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Informal Report

The Antimony-Beryllium Neutron Source for the LASL Water Boiler Reactor

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Avery M. Gage

CONTENTS

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ABSTRACT

An Sb-Be neutron source was designed for two purposes: to produce a larger neutron flux in the sphere of the Water Boiler Reactor during shut-down than that produced by the existing Ra-Be source, and to be a less hazardous source than the Ra-Be. The equilibrium strength of the Sb-Be source was calculated to be 6.2 x 10^6 neutrons/sec after 21 weeks of reactor operations. The sphere flux caused by the Sb-Be source, at equilibrium strength, was calculated to be 33,600 neutrons/cm^2-sec. Measurements of the sphere flux with indium foils and a fission counter showed that the flux caused by the Ra-Be source was 707 neutrons/cm^2-sec and that the equilibrium flux caused by the Sb-Be source was 13,000 neutrons/cm^2-sec, which was attained in about 20 weeks.

I. INTRODUCTION

This report contains the design of an Sb-Be neutron source for the LASL Water Boiler reactor, and both calculated and measured values of the neutron flux in the reactor sphere caused by the source. The instruments and techniques used in measuring the neutron flux are described. The Sb source was designed by, and the activation equation developed by R. E. Carter. The calculation and measurements of the neutron flux and the analysis of the data were done by the writer.

The Ra-Be source, previously used, was suspected of leaking radon, so a less hazardous replacement was sought. The Sb-Be neutron source was selected since it was not an alpha emitter and the active material would not migrate out of a hole in a container. In addition, the 60 day half-life of the Sb^{124} was convenient for source build-up and strength after reasonable shut-downs. Beryllium alone was not selected since its neutron emmission would be too variable, depending on previous reactor operation and the resultant gamma ray activity of the sphere. Decaying fission fragments are the main source of the sphere gamma rays, when the reactor is shut-down.

An additional requirement for the replacement source was that it produce a sphere neutron flux about 10 to 20 times that produced by the Ra-Be source. The higher source strength would simplify monitoring the neutron flux at shut-down and start-up, when the reactor power level is less than several milliwatts.

Measurements of the flux in the sphere, with the reactor shut-down, were made to check on the calculations and to ensure that the sphere flux resulting from the Sb source was greater than that from the Ra source.

II. THE Sb-Be PHOTONEUTRON SOURCE

A. Neutrons are produced by an Sb-Be photoneutron source as a result of the following reactions:

1. The two stable isotopes of Sb can capture slow neutrons in (n,γ) reactions to produce two radioactive isotopes:

$$Sb^{121} + n \rightarrow Sb^{122} + \gamma$$
$$Sb^{123} + n \rightarrow Sb^{124} + \gamma$$

The radioactive isotopes decay with the emission of beta particles and gamma rays:

$Sb^{122} \rightarrow Te^{122} + \beta + \gamma$, with a half-life of 2.75 days.

$Sb^{124} \rightarrow Te^{124} + \beta + \gamma$, with a half-life of 60 days.

2. Gamma rays or photons from Sb^{122} and Sb^{124}, with energies above 1.67 Mev, can interact with Be to produce neutrons:

$$Be^{9} + \gamma \rightarrow Be^{8} + n$$

The constants for these reactions are given in Section I, below.

B. The activity of the Sb, and hence the neutron strength, must be built up and maintained by exposure of the source to a slow neutron flux. This is done by installing the source in the reactor at some point having a reasonably high flux.

C. The usual design of an Sb-Be source is to surround Sb with Be to optimise the Be(γ,n) reaction. The layer of Be outside the Sb does not reduce the neutron flux necessary to activate the Sb, since the neutron absorption cross section for Be is very small.

III. SOURCE LOCATION AND DESIGN

A. Preliminary estimates of the neutron strength of the Sb source indicated that a one-inch diameter source in the SE Vertical Port (Figs. 1 and 2) would not produce the required increase in sphere flux. A source location closer to the sphere than the SE Vertical Port was required for two reasons. The source activity varies with the reactor neutron flux, which is larger near the sphere, and more source neutrons enter the sphere the closer the source is to the sphere.

B. An additional requirement was that none of the experimental ports should be blocked by the source. The largest available port leading to the sphere was in the North Thermal Column just below the North center line. The location and size of this port are shown in Figs. 2 and 3. The source could not be larger than the cylindrical end of the removable graphite block that contacts the sphere. This block is indicated on Figs. 2 and 3. A graphite source holder was designed from the dimension of the block as shown in Fig. 4. The exterior dimensions of the source were determined by the cylindrical end of the holder.

