REPORT CP- 400

CHAIN REACTION OF PURE FISSIONABLE MATERIALS IN SOLUTION

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January 1, 1943

ABSTRACT

The critical mass of 94-239 and the corresponding critical dimensions of homogeneous mixtures of 94-239 with various moderating media have been calculated as a function of the concentration of 94. A simple transformation makes the figures applicable to 92-235. The results are in essential agreement with preliminary estimates made independently by Oppenheimer and Serber. The problem of the stability of a chain reaction in solution and questions of protection are discussed.

CHAIN REACTION OF PURE FISSIONABLE MATERIALS IN SOLUTION

Introduction

In the chemical separation plant, 49 may occur in water solution or mixture. We must know in what quantities and concentrations the mixture would make a self sustaining chain reaction. Again, homogeneous mixtures of 49 (or 25) with suitable moderating media might be used as seeds in a production plant or as power producing units themselves. For these reasons, the critical mass of 49 and the critical dimensions of homogeneous mixtures of 49 with various moderating media have been calculated as a function of the concentration of 49.* The external boundary was treated as completely absorbing.

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*(Note: We are indebted to Oppenehimer for a letter from him received December 31, 1942 describing the results of similar calculations made independently by himself and Serber. We are taking the liberty to quote from this letter: "For a solution of 25 in water surrounded by a water case the optimal ratio of 25 absorption to hydrogen absorption is 2.9 and the mass of 25 is about 700 g. I regard this value as not too sure since some other calculations based on a slightly different treatment of the slowing down gave 450 g instead. A half a kg is a pretty good guess. For 49 the absorption ratio will be very closely the same and thus the mass may be about one-half as great. We have also looked at the boiler surrounded by ordinary uranium instead of water, but it seems doubtful whether this will reduce the amount of material needed. Because of the shorter slowing path some gain can be expected by using a saturated hydrocarbon or paraffin instead of water. ....100-g lots of 49 seemed] safe.......especially in view of]........the innocuous character of the phenomena should the reaction start during precipitation or centrifuging ....[We] have handled the problem... .....[using]....differential diffusion theory for the slow neutrons..... Otherwise the treatment is standard."

Since the function of the moderating medium is to slow the fission neutrons, it is apparent that the critical size will be of the order of the slowing down distance. The minimum concentration of 49 will be such that only one of the 2.2 neutrons per fission will be absorbed by 49, the thermal neutron absorption by 49 will be about equal to that by the moderator. The optimum concentration (minimum critical mass in a sphere) will be about three times this minimum.

For high concentrations of 49 the fast neutron reactions contribute appreciably. This effect has been treated as a small increase in V, the number of neutrons per fission. This of course underestimates the effect and overestimates the mass at very high concentration, where the contribution from fast fission is not small.

Except for the corrections at high concentrations, the results for 25 or 23 can be obtained by multiplying the masses and densities by 1.7 and 1 respectively.

The diffusion equation is written

$$\Delta n + \frac{kP_t - 1}{L^2} n = 0 \qquad (1)$$

where n is the neutron density, P_t is the probability of a neutron being slowed to thermal energies before leaking out, k is the reproduction factor for an infinite medium, and L is the thermal diffusion length. If the solution (for a sphere) is written $\frac{\sin Kr}{r}$ then

$$K^2L^2 = k\,P_t(K) - 1 \qquad (2)$$

Let the concentration of 49 be measured by

$$x = \frac{\text{thermal absorption by 49 per unit volume}}{\text{thermal absorption by moderator per unit volume}}$$

Then

$$L^2 = \frac{L_o^2}{(1 + x)\left(1 + \frac{1}{5}\frac{\sigma_{at}(M)}{\sigma_{st}(M)}x\right)} \qquad (3)$$

where L_o is the thermal diffusion length in the pure moderator and $\sigma_{at}(M)$ and $\sigma_{st}(M)$ are respectively the thermal absorption and thermal scattering cross-sections of the moderator. The second term in the denominator is a usually negligible correction to the total cross-section. It is assumed here that the presence of the 49 does not appreciably change the number of hydrogen nuclei per cm^3 of solution. Also

$$k = \frac{V_e\,x}{1 + x} \qquad (4)$$

where V_e is the effective number of neutrons per thermal fission of 49 and includes the multiplication of neutrons by fast fission. We have taken

$$V_e = V\left[1 + (V-1)\,x\,\frac{5.5\ \sigma_{at}(M)}{\xi\ \sigma_{sf}(M)}\frac{\sigma_{af}(49)}{\sigma_{at}(49)} - P_1(K)x\right] \qquad (5)$$

The fast fission was supposed to include the range where the fission cross-section is essentially constant, i.e., from 10,000 e.v. where the 1/v law meets the fast neutron cross-section of 1 up to fission energies. The number of collisions was then $\frac{5.5}{\xi}$. $P_1(K)$ is the average probability of escaping leakage for these energies. Then $\frac{\sigma_{at}(M)}{\sigma_{sf}(M)}\frac{\sigma_{af}(49)}{\sigma_{at}(49)}x$ gives the probability that a collision results in fission. Here t refers to thermal neutrons and f to fast neutrons, $\frac{\sigma_{at}(M)}{\sigma_{at}(49)}x$ is only a measure of the concentration of 49.

Substituting (3), (4), (5), in (2) we get. (6)

$$\frac{K^2L_o^2}{\left(1 + \frac{1}{5}\frac{\sigma_{at}(M)}{\sigma_{st}(M)}x\right)} = V\left[1 + (V-1)\,x\,\frac{5.5\,\sigma_{at}(M)\,\sigma_{af}(49)}{\xi\,\sigma_{sf}(M)\,\sigma_{at}(49)} - P_1(K)x\right] x\,P_t(K) - (1 + x)$$

Expanding the denominator on the left, we get a quadratic equation for x.

