[redacted]

LA REPORT-272

C.3

[redacted]

May 14, 1945

This document contains 15 pages

CRITICAL MASS OF A WATER-TAMPED 49 SOLUTION

WORK DONE BY:

Group CMR-5

D. E. Carritt
C. S. Garner

Group G-1

H. K. Daghlian
B. T. Feld
H. Hammel
J. Hughes
S/Sgt. J. Kupferberg
T/4 H. Lehr
T/3 P. Long
P. Morrison
T/4 J. Osborne
L. Slotin

REPORT WRITTEN BY:

B. T. Feld

L. Slotin

[redacted]

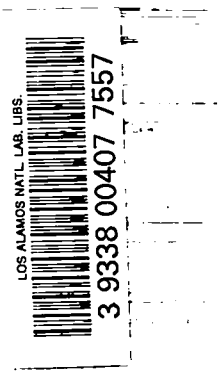
[redacted]

ABSTRACT

485 grams of 49 as $Pu(NO_3)_4$ in 1-molar nitric acid solution were tested for criticality in a water tamper. Tests were first made with 348 gms of 49 in various concentrations. It was found t at the optimum concentration was 43 ± 10 gms 49/liter, and that 348 gms, as nitrate ir water tamper, could not be made critical at any concentration. An absolute measuremen : of the multiplication of 348 gms of 49 as totravalent nitrate in 10 liters of solutio gave k = 0.87 ± 5 percent.

Starting with 348 gms in 10 liters of solution, 137 gms were added in 35-gm steps until a solution of 485 gms in 11.35 liters was obtained (485 gms was all the 49 nitrate available to the experiment). Criticality was not reached. From the inverse counting rate versus mass curves, using various detectors, the extrapolated critical mass of 49 nitrate solution in water tamper was estimated to be 606 ± 50 gms. Corrections for the nitrogen and other perturbations indicate a mass of 518 ± 50 gms for pure 49 in water solution with infinite water tamper.

The 485-gm solution was made critical by addition of a crudely stacked 2" layer of BeO bricks in the water around the outside of the bottle containing the solution.



CRITICAL MASS OF A WATER-TAMPED 49 SOLUTION

This experiment was designed to measure the critical mass of 49 in solution, in order to determine the amount of 49 that can be handled with safety. To contain the solution, four bottles with capacities 2.25, 3.6, 7.7, and 11.6 liters (labeled respectively A, B, C, and D) were used. These were commercial soda-glass bottles, as nearly equilateral cylinders as could be obtained to give the above range of capacities. To obtain an ideal spherical configuration would have required more shop time than this experiment was deemed to warrant. Each bottle was provided with a closure containing a re-entrant steel tube, which extended to the center of the bottle. This tube served as a channel for a neutron source as well as for a detecting chamber. In addition, the closure was provided with a capillary gas leak as an outlet for the decomposition gases given off from the 49 solution.

To provide for the possibility of breakage of the glass bottles, each bottle was enclosed in a water-tight lucite container. The bottles in their containers are shown in the accompanying photograph.

These bottles, when filled with 49 solution, were placed at the center of a cylindrical tank, 4-1/2 ft. in diameter and 4-1/2 ft. high. This tank could be filled with Los Alamos water to provide the tamping.

The monitoring instruments consisted of:

(1). A G.M. tube set half-way up the tank -- at approximately the same height as the center of the bottles -- on the inner periphery of the tank. This detector proved to be excessively sensitive to any source of gamma radiation in the building, and could therefore not be relied upon when the 25 water boiler or a 25 metal critical assembly was also in operation, nor for some time after the end of a period of operation of one of the critical assemblies.

(2). A cylindrical, boron-coated proportional counter, pointing at the center of the

bottle, its end about 6" to 8" from the outside of the smallest bottle. The efficiency of this instrument for counting neutrons was very high, so that care had to be exercised that no other neutron sources were present in the vicinity during the measurements.

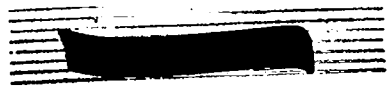
(3) A 4-inch square, flat 25 fission chamber was placed underneath the bottle position in contact with the bottom of the lucite case. When operating, this chamber proved to be the most reliable of all the detectors.