C. The interior arrangement of the Sb source was determined from the following considerations:

1. The Be should surround the Sb to give the longest practical path for gamma absorptions.

2. The Sb should not be much thicker than 1 or 2 cm. to avoid self shielding for neutron absorption and gamma emission.

3. The Be, Sb, and container should be easy to fabicate. The design of the source is shown in Fig. 5.

D. <u>Antimony</u>

The four Sb rods were cast oversize and machined to an average length and diameter of 1.497 in. and 0.246 in. respectively and average volume of 1.171 cm^3. The mass of each was 7.60 gms and the density 6.49 gms/cm^3. The Sb used was of analytical reagent grade.

1. The mean free path of thermal neutrons in Sb was calculated to ensure that the Sb was not so thick that the center of the rods would not be activated:

$$\lambda = \frac{1}{\Sigma a}$$

$$\Sigma_a = \frac{\rho Na}{A}$$

$$\Sigma_a = 0.177 \text{ cm}^{-1}$$

$$\lambda = 5.6 \text{ cm}$$

$$\frac{\phi}{\phi_o} = e^{-\Sigma a^x} = 0.89$$

Thus about 11 percent of the thermal neutrons passing diametrically through an Sb rod will be absorbed.

2. The total number of Sb atoms N(t) in the source and the number of atoms of each isotope N(3) and N(1):

$N(t) = 1.504 \times 10^{23}$ atoms
$N(3) = 6.43 \times 10^{22}$ atoms
$N(1) = 8.61 \times 10^{22}$ atoms

E. <u>Beryllium</u>

1. The mass of Be in the source is 179 gms and the density is assumed to be standard, 1.85 gms/cm^3.

2. The number of Be atoms per cm^3 is:

$$n = \frac{\rho Na}{A} = 1.24 \times 10^{23} \text{ atoms/cm}^3$$

3. From the geometry of the source it is assumed that the average path of an Sb gamma through the Be is 1 in.

IV. CALCULATION OF SOURCE ACTIVATION

A. Equilibrium activity of Sb^{124}, 60 day half-life:

1. Water Boiler operating cycle and averages: The Water Boiler is usually operated during normal working hours, 0800 to 1700, five days a week. Occasionally it is operated at night or on weekends. During the first five months of 1955 the reactor was operated on the average of 32.3 hours per week or 6.46 hours per day. The hours operated per week varied from a low of 7 to a high of 60. The reactor power during operation is usually 25 kw.

2. Activation equation (developed by R. E. Carter):

a. Definition of symbols:

N_3 = number of Sb^{123} atoms, essentially constant

ϕ = average thermal neutron flux, assumed constant

The center of the Sb source is 7.5 in. from the center of the sphere, and the thermal flux at this position is 5.5×10^{11} neutrons/cm^2-sec at 25 kw, Ref. 1.

σ_3 = microscopic thermal neutron activation cross section in barns for Sb^{123} $(n,\gamma)Sb^{124}$, 60 day half-life

λ_4 = decay constant for the 60 day activity of Sb^{124} per day

td = reactor operation time per day, day

tn = reactor shut-down time per day, day

A = activity = $\frac{\text{disintegrations}}{\text{sec}}$

σ_4 = the Sb^{124} thermal neutron absorption cross section, in barns

N_4 = number of Sb^{124} atoms

b. The number of Sb^{124} atoms formed:

$$\frac{dN_4}{dt} = N_3\,\sigma_3\,\phi - \lambda_4 N_4 - \sigma_4 N_4\,\phi$$

σ_4 is not known, but assuming it to be about the same as σ_3 or 5 barns,

$$\sigma_4\,\phi = 2.36 \times 10^{-7}\text{/day}$$

$$\lambda_4 = 1.16 \times 10^{-2}\text{/day}$$

Therefore $\sigma_4\,\phi$ may be neglected in comparison to λ_4:

$$\frac{dN_4}{dt} = N_3\,\sigma_3\,\phi - \lambda_4 N_4$$

$$N_4 = \frac{N_3\sigma_3\phi}{\lambda_4}\,(1 - e^{-\lambda_4 td})$$

The activity after the first day's operation is (omitting the subscripts on N, λ, and σ):

$$A = N_4\,\lambda_4 = N_3\,\sigma_3\,\phi\,(1 - e^{-\lambda td}),$$

And after the first night shut-down, on Tuesday at 0800, A(1) being the activity after one day of operation and shut-down:

$$A(1) = N\,\sigma\,\phi\,(1 - e^{-\lambda td})\,e^{-\lambda tn}$$

After the second day of operation and night shut-down, on Wednesday at 0800:

$$A(2) = N\sigma\phi(1-e^{-\lambda td})\,e^{-2\lambda tn} + N\sigma\phi\,(1-e^{-\lambda td})\,e^{-\lambda tn}$$