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$$\frac{\nu(\nu-1)\,.5\,\sigma_{at}(M)\,\sigma_{af}(49)}{\xi\,\sigma_{sf}(M)\,\sigma_{at}(49)}\,P_1(K)P_t(K)x^2 + \left[\nu P_t(K) - 1 + \frac{1}{3}\frac{\sigma_{at}(M)}{\sigma_{st}(M)}K^2L_0^2\right]x - \left[1+K^2L_0^2\right] = 0 \qquad (7)$$

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When Fermi's concept of neutron age applies in the slowing down procedure, so that the distribution of nascent thermal neutrons from a point source of fast neutrons can be written $e^{-\frac{r^2}{4\tau}}$, then $P_t(K) = e^{-K^2\tau}$ and $P_1(K) = e^{-K^2\tau_1}$ where τ_1 is the appropriate age of the fast neutrons making fast fission. In water, the distribution of energetic neutrons from a fission source is $\frac{1}{r^2}e^{-\frac{r}{l}}$. After the first few collisions the distribution spreads in an approximately Gaussian manner with an age τ from this lower energy to thermal energies. This consideration leads to $P_1(K) = \frac{\tan^{-1}Kl}{Kl}$ and $P_t(K) = \frac{\tan^{-1}Kl}{Kl}e^{-K^2\tau}$.

The equation (7) for x was solved for various values of K. Then the density of 49 which is proportional to x is known as a function of the critical dimensions of the mixture. For a sphere $R_s = \frac{\pi}{K}$, for a cylinder of infinite length $R_c = \frac{2.404}{K}$, and for a slab the thickness $T = \frac{\pi}{K}$.
This permits calculation of the critical mass, mass/cm, and mass/cm^2 of 49 respectively for sphere, cylinder, and slab as a function of the density of 49 or as a function of the dimensions. Except for the region of large density, the critical mass of 25 or 23 is greater than that of 49 by the factor $\frac{\sigma_{at}(49)}{\sigma_{at}(25)}$ or $\frac{\sigma_{at}(41)}{\sigma_{at}(23)}$, i.e., by 1.7 or 1 for the same dimensions of mixture.

We took $\nu = 2.2$, $\sigma_{af}(49) = 1 \times 10^{-24}\text{cm}^2$ (fast fission),

$\sigma_{at}(49) = 1090 \times 10^{-24}\text{cm}^2$.

Water: $\sigma_{at}(M) = 0.4 \times 10^{-24}\text{cm}^2$, $\xi\sigma_{sf}(M) = 7.6 \times 10^{-24}\text{cm}^2$,

$\sigma_{st}(M) = 43 \times 10^{-24}\text{cm}^2$, $l = 7$ cm, $L_0^2 = 8.3$ cm^2,

$\tau = 10.7$ cm^2.

These constants give for the ratio x the relation

$$7\times10^{-4}\left(\frac{\tan^{-1}Kl}{Kl}\right)^2 e^{-.22K^2l^2}x^2 + \left[2.2\,\frac{\tan^{-1}Kl}{Kl}e^{-.22K^2l^2} - 1 + 3.1\times10^{-4}K^2l^2\right]x - \left[1 + .17K^2l^2\right] = 0,$$

where the density of 49 in gm/cm^3 is

$$\rho_{49} = \frac{239 \times 4}{9 \times 1090}\,x = .00977x.$$

In fig. 1 is plotted the critical mass of 49 in a sphere of water as a function of the density of 49. Fig. 2 is the critical mass of 49 as a

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function of the radius of the sphere. Fig. 3 is the critical mass/cm of 49 as a function of the radius of the infinite cylinder container. Fig. 4 is the critical mass/cm^2 of 49 as a function of the thickness of the slab of mixture. Fig. 5 gives mass of 49 contours for spherical geometry on a ρ-R plot.

Heavy Water ρ assumed = $\frac{10}{9}$

$$\sigma_{at}(M) = .0057 \times 10^{-24} \qquad \xi\sigma_{sf}(M) = 2.21 \times 10^{-24}\ cm^2, \quad \sigma_{st}(M) = 7.53 \times 10^{-24}$$

$$L_0^2 = 1940\ cm^2, \qquad \tau = 130\ cm^2, \qquad \tau_1 = 50\ cm^2.$$

This choice of constants gives

$$3.45 \times 10^{-5}\, e^{-1.38 K^2 \tau}\, x^2 + \left[2.2\, e^{-K^2 \tau} - 1 + 2.27 \times 10^{-3} K^2 \tau\right] x - (1 + 15 K^2 \tau) = 0$$

$$\rho_{49}' = \frac{239 \times .0057}{9 \times 1090}\, x = 1.39 \times 10^{-4}\, x$$

Fig. 6 gives the critical mass of 49 as a function of its density in heavy water.

Graphite
(AGOT, ρ = 1.62)

$$\sigma_{at}(M) = .00493 \times 10^{-24}\ cm^2, \qquad \xi\sigma_{sf}(M) = .55 \times 10^{-24}\ cm^2.$$

$$\sigma_{st}(M) = 4.8 \times 10^{-24}\ cm^2, \quad L_0^2 = 2520\ cm^2, \quad \tau = 370\ cm^2, \quad \tau_1 = 133\ cm^2.$$

These values give

$$1.2 \times 10^{-4}\, e^{-1.36 K^2 \tau}\, x^2 + \left[2.2\, e^{-K^2 \tau} - 1 + 1.4 \times 10^{-3} K^2 \tau\right] x - \left[1 + 6.8 K^2 \tau\right] = 0$$

$$\rho_{49} = \frac{239 \times .00493 \times 1.62}{12 \times 1090}\, x = 1.46 \times 10^{-4}\, x.$$

Fig. 7 gives the critical mass of 49 as a function of its density in graphite.