(4) A 1/2-inch-diameter cylindrical 25 fission chamber fitted the re-entrant tube. The source was attached to this chamber and separated from it by 2-1/2 inches of polythene. This chamber had rather small efficiency so that the data obtained from it did not have good statistical accuracy.

(5) A boron-coated chamber connected to a DC amplifier, which drove a recording milliammeter was provided for measurements on a critical assembly. Since criticality was not reached (except at the very end, by using a BeO tamper), this instrument did not take any part in the actual measurements. All the instruments were provided with water-tight casings and were used submerged in the water tamper. Their positions were unchanged during the series of measurements.

In order to check on the constancy of the instruments and to provide a means of correcting for changes in their efficiencies, a "standard" reading was taken before and after each measurement on the active material. This was done by placing the source at the center of the tank, filled with water, and measuring the counting rates of the detectors. The inverse counting rate listed in the table is then the standard reading divided by the reading with active material.

A view of the inside of the tank, with a bottle and some of the instruments in place, is shown in the second photograph.

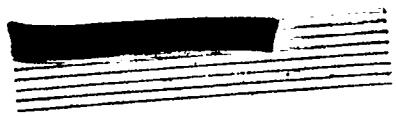


A total of 485 gms of 49 as +4 nitrate in one-molar nitric acid solution was available for the experiment. Analyses of the stock solutions involved indicated the presence of 1 g-atom of sulfur for each g-atom of plutonium, the sulfur originating from the sulfate coprecipitated with the plutonium peroxide which was evaporated down in nitric acid solution to give the nitrate stock. All amounts of 49 given in this report are based on the Group CMB-9 chemical titration method which is believed to be the most precise method available at this writing; the precision is of the order of 0.1 percent, and the amounts are believed accurate to 1 percent. The required transfers and dilutions with 0.996-molar nitric acid were effected by a vacuum transfer system. The various volumes and concentrations used are shown in Table I.

TABLE I

49 Solutions -- $Pu(NO_3)_4$ in 1 Normal Nitric Acid

Assembly	Concentration gms 49/liter	Total Volume Liters	Total gms 49	Bottle No.
I	127	2.25	287.0	A
II	99.93	3.482	348.0	B
III	46.4	7.5	348.0	C
IV	34.8	10.0	348.0	D
V	37.5	10.2	382.9	D
VI	40.2	10.4	417.8	D
VII	42.7	10.6	452.7	D
VIII	42.7	11.35	484.6	D



[redacted]

Determination of Optimum Concentration:

Starting with the solution containing 348 gms of 49 in 3.48 liters of solution (solution II) a series of measurements were made to determine the variation of reactivity with concentration. For these measurements, three different concentrations were used, corresponding to solutions II, III, and IV in Table I. The data are shown in Table II, which includes all the data of the experiment. In Fig. I the inverse counting rate is plotted against the concentration. From these curves it may be seen that the concentration corresponding to maximum multiplication is 43 ± 10 gms 49 per liter of solution. It is also evident from the curves that in the configuration used in the experiment, 348 gms of 49 nitrate cannot be made critical at any concentration.

An attempt was made to measure the multiplication of solution IV. A solution containing 130 gms of boric acid in 10 liters of water was substituted for solution IV in the tank filled with water. This solution was chosen to have the same absorption - in the same geometry - as solution IV without, at the same time, having any multiplication properties. From a comparison of counting rates of the various detectors for the boron solution with their counting rates for solution IV, a multiplication of 7.9 ± 20 percent was deduced for solution IV. Since the multiplication is equal to $1/(1-k)$, this corresponds to a $k = 0.87$. The estimated uncertainty of this measurement of k is around 5 percent.

Determination of Critical Mass:

To solution IV in bottle D, 35-gm "spikes" of 49 were added until 485 gms of 49 in 11.35 liters of solution were obtained (solution VIII). After each addition, the bottle was placed in the tank and the counting rates of the various detectors measured in their standard geometry. The data of these measurements are included in Table II.