After five days of operation, Monday through Friday, on Saturday at 0800:

$$A(5) = N\sigma\phi\,(1-e^{-\lambda td})\,(e^{-\lambda tn} + e^{-2\lambda tn} + e^{-3\lambda tn} + e^{-4\lambda tn} + e^{-5\lambda tn})$$

The decay time over Saturday and Sunday of 48 hours is 2.74 times the average night shut-down time. Therefore, on Monday at 0800 the activity is:

$$A(7) = N\sigma\phi(1-e^{-\lambda td})\,(e^{-\lambda tn}+\ldots+e^{-5\lambda tn})\,e^{-2.74\lambda tn}$$

A(7) = K, a constant

During the next 7 days the same amount of activity is added on the average, but the first week's activity has decayed for 7 days or 9.58 times tn.

$$A(14) = K(1+e^{-9.58\lambda tn})$$

$$A(21) = K(1+e^{-9.58\lambda tn} + e^{-2 \times 9.58\lambda tn})$$

If (p + 1) = the number of weeks after start of irradiation,

$$A(p+1) = K(1+e^{-9.58\lambda tn} + \ldots + e^{-9.58\,p\lambda tn})$$

Let $\gamma = e^{-9.58\lambda tn}$

$$A(p + 1) = K\,(1 + \gamma + \gamma^2 + \ldots + \gamma^p)$$

As p becomes large, A approaches Ae, the equilibrium activity.

$$Ae = K\,(1 + \gamma + \gamma^2 + \ldots) = K\,\left(\frac{1}{1 - \gamma}\right),$$

for $\gamma < 1$.

Letting $\alpha = e^{-\lambda tn}$,

$$Ae = \frac{N\sigma\phi}{1-\gamma}\,(1-e^{-\gamma td})\,(1 + \alpha + \alpha^2 + \alpha^3 + \alpha^4)\,\alpha^{3.74}$$

3. Saturation and equilibrium values: If the irradiation was continuous and for many half-lives the saturation activity, As, would be approached:

$$As(4) = N_3 \sigma_3 \phi = 6.84 \times 10^{10} \frac{dis}{sec}$$

$$As(4) = 2.39 \text{ curies}$$

The equilibrium activity, Ae, resulting from the intermittent operation, is less than As.

$$\alpha_4 = e^{-\lambda_4 tn} = 0.99160$$

$$\gamma_4 = e^{-9.58\lambda_4 tn} = 0.92233$$

$$e^{-\lambda_4 td} = 0.99690$$

$$\frac{1}{1-\gamma_4} = 12.875$$

$$(1 + \alpha_4 + \alpha_4^2 + \alpha_4^3 + \alpha_4^4) = 4.9167$$

$$\alpha_4^{3.74} = 0.96893$$

$$Ae(4) = 2.39\ (0.1801) = 0.454 \text{ curies}$$

4. Time to 90 percent of Ae: The Sb^{124} activity will be 90 percent of the equilibrium value when:

$$\Sigma_1^{p+1} (1+e^{-9.58\lambda tn} + \dots + e^{-px9.58\lambda tn}) = \frac{0.90}{1-\gamma} = 11.5875$$

Each term in the series represents one week.

$$\Sigma_1^{28} (1 + e^{-9.58\lambda tn} + \dots) = 11.64$$

Thus the Sb^{124} activity would reach 90 percent of its equilibrium value in 29 weeks after start of irradiation.

B. Equilibrium Activity for Sb^{122}:

1. Saturation and equlibrium activity:

$$e^{-\lambda_2 td} = 0.93442$$

$$\gamma_2 = e^{-9.58\lambda_2 tn} = 0.17130$$

$$\alpha_2 = e^{-\lambda_2 tn} = 0.83180$$

$$(1 + \alpha_2 + \alpha_2^2 + \alpha_2^3 + \alpha_2^4) = 3.57791$$

$$\alpha_2^{3.74} = 0.5025$$

$$As(2) = N_1 \sigma_1 \phi = 3.22 \times 10^{11} \frac{dis}{sec} = 8.70 \text{ curies}$$

$$Ae(2) = 8.70\ (0.1422) = 4.58 \times 10^{10} \frac{dis}{sec}$$

$$Ae(2) = 1.24 \text{ curies}$$

2. Time to 99 percent of Ae:

$$\Sigma_1^{p+1} (1 + \gamma_2 + \dots + \gamma_2^{p}) = \frac{0.99}{1 - \gamma_2} = 1.195$$

$$\Sigma_1^{2} (1 + \gamma_2 + \gamma_2^{2}) = 1.2001$$

Thus, after three weeks the Sb^{122} activity is over 99 percent of its equilibrium value. However, for a 2.75 day half-life the equation based on an average week's operating time is really not applicable.