Chain Reaction

An amount of 94 greater than the critical mass will not be chain reactive when diluted with a sufficiently large quantity of water. As the water evaporates away, a concentration will however be reached at which the effective multiplication factor will exceed unity by a small amount. The neutron density will thereupon increase from a negligible value to a figure sufficient to produce a very appreciable liberation of heat. The recoiling fission fragments will impart their energy to the

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water in a time of the order of magnitude of 10^{-11} sec., short in comparison with the time of reproduction of one generation, $\sim 10^{-4}$ sec. Consequently the rate of rise of the temperature of the water will respond at once to the level of neutron density. As the temperature rises, the density of the water decreases. The leakage of neutrons is consequently increased. The effective multiplication factor drops to unity. The neutron density becomes stationary for a short interval. Heat is at this moment being produced at its maximum rate. The multiplication factor continues to fall and drops appreciably below unity. The neutron density dies off only after a finite time interval. An additional temperature rise occurs on this account. At the end of the first act the temperature of the water has risen by a finite amount, the multiplication factor is less than unity by a finite amount, and the neutron density is again negligible.

The water will begin to cool. The density will increase to the point where there will be a second, smaller, surge of activity, a reheating of the solution, and a repetition of the first cycle of events. The second cycle will be followed by a third, a fourth, etc., each of decreasing amplitude. Finally the solution will settle down to a steady state. For the given composition there will be a critical density for which the effective multiplication factor will be exactly unity. Corresponding to this density will be a critical temperature at which the solution will maintain itself. Whatever heat is lost owing to the temperature difference between water and surroundings will constantly be made up by the nuclear reaction. The solution acts as a thermostat. Well insulated, it produces a negligible amount of heat. Penetrated by ducts arranged to give the maximum possible transfer of heat to a suitable cooling fluid, the solution acts as a power plant whose output varies over a range whose limit is set entirely by the possibilities for heat transfer. In order that the working temperature should remain constant in such a low temperature plant, it is naturally necessary that further evaporation of the water should be prevented and consequently that the solution should be enclosed.

When the solution is not enclosed, further evaporation will take place as time goes on. The concentration of 94 will slowly increase, the critical temperature will rise. More heat will be transferred to the surroundings per unit of time. Consequently the nuclear reaction will have to proceed at a higher rate in order to maintain the temperature at the critical level.

After a time determined entirely by the rate of evaporation, the solution will reach the boiling point and bubbles will form. The rate of evaporation will thereby be greatly increased. In contrast to the previously discussed stage of events, where the rate of evaporation, and consequently the rate of temperature rise, depended upon such external factors as air velocity and humidity, the present occurences will be more nearly independent of conditions outside the solution, and will run through their course in a much shorter time. The nuclear reaction will be maintained at a level to balance the losses of heat through evaporation by adjustment of the density, as before. How-

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however, the density will be controlled, not by the temperature, which remains constant, but by the proportion of bubbles to liquid. Let the solution lie at the bottom of a large vessel. If the opening at the top is large, the rate of loss of heat through evaporation will be great. The solution will quickly boil down. The concentration of 94 will rise to a level where the solution is no longer chain reactive and the boiling will stop, the solution cool off. If the opening at the top of the container is small, the same chain of events will require a longer stretch of time. In both cases the total liberation of nuclear energy will be the same and will equal the latent heat of evaporation of the excess water. To liberate a really large quantity of heat, the system will be designed to avoid any net loss of water. Fresh water may be pumped in at the bottom and steam removed at the top of an otherwise closed container. Such a boiler is safe, for operation will cease shortly after the flow water is cut off. Even if the steam outlet is blocked by mistake, an explosion will not necessarily occur. Boiling indeed will soon cease but evaporation will build up the vapor pressure of water in the free space at the upper part of the container. The resulting increase in concentration of the solution will suffice to stop the chain reaction before the pressure reaches a dangerously high level, provided that the vessel is sufficiently large.

Quantitative relationships between critical mass and density and induced radioactivity will supplement the foregoing qualitative picture of a chain reaction in an aqueous solution of 94. When boiling is possible, the solution by frothing will adjust its density so that the effective factor of multiplication is very close to unity for the given relative concentrations of 94 and moderator. A decrease of density by the factor f from the normal value ρ_0 to the new value of ρ_0/f will increase all mean free paths by the same factor f. The new solution will have the same leakage, and therefore the same effective multiplication factor, as the system of normal density, if all its linear dimensions are increased by the factor f. The critical mass required for a chain reaction in the frothing solution will therefore be greater than that for the normal solution by the factor $(1/f)f^3 = f^2$, provided that the boiling expands all the dimensions of the mass by the same factor. Actually we will be more interested in the case where the liquid lies in a rigid tank whose diameter is much greater than the depth of the solution. The neutronic leakage will be independent in first approximation of the diameter. Consequently a solution whose density is low by the factor f will only have to have a depth greater by the same factor f if it is to have the same effective multiplication factor as a solution of normal density. The critical masses of the two solutions will be related by the factor f/f or in other words will be equal. Consequently the stabilizing action of the frothing is not apparent in the first approximation. Actually a shallow solution of normal density, having appreciably more than the critical mass, will have to boil up until its depth is comparable with the diameter of the tank before the neutron density reaches equilibrium. Introduce the symbols

- k, multiplication factor
- $\mathcal{A}$, effective migration area
- D, diameter of tank
- h, h', height of normal, of boiling solution
- r = h'/h, critical mass for boiling solution relative to normal solution.