[redacted]

TABLE II

[redacted]

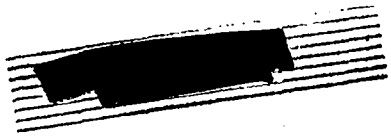
Assembly	G.M. Counting Rate			Boron Prop. Counting Rate			25 Flat Counting Rate			1/2" 25" Counting Rate		
	Std.	49 Counts	Std./counts	Std.	49 Counts	Std./counts	Std.	49 Counts	Std./counts	Std.	49 Counts	Std./counts
II	5.34	7.08	0.76	34.3	81.4	0.42	----	----	----	1.94	0.67	2.90
III	6.90	16.4	0.42	34.4	234	0.14	11.25	41.9	0.27	1.76	2.50	0.70
IV	7.65	14.3	0.54	5.63*	32.8*	0.17	11.3	42.0	0.27	1.73	3.01	0.57
Boron Solution	----	----	----	----	----	----	9.21	4.21	2.2	1.85	0.418	4.4
IV	5.36	13.0	.41	5.45	30.3	0.18	11.2	42.3	0.27	1.72	2.95	0.58
	5.21	15.1	0.35	6.14	38.5	0.16	11.3	50.5	0.22	1.75	3.25	0.54
I	4.38	17.5	0.25	6.10	5.07	0.12	11.5	61.1	0.19	1.79	3.71	0.48
II	4.45	20.6	0.22	6.30	67.9	0.093	11.6	75.0	0.16	1.86	4.36	0.43
II	4.43	25.9	0.17	6.36	76.9	0.083	11.6	91.0	0.13	1.91	6.33	0.3
II**	----	----	----	5.56	101.8	0.055	11.5	91.0	0.13	1.70	6.70	0.25

* Geometry of B prop. altered starting with this run

** Lucite case completely filled with water

[redacted]

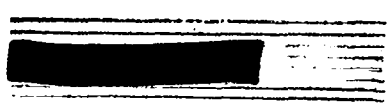
APPROVED FOR PUBLIC RELEASE



In Fig. 2 are plotted the inverse counting rates against the mass of 49 in solution. Through the points corresponding to each detector a smooth curve is drawn. These curves have been extrapolated to infinite counting rates by drawing a straight line tangent to the curves, starting at a point corresponding to 500 gms of 49. It is evident from the shapes of the curves that this method yields an underestimate of the extrapolated critical mass for all detectors except the 1/2" 25 chamber. The average value of the extrapolated critical mass, from the curves of Fig. 2, is 642 gms of 49.

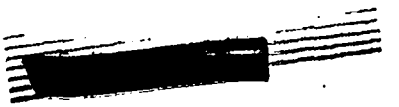
Fig. 3 is a plot of the inverse counting rate versus the reciprocal of the mass. The curves obtained are seen to be straight lines - as closely as can be judged from the data - for all but the 1/2" 25 chamber, and therefore lend themselves much more readily to a reasonable extrapolation to infinite counting rate. Extrapolating in the same manner as described for Fig. 2, the average value of the critical mass is found to be 678 gms of 49.

All of the experiments described above were performed in a configuration such that there was an air space of about 2.7 liters at the top of the lucite bottle container In order to determine the effect of this perturbation, the measurement of the last solution was repeated with this space filled with water. The results of this measurement are plotted in Figs. 2 and 3 as circled points. Since the P.M. tube was not operating well at the time of this measurement and since the flat 25 chamber, being located directly underneath the bottle, is particularly insensitive to filling the top space with water, only the boron proportional counter and the 1/2" 25 chamber measurements were used to estimate the effect of removing the perturbing air space. By estimating the distance of the circled points from the smooth curves drawn, the effect of removing the perturbation seems to correspond to 42 gms on the mass plot and to 65 gms on the reciprocal mass plot. This leads to 600 gms and 613 gms, respectively, for the



critical mass of a 49 nitrate solution completely tamped by water. Averaging the two, the figure of 606 ± 60 gms is obtained for the critical mass of a fully tamped nitrate solution in this geometrical configuration.