C. Equilibrium activity of Sb by the average flux Method: The Sb^{-124} activity may also be estimated from an assumption of an average continuous flux. The average operating time per week is 32.3 hours and the normal power is 25 kw. Thus, the average continuous power and flux would be:

$$Pav = 4.81 \text{ kw}$$

$$\phi av = 1.08 \times 10^{11} \frac{neuts}{cm^2\ sec}$$

$$Ae(4) = 1.70 \times 10^{10} \frac{dis}{sec} = 0.46 \text{ curies}$$

This result is nearly identical with the previous calculation. The intermittent daily operation time and weekly cycle are small compared to the 60 day half-life.

The Sb^{122} activity by the average flux method is:

$Ae(2) = 6.20 \times 10^{10} \frac{dis}{sec} = 1.68$ curies, which is 35 percent larger than by the intermittent operation equation.

D. Neutron strength of source: From Section I below, about 30 percent of the gammas emitted by Sb^{122} and Sb^{124} are above the $Be(\gamma,n)$ threshold of 1.67 Mev. Thus the effective curie strength of the Sb at equilibrium is:

$$Ae(2) = 1.\ 24 \text{ curies} \times 0.30 = 0.37 \text{ curies}$$

$$Ae(4) = 0.454 \text{ curies} \times 0.30 = \underline{0.14}$$

$$\text{total} = 0.51 \text{ curies}$$

From the equation for gamma ray attenuation $I_x = I_o e^{-n\sigma x}$ the number of (γ,n) reactions is $I_o - I_x = I_o (1 - e^{-n\sigma x})$

$$n = 1.24 \times 10^{23} \text{ atoms/cm}^3$$

$\sigma = 1 \times 10^{-27}$ cm^2, from Section I, below.

The average gamma-ray path through Be surrounding each Sb rod is assumed, from the shape of the source, to be about one inch.

Source stength = 6.23×10^6 neuts/sec on Mondays at 0800.

V. CALUCLATION OF SPHERE FLUX FROM SOURCE NEUTRONS

A. Summary: The neutron flux in the sphere caused by neutrons from the Sb(γ,n)Be reactions may be estimated by a calculation based on the assumption that the neutron flux is uniform throughout the volume of the solution in the sphere.

B. Fraction of Source neutrons entering the sphere:

The number of source neutrons entering the sphere is assumed to be proportional to the solid angle at the source that is subtended by the sphere. A more accurate calculation would involve neutron diffusion theory beyond the scope of this paper.

The center of the source is about 1.5 in. from the surface of the sphere, which is 12 in. in diameter. Thus the solid angle subtended by the sphere is:

$$\Omega = 0.8\ \pi$$

The fraction of the source neutrons striking the sphere is:

$$F = \Omega/4\pi = 0.2$$

The absorption of neutrons in the graphite is negligible.

C. The Uniform flux equation:

The neutron flux in the sphere may be calculated from the following expression,

$$\phi = SFml(1/V)v$$

The symbols are:

ϕ = thermal neutron flux, neutrons/cm^2 - sec

S = Sb-Be source strength = 6.23×10^6 neutrons/sec

F = fraction of source neutrons entering sphere = 0.2

m = the subcritical multiplication of the sphere with all the control rods in

l = lifetime of fission neutrons in the sphere = 10^{-4} sec

V = volume of solution in sphere = 12.7 liters

v = velocity of thermal neutrons = 2.2×10^5 cm/sec

The subcritical multiplication of the sphere is

$$m = \frac{1}{1 - k_{eff}} = \frac{1}{-k_{ex}}, \text{ for } k_{eff} < 1$$

The excess multiplication factor for the reactor with all the control rods in may be estimated as follows: The position of each of the rods has been calibrated in terms of the number of grams of U^{235} in the sphere. Also, it has been determined that 30 grams of U^{235} in the sphere is equal to an excess multiplication factor of 0.01. The critical condition of the reactor during the period that the measurements were taken was:

Rod	Indicator	Grams equivalent	k_{ex}
East Cd	350	30	0.01
West Cd	350	30	0.01
Safety Rod (Cd)	out	30	0.01
West B	970	80	0.027
East B	237	22	0.007
	Total	192	0.064

Thus, with all the rods in k_{ex} would be:

$$k_{ex} = -0.064$$

The multiplication with the rods in is thus:

$$M = \frac{1}{0.064} = 15.6$$

Substituting the values given the thermal neutron flux in the sphere is:

$$\phi = 3.36 \times 10^4 \text{ neutrons/cm}^2\text{-sec}$$

VI. MEASUREMENTS OF NEUTRON FLUX IN SPHERE

A. Summary

It was originally planned to determine the effect of the Sb source relative to Ra source, and the counting rates from a fission chamber in the sphere were to be used for this purpose. After the Sb source had been installed, irradiated for 2 weeks, and relative counting rates taken with the fission chamber, it was decided to determine the sphere flux with indium foils. The In foils and the counting equipment were calibrated by exposing the foils to a known neutron flux in the LASL Sigma Pile, i.e., a pure graphite pile containing a neutron source of known strength. The fission chamber counting rate was then related to the sphere flux as determined by the In foils.

The following sections describe the fission chamber, the In foils and counting equipment, the In foil technique, and give the results of the measurements.

B. The design of the fission chamber used is shown in Figs. 6 and 7. The chamber and tube are covered with insulating tape, so that the equipment is grounded only at the amplifier and scaler. The chamber operates on 150 volts from a modified Model 101 Preamplifier. From the preamplifier the pulses go to a Model 101 amplifier and are recorded on a Model 700 scaler. These items are standard LASL equipment.

The geometry of the chamber is largely determined by the 1 in. internal diameter of the reentrant tube in the sphere, and the need for a large area of U^{235} to give a reasonable counting rate. A satisfactory plateau of counting rate vs. discriminator setting, Fig. 8, was obtained by using a high gain on the amplifier.

C. The In foils used were the standard size used at LASL: 1.60 in. wide, 2.56 in. long, 0.005 in. thick, and weighed about 2.30 grams. A foil holder, Fig. 9, and tube were used to position the foil in the sphere. The counting equipment consisted of a Mark 1, Model 10A, Radiation Counter Laboratories, Geiger tube in a Model S85, Technical Associates, lead pig, and a Model 700 scaler that is also a high voltage supply for the Geiger tube. For exposure, the foil was smoothed in the annulus inside the holder, Fig. 9. For counting, the foil was smoothed in a standard brass tube so that the outer surface of the exposed foil was next to the Geiger tube when the brass and foil were positioned around the Geiger tube. A counting rate versus voltage plateau for the Geiger tube is shown in Fig. 10.

D. The following outline of the In foil method of determining neutron fluxes was adapted from the developments given in Froman and Graves, Ref. 2, and Glasstone, Ref. 3.

Natural In contains 95.8 percent In^{115} and 4.2 percent In^{113}. The slow neutron reactions of interest are:

$$In^{115} + n \rightarrow In^{*116} \rightarrow Sn^{116} + \beta + \gamma$$

In^{*116} decays with two half-lives: one of 15 sec. with a β of 2.8 Mev and no gammas, the other of 54 min. half-life, a β of 0.85 Mev and 4 gammas from 0.43 to 2.32 Mev.

N_6 = number of In^{116} atoms

N_5 = number of In^{115} atoms

$$dN_6/dt = N_5 \sigma_5 \phi - \lambda_6 N_6$$

In^{116} neutron captures being insignificant.

te = time of foil exposure to neutron flux

$$N_{6_{te}} = N_5 \sigma_5 \phi(1 - e^{-\lambda 6te}) / \lambda_6$$

In^{116} activity = $N_6 \lambda_6$ = dis/sec

Omitting the subscripts 5 and 6,

$$A = N \sigma \phi (1 - e^{-\lambda te})$$

$As = N \sigma \phi$, The saturation activity.