Then the condition for a stable reaction gives the condition

$$(4.8096/D)^2 + (\pi/h)^2 = (k - 1)/\mathcal{L}$$

for the normal solution, and for the frothing solution

$$(4.8096/D)^2 + (\pi/h')^2 = (k - 1)/\mathcal{L} f^2$$

Multiplying through the first equation by $(h'D/4.8096\ f)^2$, the second by $(h'D/4.8096)^2$, equating left hand numbers and solving for h', we find

$$h' = \left\{ (r^2 - 1)(\pi D/4.8096)^2 + r^2h^2 \right\}^{\frac{1}{2}}$$

as relation connecting effective height of the frothing solution with the ratio, r, of its mass to the mass of a solution of normal density and the same concentration ratio, which is sufficient in amount just to be on the verge of reacting when lying in a vessel of the diameter D filled to the height h. When D is very large in comparison with h, and the factor, r, of excess over the critical mass for normal density is considerable, the solution will boil up to a height, h', approximately equal to 0.654 rD, provided that the tank is not closed.

When free boiling is possible, the total energy released by nuclear fission will be very little more than that required to evaporate off the excess water and raise the concentration of 94 to a stable value. Under these conditions the integrated dosage due to exposure to gamma rays can easily be estimated in order of magnitude. Of the energy release of ∽ 200 Mev per fission, an amount of the order of 15 Mev will be given out in the form of gamma radiation. For every liter of water boiled away, the integrated dosage in roentgen units at an unshielded point at a distance of x centimeters will be given by the product of the following factors:

- 1000 grams per liter
- 540 calories per gm, latent heat of evaporation
- 4.18×10^7 ergs/calorie
- 15/200 fraction of energy in gamma rays
- $1/4\pi x^2$ fraction passing through 1 cm^2 at x cm
- 3.5×10^{-5} fraction of energy of one quantum converted into electronic energy in 1 centimeter (factor nearly independent of energy from 2 Mev to 70,000 ev).
- 300/32 e.s.u. of ion pairs produced by 1 erg when 32 ev will make one ion pair.

Multiplication gives $4.4 \times 10^7/(x \text{ in cm})^2$ e.s.u. of charge per cm^3 of normal air at a distance of x cm, per liter of water boiled away. An unshielded individual 10 meters from a solution from which 1 liter of water boils away without warning will get an integrated dosage of 44 roentgen units. This figure in itself is not considered dangerous. Absorption of gamma rays in the solution will reduce the effect. However, neutrons escaping from the unit will add somewhat to the dosage.

The chance for an energetic neutron to escape from the solution will be of the order

$$1 - (K\ell)^{-1} \text{ arc tan } K\ell \doteq (K\ell)^{2/3}$$

$$= (\pi \ell / \text{thickness of slab})^{2/3},$$

in terms of the notations used above. With a value of 7 cm for the quantity ℓ, and with a depth of solution corresponding to the critical region in Fig. 4, we estimate that 1/20 of the fast secondary neutrons will escape with an average evergy of the general order of magnitude of 1 Mev. To compute the dosage, we shall take the physiological effects of gamma rays and neutrons to be equal if equal amounts of energy are liberated per cm^3 of body tissue -- or water -- an assumption that may be in error by as much as a factor of 3. Then when the energy of fission evaporates one liter of water, the effect of the neutrons translated into roentgen units will be given roughly by the product of the following factors:

1000 gms per liter

540 calories per gm

4.18×10^7 ergs/calorie

$1/20 \; 2.2 \; \dfrac{1 \text{ Mev}}{200 \text{ Mev}}$, fraction of energy carried by energetic escaping neutrons ($\nu = 2.2$)

$1/4 \pi x^2$, fraction striking cm^2 at x cm.

$2.5 \times 10^{-4} cm^{-1} = (\dfrac{2}{16+2} \; 6.02 \times 10^{23} gm^{-1})(0.001293 \; gm/cm^3) \cdot (3 \times 10^{-24} cm^2)$

approximately average fraction of energy converted into energy of recoil protons per cm of path of a neutron through water vapor of same density as standard air.

300/32 e.s.u. of ion pairs produced by one erg when 32 ev will make one ion pair.

Multiplication gives $2.4 \times 10^6/(x \text{ in cm})^2$ as our order of magnitude estimate for the number of equivalent roentgens of dosage produced by the neutrons emitted during the boiling away of one liter of water. The effect of the slow neutrons should not increase the result by a factor more than two. We conclude that the effect of the neutrons is less important than that of the gamma rays.

The radioactive gases carried out by the boiling process form a third source of danger. However, the amount of such activity in solution is very much less in the present instance than it is in the case of uranium dissolved up after a long irradiation in a plant for the rapid production of 94. Consequently the problem of protection can be solved by known and apparently quite adequate precautions.