It is of interest to evaluate the critical mass of pure 49 in water solution in the absence of nitrogen, glass, boron, etc. These, as well as the hydrogen present in the solution, compete with the 49 for the absorption of the neutrons. Since the major part of the "foreign absorption" is due to the hydrogen - which must, in any event, be present in the solution - removal of the other absorbers does not effect a very great change in the amount of foreign absorption. Hence, to a sufficiently close approximation, the effect of foreign absorbers in the solution may be evaluated in the following way: To compensate for the addition or removal of foreign absorbers to a critical configuration (which it is wished to maintain as critical), 49 must be added or removed in amounts sufficient to maintain the same ratio of 49 to foreign material absorption. Thus, for a change in foreign absorption corresponding to ΔA_f, there corresponds a change in 49 mass, ΔM, such that $\Delta M/M = \Delta A_f/A_f$. In this way, the effects of removing the nitrogen from the solution, of the iron re-entrant tube and of the absorption of the glass bottle were estimated. The solution was considered to be a critical mass of 606 gms of 49 (as a compound containing 2N atoms and one S atom per atom of 49) in 1 normal nitric acid solution. The conventional thermal neutron absorption cross sections were used. The effect of the absorption of the bottle was considered to be 1/2 as much as if the absorber had been distributed uniformly throughout the solution, in agreement with measurements made on the 25 water boiler. The boron content of the water and glass was estimated by Group CM-9 to be less than 0.1 part per million for the water and 25 ppm for the glass. The concentrations of Li, Cd, Cl, and N_2 in the tamper water were measured by CM-9. They were 0.1, 0.002, 2 and 10 parts per million respectively. These were neglected. The calculated reductions



in critical mass are shown in Table III. The total reduction of mass for removing the foreign absorbers is 68 gms.

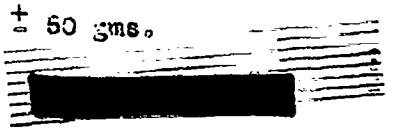
TABLE III

Reduction of M_0 Due to Removal of Foreign Absorption

Absorber	Effective No. of gm Atom	X-section(in b)	Prod.	ΔM
49	2.54			
H	1233	0.3	369.9	
N	16.1	1.75	28.2	41.0
S	2.54	0.53	1.34	1.9
Fe	1.0	2.5	2.5	3.6
Glass (Na_2O-CaO-7SiO_2)	3.62	3.2	11.6	16.9
B (in glass)	.0047	720	3.36	4.9
Total			417	68.3

In addition, it is estimated that the superior scattering properties of water as compared to glass corresponds to $\approx$ 20 gms, making ΔM = 88 gms and leaving for the critical mass of 49 in 11.5 liters of water solution the value of 518 gms. This corresponds to a concentration of 44 gms/liter, and is approximately the optimum concentration. No correction has been applied for the fact that the solution was contained in a cylindrical, rather than a spherical, shape.

Thus, the experiment yields for the best value of the critical mass of pure 49 in aqueous solution in an infinite water tamper, 518 $\pm$ 50 gms.



[redacted]

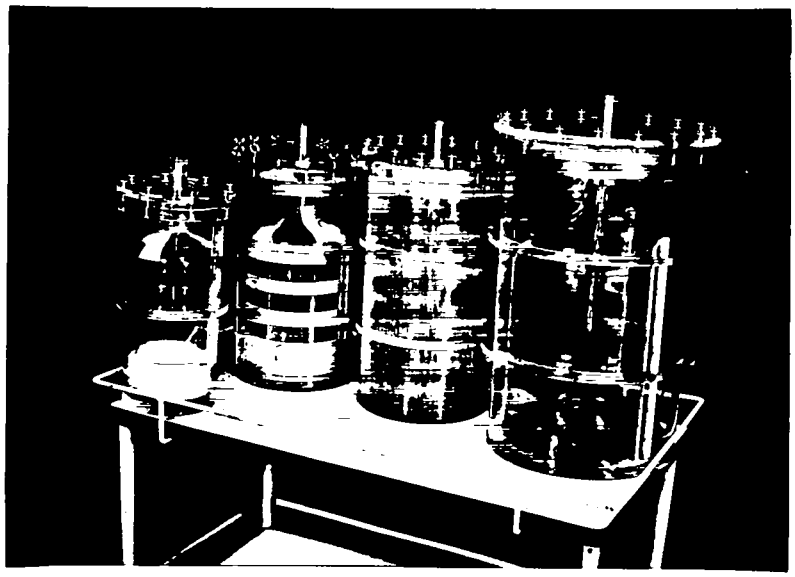
Criticality in a BeO Tamper:

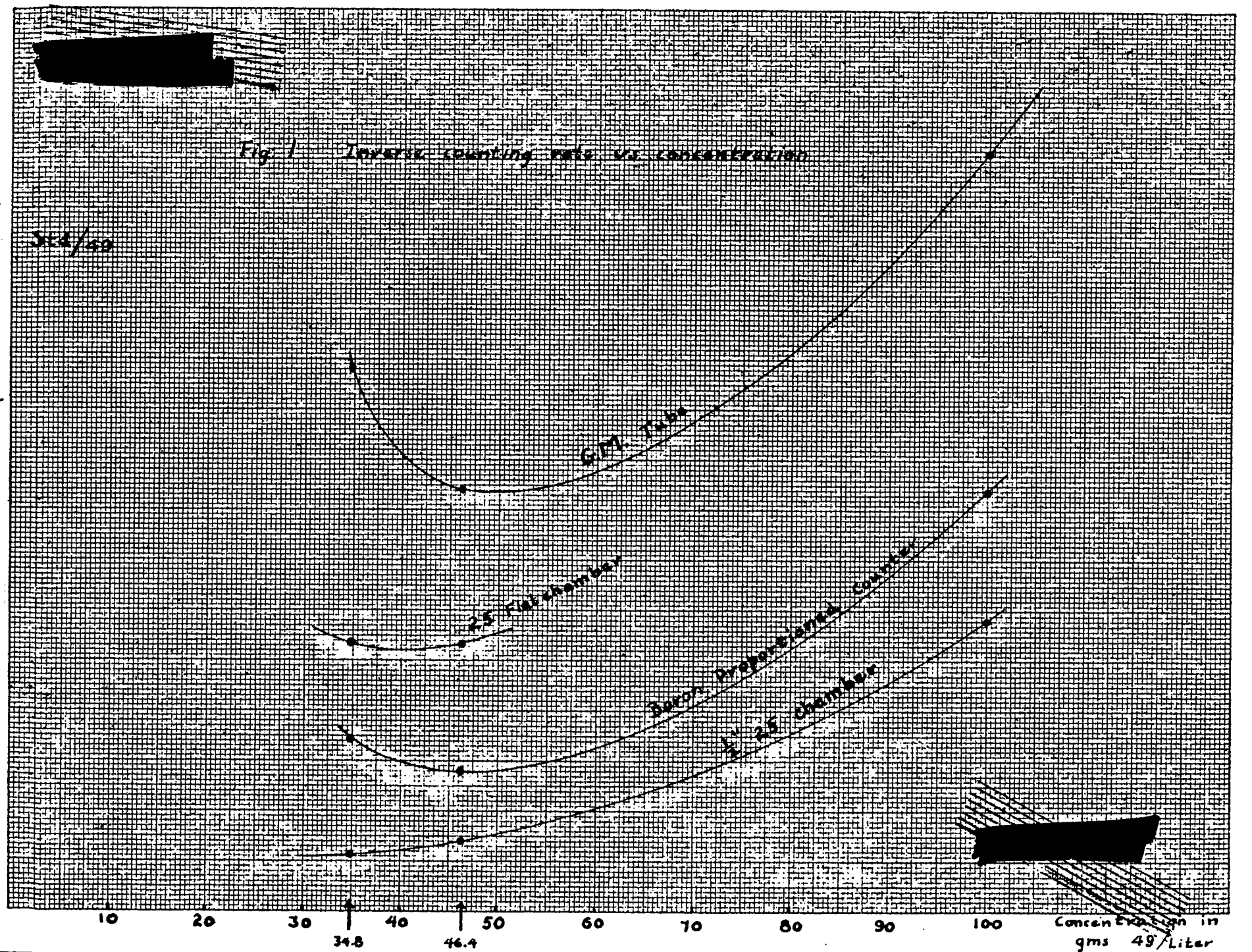
The solution containing 485 gms of 49 in 11.35 liters of solution was made to reach criticality by the addition of a roughly stacked 2" thickness of BeO blocks around the bottle in the water tamper. The blocks (1" x 4" x 4") of density 2.7 were stacked in the form of a 2" pedestal, on which the solution rested, and then in the form of a mosiac-like wall around the outside of the cylindrical container. The stacking of square blocks around a circular contour left, of necessity, considerable gap space, which was occupied by water. In addition, blocks were placed on the top of the solution containing bottle inside the lucite container.