After a waiting time, (tw), the number of In^{116} atoms, is

$$Ntw = Nte\ e^{-\lambda tw}$$

$$Atw = N_5 \sigma_5 \phi(1 - e^{-\lambda te}) e^{-\lambda tw}$$

If Nd is the number of disintegrations in a time interval,

$$A = \frac{dNd}{dt}$$

$$\int_0^{Nd} dNd = N\sigma\phi(1-e^{-\lambda te}) \int_{tw}^{tw + tc} e^{-\lambda t}dt,$$

where tc is the counting time.

$$Nd = (N\sigma\phi/\lambda)(1-e^{-\lambda te})(e^{-\lambda tw})(1-e^{-\lambda tc})$$

$$As = Nd\lambda / (1-e^{-\lambda te})(e^{-\lambda tw})(1-e^{-\lambda tc})$$

This equation is used to calculate the saturation activity of a foil exposed to an unknown neutron flux, ϕ. "te" is selected to give a satisfactory counting rate, and tc to get a total count required for the statistical accuracy desired. A tw of 3 min. is used to allow the 13-sec. activity to decay. Nd, the total number of disintegrations in tc, is proportional to the total number of counts of the exposed foil recorded by the counting equipment.

E. Calibration of the foils and counting system:

The known flux in the Sigma Pile is used to calculate the constant, K, in the following equation. K is an inverse efficiency factor for the counting system, since Nd, the number of disintegrations in tc, is greater than the number of counts recorded:

$$\phi th = K_1 \text{ (neuts/cm}^2\text{ct) } As \text{ (ct/sec)}$$

As indicated in the calibration data below, K_1 is corrected to K_2 to give the thermal neutron flux, since the Sigma Pile and sphere fluxes are not completely thermalized.

1. Sigma Pile Neutron Flux in Tray #10:

Source #40 $Q = 5.79 \times 10^6 \pm 2\%$ (8/54)

#44 $Q = \underline{6.08 \times 10^6}$

Sum $= 11.87 \times 10^6$ n/sec

Flux in Tray #10:

ϕ (thermal, for 11.48×10^6) =
4658 neuts. th/cm^2 sec

ϕ (thermal, for 11.87×10^6) =
4658 (11.87/11.48)

ϕ (thermal, for 11.87×10^6) =
4816 n.th./cm^2 sec

Activities of In foils exposed in Sigma Pile, for ϕ = 4816.

Ab is the activity of an In foil exposed "bare", or directly to a neutron flux. Acd is the activity of an In foil covered with Cd and exposed to a neutron flux. Ath is defined as Ab-Acd.

Ab = 50,313 cts
Acd = $\underline{\ 2,822}$
Ath = 47,491

cts th/cts b = 47,491/50 313 = 0.9439 Ath/Ab

2. Calculation of K for P-2 In counting system:

As(th) = (1/K) ϕth (Σ-Pile)

K = ϕth/As(th) neuts/cm^2 sec/cts/sec

$$As = \frac{Nc(cts)\ \ \lambda(1/min)}{60\ sec/min\ (e^{-\lambda tw})(1-e^{-\lambda te})(1-e^{-\lambda tc})} = cts/sec$$

In-A foil:

te = 5 min
tw = 45 min
tc = 10 min

N_T = 16,331
Bgd = - 322
Nc = 16,009

As = 815 cts-b/sec

K_1 = 4816 neuts th/cm^2sec/(815 cts-b/sec) x (0.944 cts-th/cts-b)

K_1 = 4816/769 = 6.26 th neuts/cm^2)(cts-th)

ϕwb = K_1 (th neuts/cm^2 cts-th) x As(cts-b/sec) x Ath wb/Ab wb

Cd ratio (for 5 mil In foils) in Water Boiler sphere = 3.05, Ref .14.

Cd ratio = Ab/Acd

Ath = Ab - Acd

$$Ath/Ab = 1 - Acd/Ab = 1 - 1/Cd\ ratio = \frac{(Cd\ ratio\ -1)}{Cd\ ratio} = 0.672$$

ϕwb = K_2 As

ϕwb = 6.26 x 0.672 x As th neuts/cm^2sec = 4.21 As

K_2 = 4.21 th neuts/cm^2 cts-b

F. Counting and Foil Exposure Procedures:

1. The measurements of the sphere flux were made on Sunday or Monday mornings, 1-3/4 to 2-3/4 days after the reactor had been shut down on Friday afternoon. This was necessary to allow the neutron background in the sphere to decay to its normal level, and for the sphere gammas to decay. Counts were taken every week after the new source was first installed for four weeks and then about once a month.

2. Equipment warm-up: The counting equipment was turned on and allowed to warm up about one hour before counts were recorded. No calibration check was made for the fission counter, but a U-glass source was used to check the In foil counter.

3. Counting Rates and the total number of counts: For the fission counter the counting rates not in excess of 1200 cpm were within the capabilities of the electronic equipment. Three determinations of the counting rate were made, each with a total count of not less than 1000 counts, which gave a standard deviation of about 2 percent.