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FIG. 1

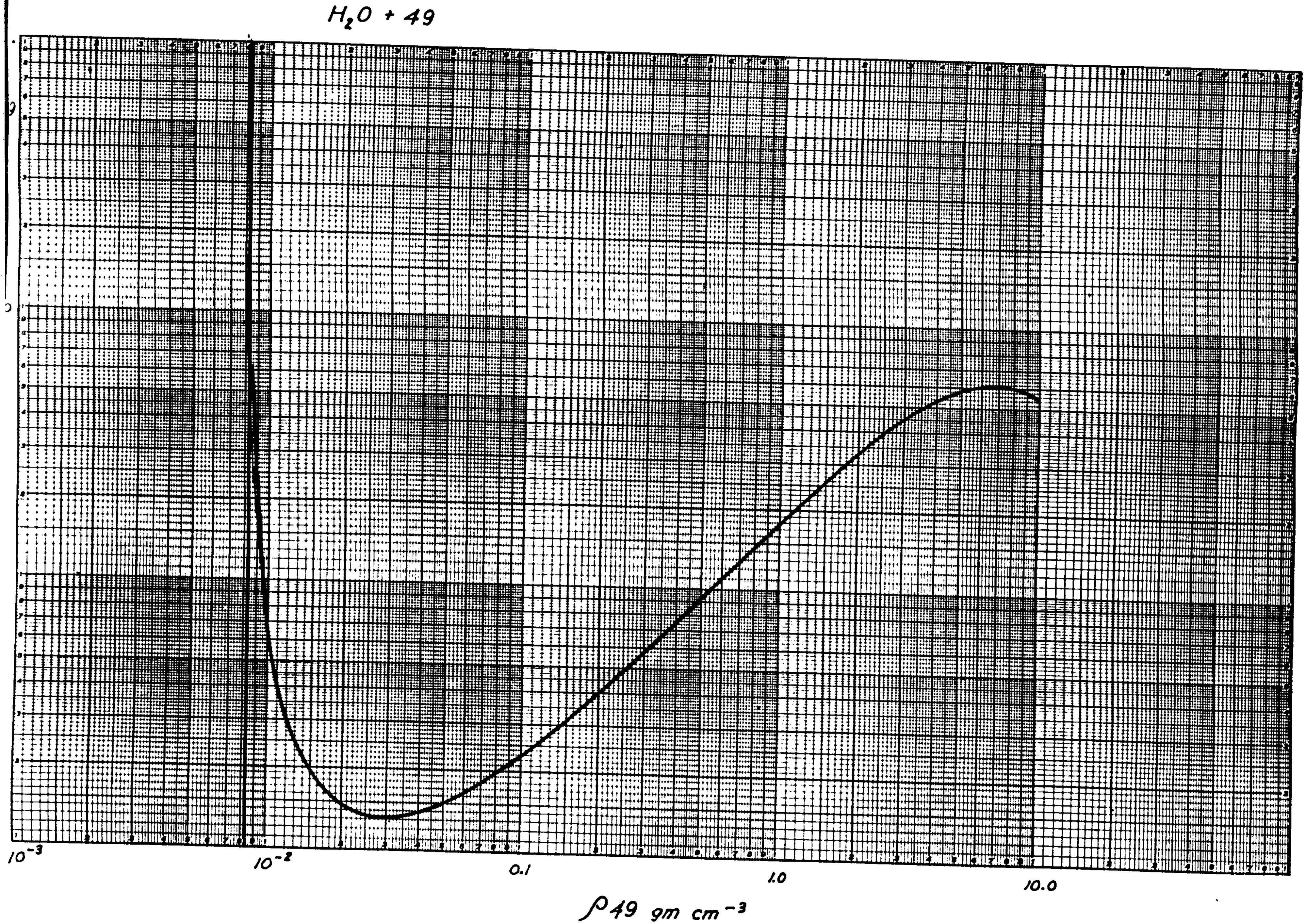

FIG. 2

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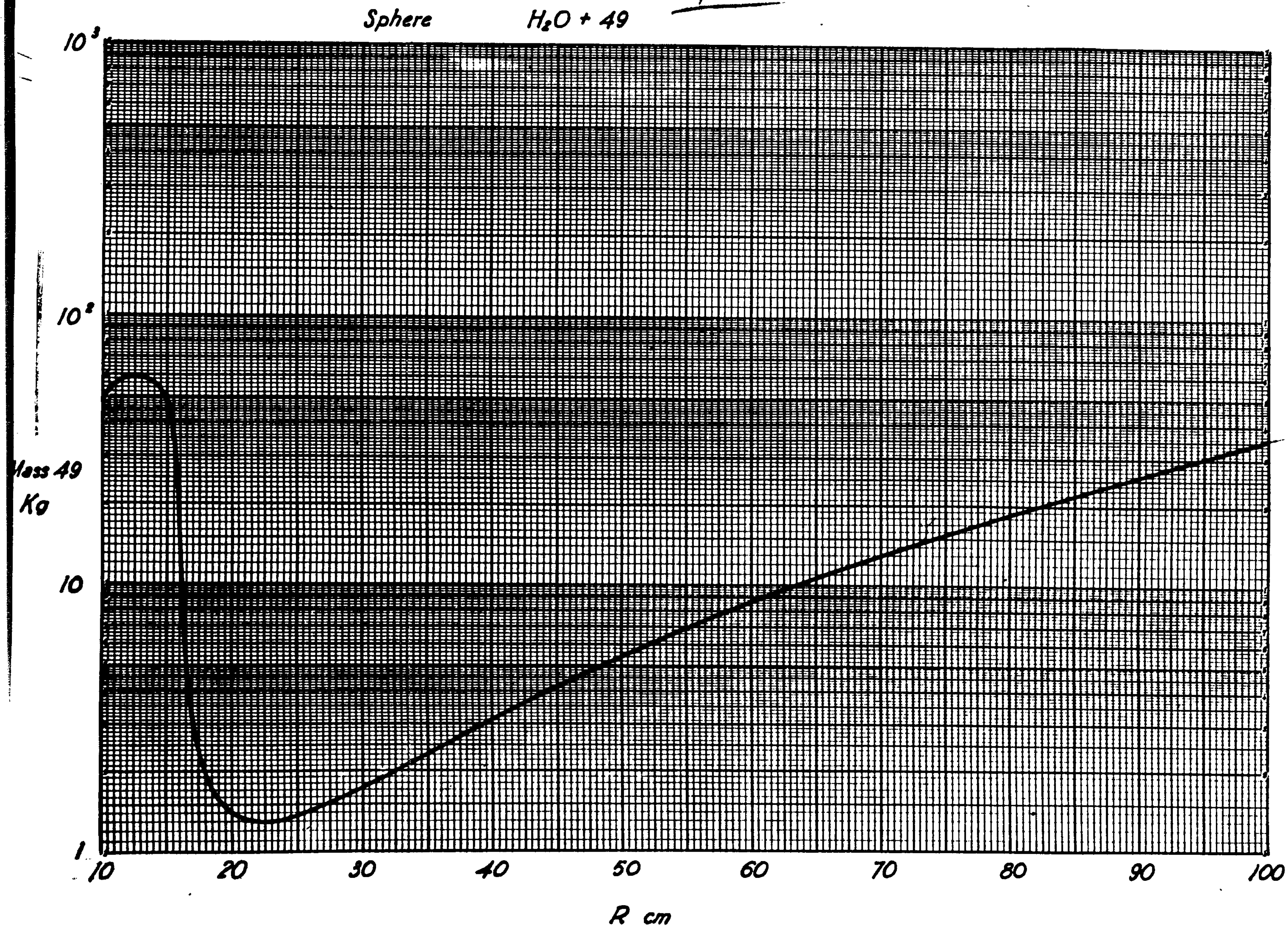


