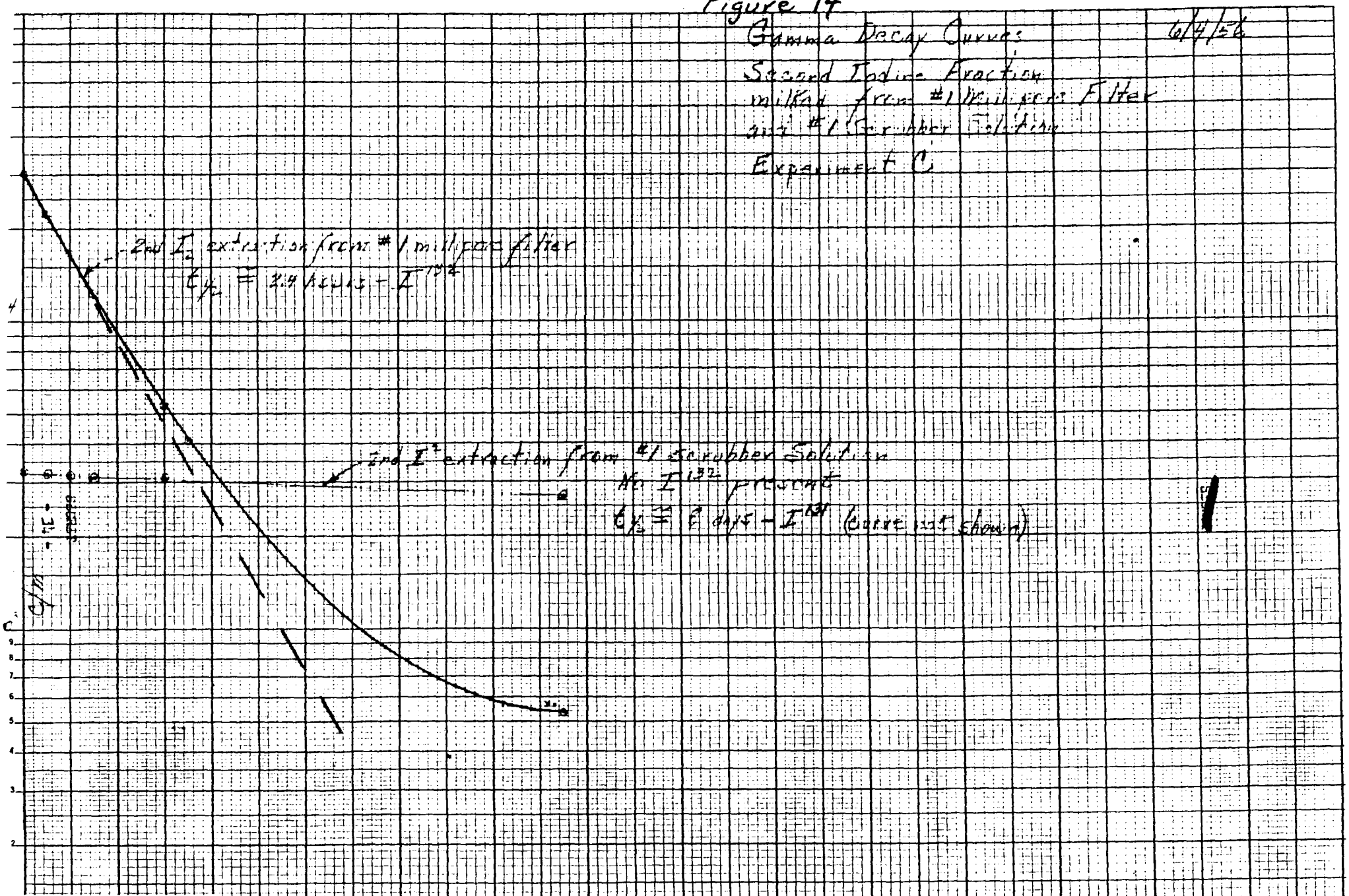
[redacted]