For the Geiger counter the total number of counts in ten minutes had extreme values of 7000 and 129,000.

4. Initial counting: The initial counting rates of the Ra source alone, the Sb source alone, and the two sources together were made with the fission counter only, since it had not been decided to use foils at that time. The initial neutron emission of the Sb source was caused by gamma rays from the sphere and not from the Sb in the source. By subtracting the initial counting rates from subsequent rates the change in activity of the source could be determined and the Sb source compared to the Ra source.

The fluxes measured with the fission counter included the background flux from the sphere, ϕ(Sp), caused by spontaneous fissions, (γ,n) reactions, cosmic rays, etc. The background flux can be calculated from the initial flux:

$$[\phi(Ra + Sp) - \phi(Sp)] + [\phi(Sb + Sp) - \phi(Sp)] = \phi(Sb + Ra + Sp) - \phi(Sp)$$

$$\phi(Sp) = \phi(Ra + Sp) + \phi(Sb + Sp) - \phi(Sb + Ra + Sp)$$

From the values given in Table I, Data Summary:

$\phi(Sp) = 955 + 1183 - 1890 = 228$ th neuts/cm^{2}sec

Unfortunately the counting rate in the sphere was not determined with both sources removed, hence the value for $\phi(Sp)$ was not determined directly. But, the neutron background in the sphere is included in all the counts, and is presumably eliminated in the differences.

After installation it would have been inconvenient to remove the Sb source, but the Ra source could be withdrawn to the top of the shield. Consequently all counts after the initial counts were made of the Sb and Ra sources together and of the Sb alone.

The radium source in the withdrawn position was separated from the graphite tamper by 70 in. of concrete and steel, compared to its normal position in the graphite of 20-1/4 in. from the sphere center. It is estimated that the shielding reduced the neutron flux entering the graphite from the Ra source by a factor 10^5 or 10^8. The strength of the Ra source, #34, of 200 millicuries was 1.66×10^6 neuts/sec. Thus the neutrons from the Ra source should not have been detected when the Sb source was counted alone.

5. Subsequent counting: The procedure for all counting after the initial ones was as follows:

a. Fission counter:

(1) Counting rate of the sphere flux with the Sb and Ra sources in position.

(2) Counting rate of the sphere flux with the Ra source withdrawn.

b. In foils:

(1) Expose a foil with the Ra source withdrawn and count the foil activity.

(2) Expose a foil with the Ra and Sb sources in position and count the foil activity.

G. Analysis of the Data:

1. Thermal flux from In foils: Measurements of the sphere flux were made at twelve different times from December, 1954, to September, 1955. The first three measurements were made with the fission counter and the last nine with the fission counter and In foils.

For each foil exposure the saturation activity, As, was calculated from the equation given above using the total number of counts, Nc, during the counting time. Nc is related to Nd, the number of disintegrations through the counting system constant K_2 as given above. The constant K_2 also contains the cadmium ratio correction. Thus the thermal neutron flux for each exposure was:

ϕ th wb = K_2(th neuts/cm^2-cts b) x As(cts b/sec)

2. Calibration of the fission counter: For the last nine measurements the average ratio of flux to fission counting rate was determined:

$R = \phi(Sb + Sp)/Ct(Sb + Sp) = 13.06$ th neuts/cm^2 sec/ f ct/min

This ratio applied strictly to one foil, In-A, since it was the only one calibrated in the Sigma Pile. Although the ratio for the other foils used was within 4% of the In-A value. Also, the value of R was determined from the fission counting rates and saturation activities resulting from the Sb source only. Thus the flux is:

$\phi(Sb + Sp) = R \times Ct(Sb + Sp)$

3. Neutron flux caused by the Sb activity of the source: The fission counting rate from the Sb source and the ratio, R, were used to determine the sphere flux for all fission counting rates, since the initial Sb flux had to be determined this way. The change in activity of the Sb is related to the difference between a subsequent flux and the initial flux. The multiplication of the sphere, the gamma flux from the sphere, and the neutron background in the sphere, after a week-end shutdown being nearly constant. The change in flux due to Sb activity is:

$\phi(Sb) = \phi(Sb + SP) - \phi(Sb + Sp)0$

A summary of the data is given in Table I, and a graph of the sphere flux due to Sb activity vs. Kw Hrs of reactor operation is given in Fig. 11.