It is evident that the critical mass of a 49 aqueous solution inside infinite BeO tamper is considerably less than 485 gms.

This first 49 chain-reacting assembly was operated super-critical for a short time. Its time behavior, response to control elements, etc.; was qualitatively like that familiar from 25 assemblies. No quantitative measurements were made.

[redacted]





APPROVED FOR PUBLIC RELEASE

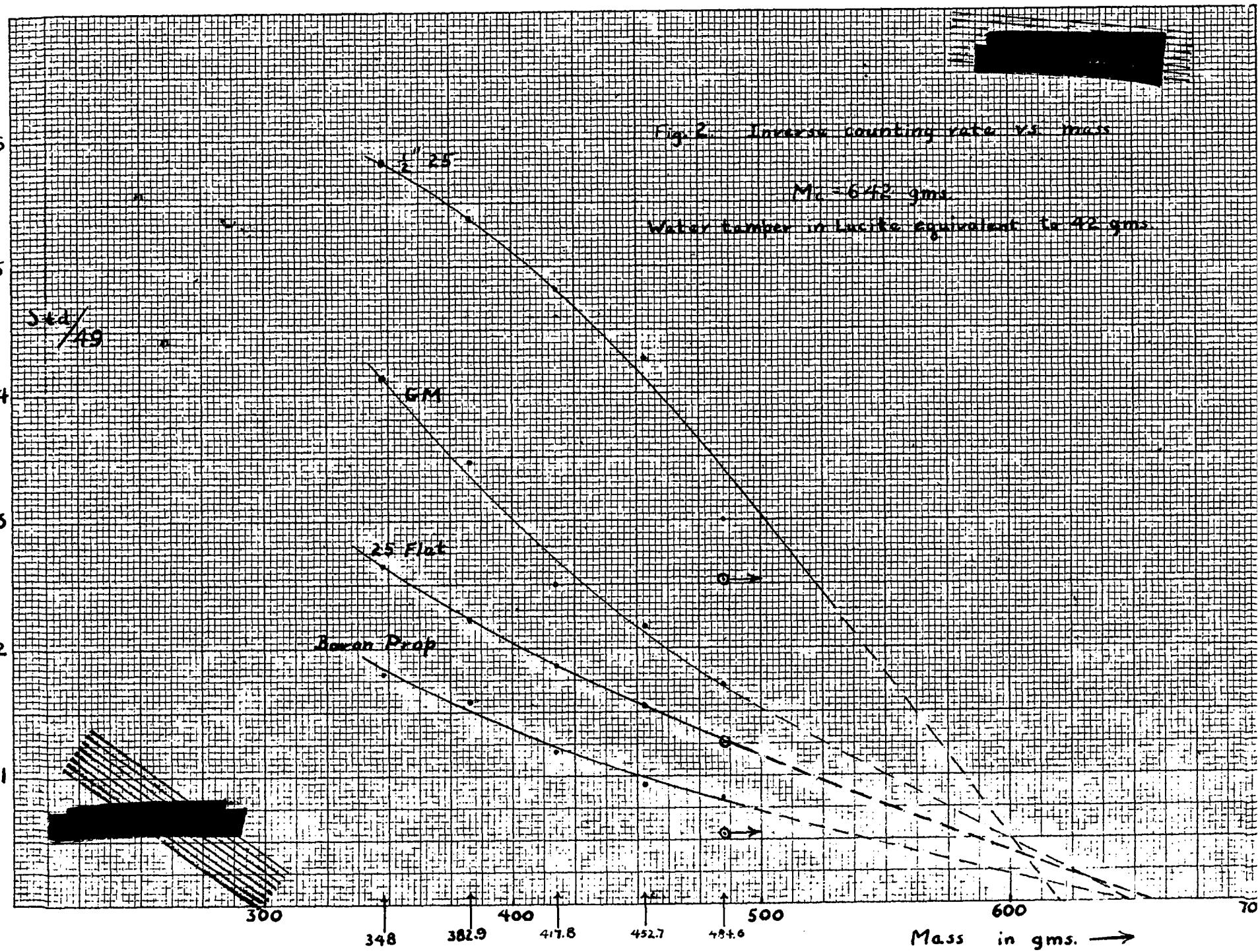
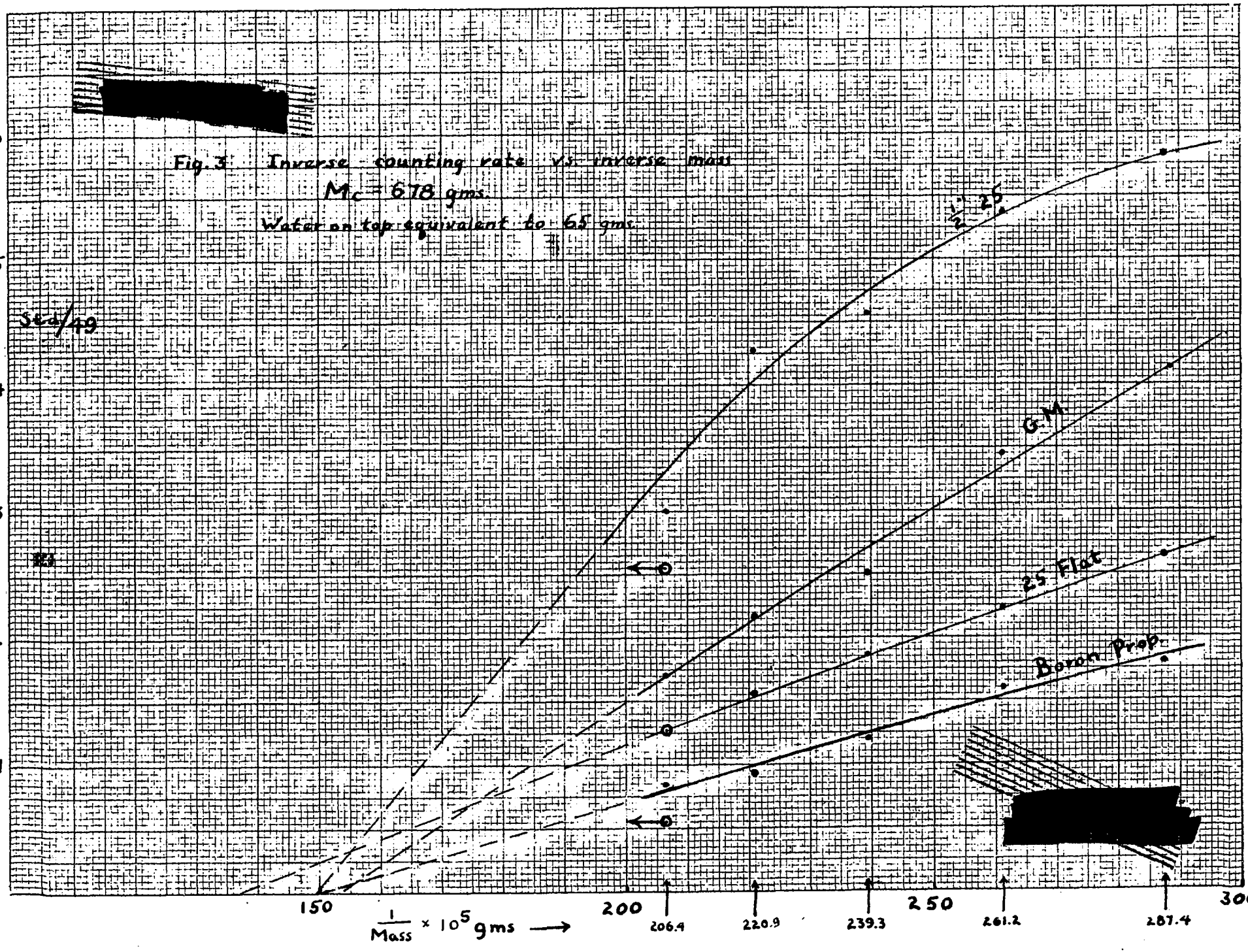


Fig. 2. Inverse counting rate vs. mass

M_c = 6½ gms.

Water tamper in Lucite equivalent to 42 gms.

APPROVED FOR PUBLIC RELEASE



APPROVED FOR PUBLIC RELEASE

UNCLASSIFIED

UNCLASSIFIED

[redacted]