1187298

Figure 14

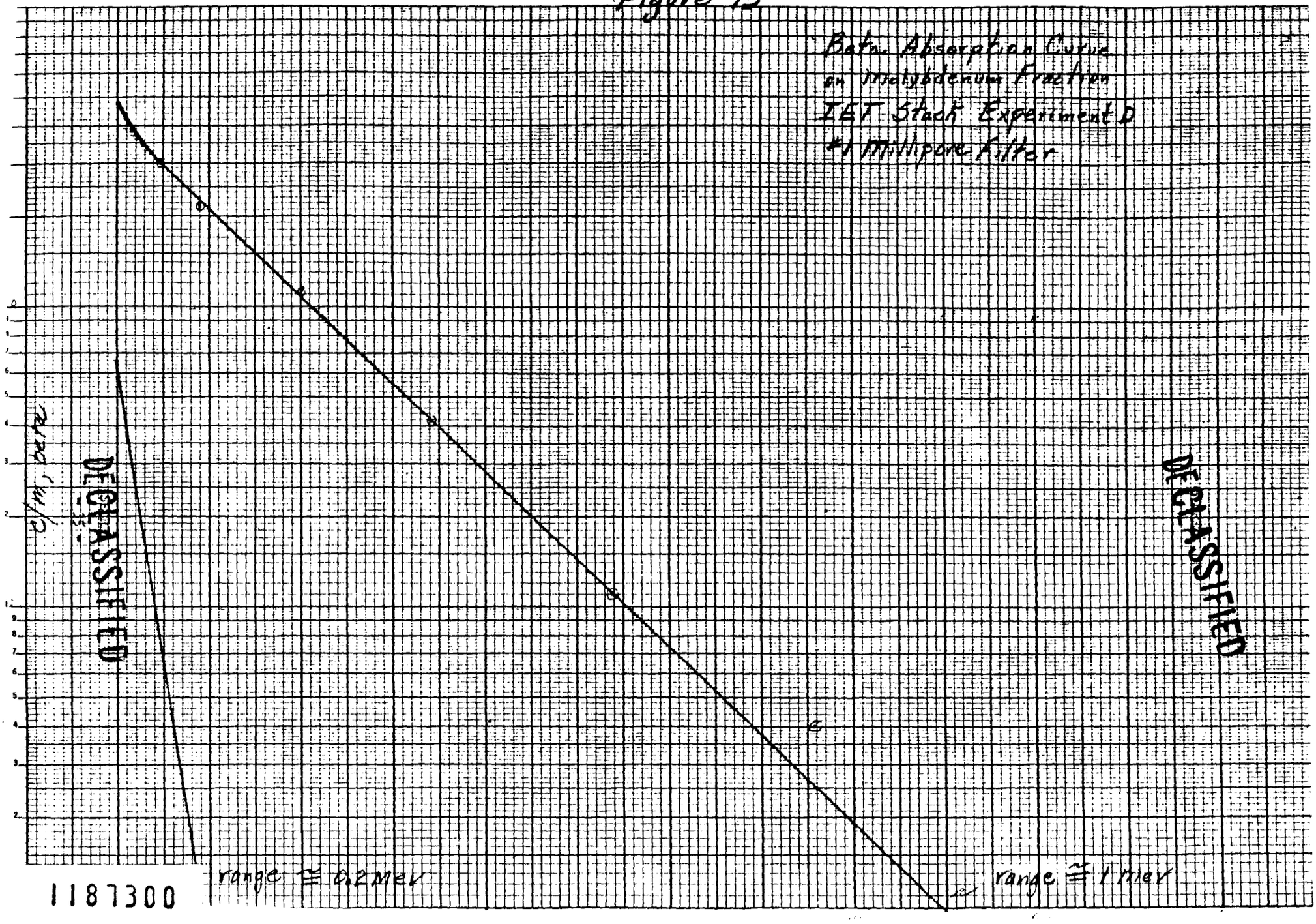
Gamma Decay Curves
Second Iodine Fraction
milked from #1 Millipore Filter
and #1 Scrubber Solution
Experiment C

6/4/56



1187299

Figure 15



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1187300