Sphere
H_2O + 49
10^3
10^2
10
1
Mass 49
Kg
10
20
30
40
50
60
70
80
90
100
R cm

FIG. 3

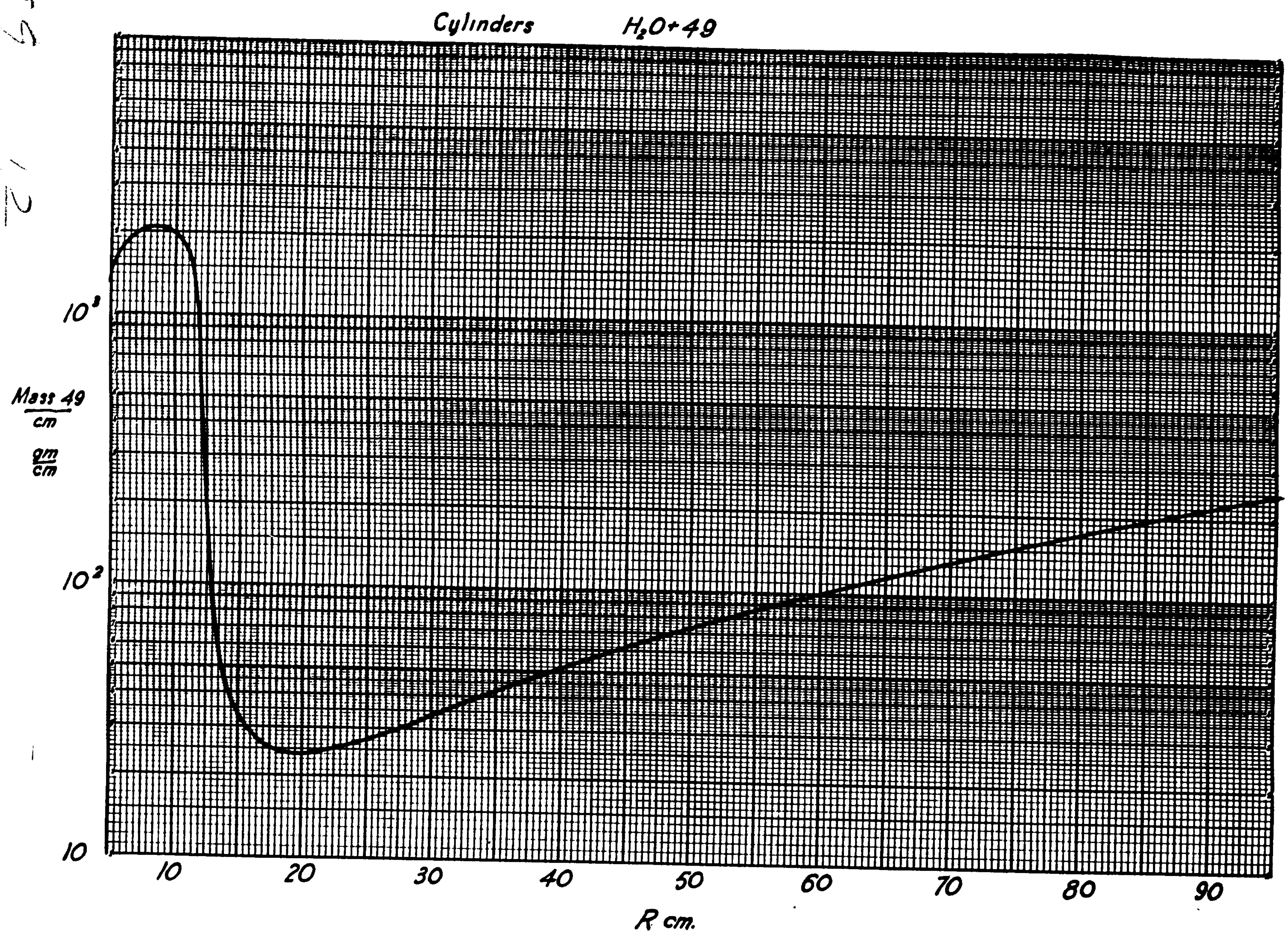
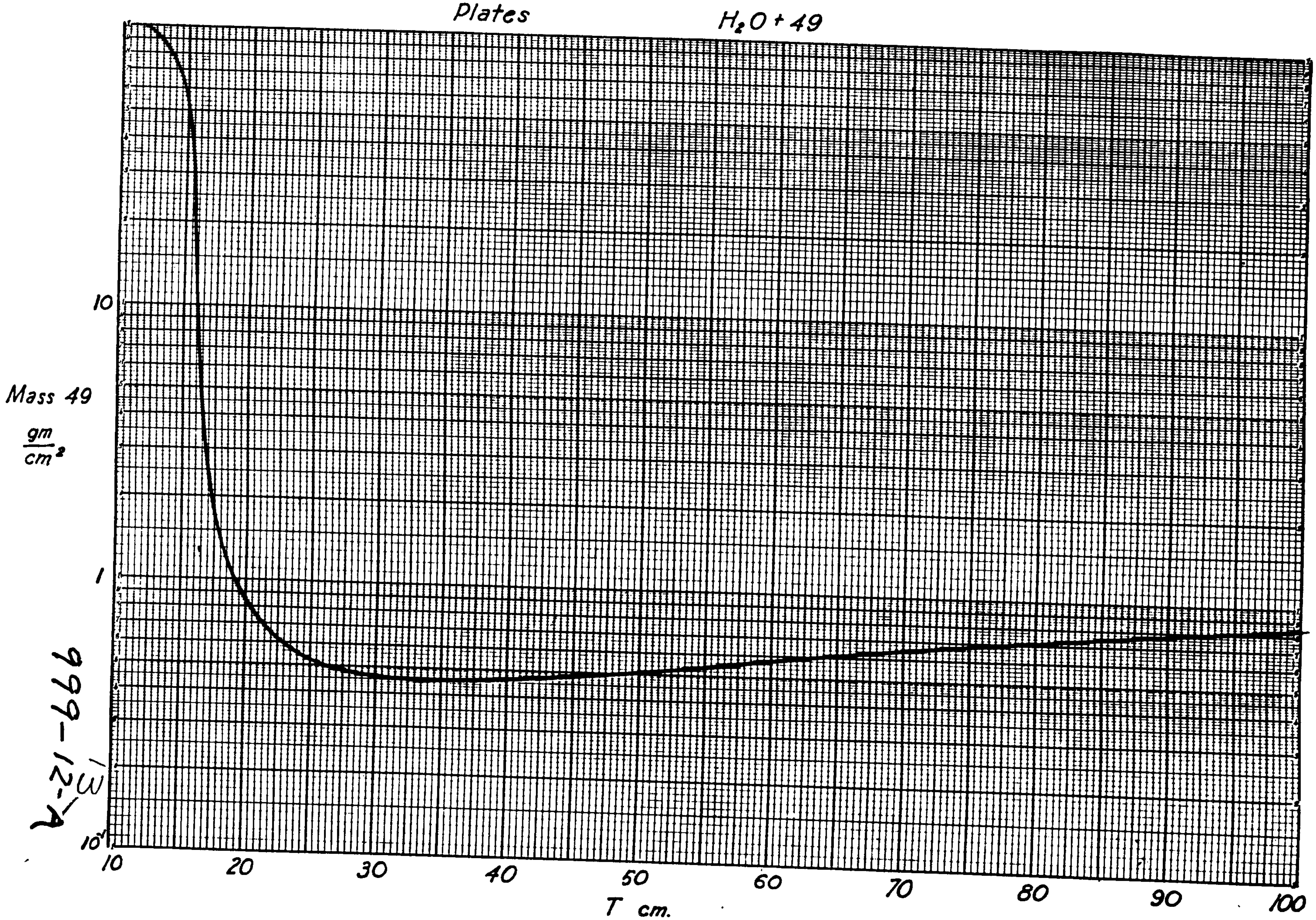

Cylinders
H_2O+49
10^3
10^2
10
Mass 49 / cm
gm / cm
10
20
30
40
50
60
70
80
90
R cm.

FIG. 4



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Course of chain reaction in water

Contours of constant mass of 49

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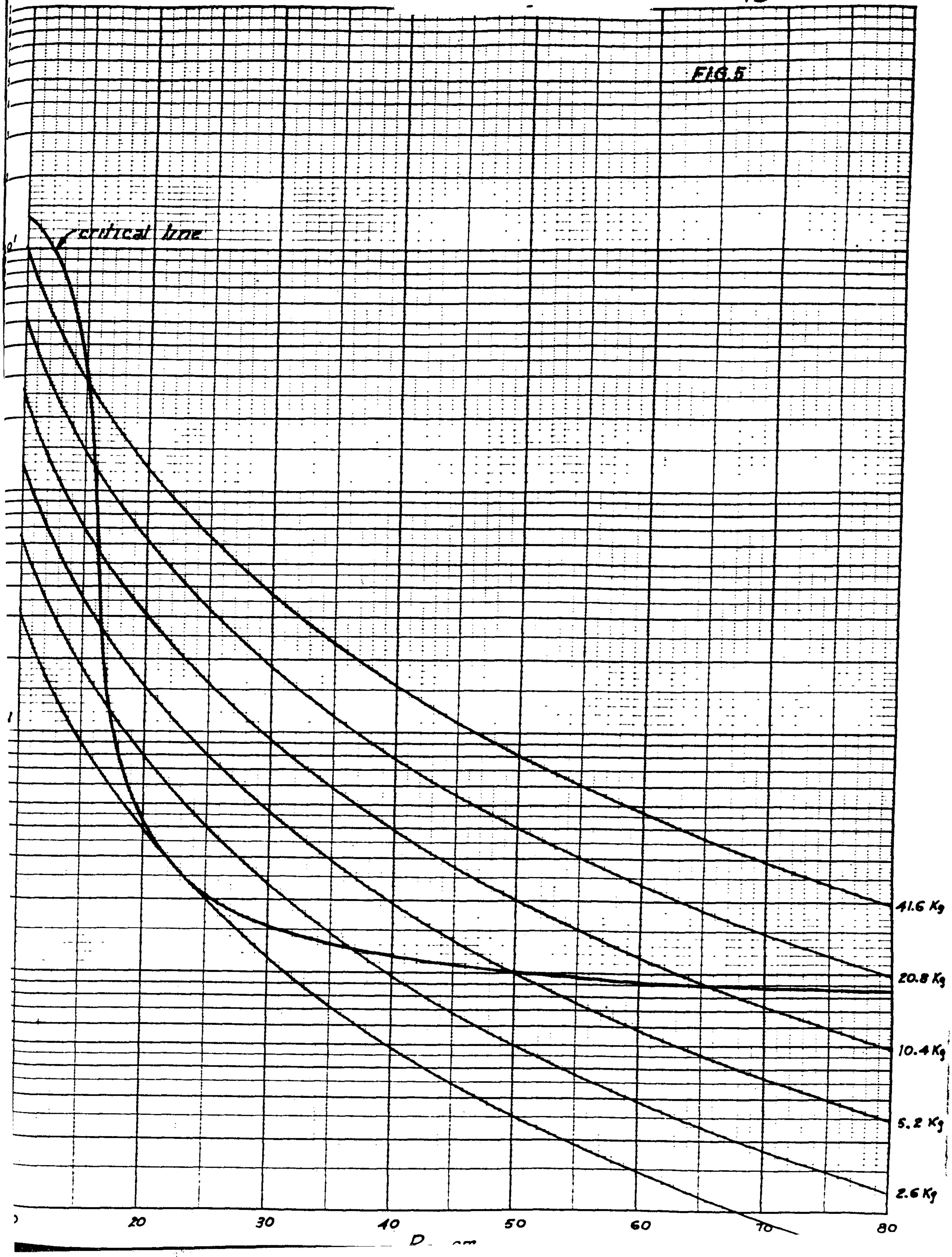

FIG. 5

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FIG. 6

$D_2O + 49$

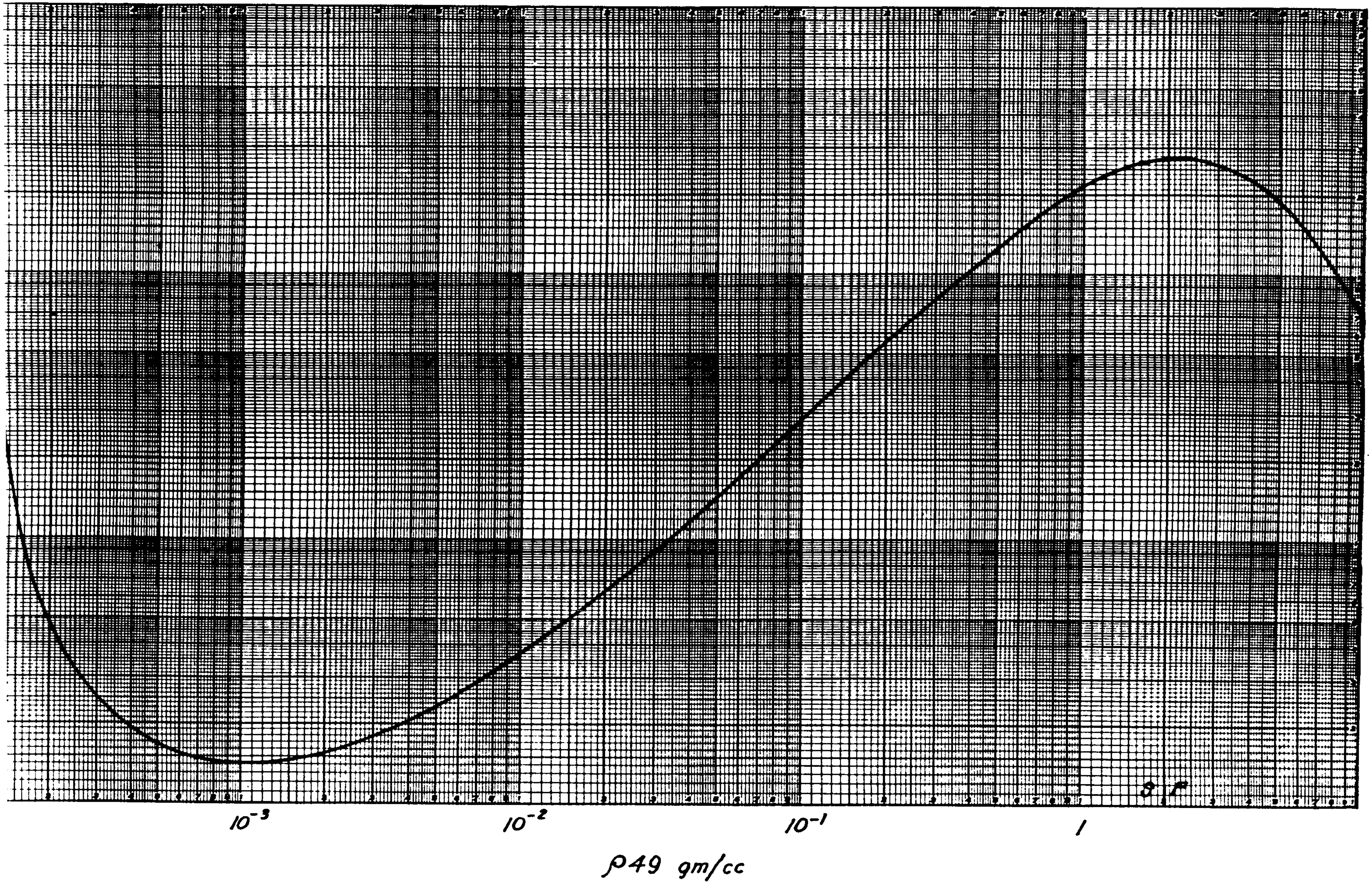

FIG.7