An average value of the measured equilibrium Sb flux, obtained from the last four measurements, is:

$\phi(Sb)m = 13{,}000$ th neuts/cm^{2}sec

This is to be compared with the calculation of the equilibrium Sb flux:

$\phi(Sb)c = 33{,}600$ th neuts/cm^{2}sec

The equilibrium date cannot be estimated closely from the curve, Fig. 13, since no measurements were taken between 3/27/55 and 6/12/55. But estimating 95% equilibrium at 17,000 kw-hrs, the Water Boiler records showed that this occurred on 5/23/55 - 140 days after first significant operation of the reactor with the Sb source. The times to equilibrium calculated above, for each isotope individually, were:

$Te(2) = 3$ weeks to 99% $Ae(2)$

$Te(4) = 29$ weeks to 90% $Ae(4)$

The time to equilibrium for the Sb source, which contains both isotopes, can be calculated as follows: On a Monday at 0800 the equilibrium activities were previously calculated to be:

$$Ae(2) = 1.24 \text{ curies}$$
$$Ae(4) = \underline{0.45} \text{ curies}$$
$$Ae(T) = 1.69 \text{ curies}$$

After the first three weeks the growth of source activity is essentially the Sb^{124} growth. The activity of the source at the 95% of equilibrium is:

$$0.95\ Ae(T) = 1.61 \text{ curies}$$
$$-\ Ae(2) = \underline{1.24} \text{ curies}$$
$$(y\%)\ Ae(4) = 0.37 \text{ curies}$$
$$y = 0.37/0.45 = 82\%$$
$$.82[1/(1-\gamma_4)] = .82(12.875) = 10.6$$

The sum of the series $(1 + \gamma + \gamma^2 \ldots)$ to 21 terms is 10.5. Thus the time to 95% equilibrium is 21 weeks, which agrees with the estimate from the curve on Fig. 11 of 20 weeks.

VII. CONCLUSIONS

A. Initially the Sb-Be source produced a total neutron flux in the sphere larger than that produced by the Ra-Be source.

$$\phi(T)_0 = \phi(Sb + Sp)_0 / \phi(Ra + Sp)0 =$$
$$1183/935 = 1.27$$

B. The initial ratio of the fluxes caused by the sources, i.e., subtracting the sphere background flux from the total flux:

$$\phi(S)_0 = \frac{\phi(Sb + Sp)_0 - \phi(Sp)_0}{\phi(Ra + Sp)_0 - \phi(Sp)_0} =$$
$$1183-228)/ (935-228) = 1.35$$

C. The equilibrium ratio of the fluxes caused by the Sb source and the Ra source is:

$$\phi(S)_e = \frac{\phi(Sb + Sp)_e - \phi(Sb + Sp)_o}{\phi(Ra + Sp)_o - \phi(Sp)} =$$
$$\frac{14{,}443-1183}{935-228} = \frac{13{,}260}{707} = 18.8$$

D. The equilibrium ratio of the total flux in the sphere for the two sources is:

$$\phi(T)_e = \phi(Sb + Sp)_e / \phi(Ra + Sp)_o =$$
$$14{,}443 / 935 = 15.4$$

It is a flux from the total sphere flux that is monitored during shut-down and start-up.

E. A comparision of the calculated and measured values of the sphere flux shows that useful estimates of the effect of the Sb source could be made.

F. About 73% of the neutron emission of the source on Monday at 0800 is caused by Sb^{122} activity; only the Sb^{124} activity is considered in references 4 and 5.

VIII. NUCLEAR REACTIONS AND CONSTANTS INVOLVED

A. $\underline{Be^9(\gamma,n)Be^{8*}}$ reaction:

1. The threshold energy for the $Be^9(\gamma,n)$ reaction is given by Wattenberg, Ref. 4, as 1.83 Mev, and 1.67 Mev by Friedlander and Kennedy, Ref. 5.

2. The cross section for the reaction varies with the gamma energy. Two values given by Russell, et al., Ref. 6, are:

$$\sigma(1.67 \text{ Mev}) = 9.7 \times 10^{-4} \text{ barns}$$
$$\sigma(2.76 \text{ Mev}) = 7.0 \times 10^{-4} \text{ barns}$$

An average value of the cross section for the gamma rays emitted by Sb can be calculated from the neutron yield of an Sb-Be source given by Wattenberg, Ref. 4.

$$y = 1.9\text{x}10^5 \ \frac{\text{neuts}}{\text{sec-curie-gm Be at 1 cm}} = N\sigma\phi$$
$$\sigma = 9.7 \times 10^{-4} \text{ barns}$$

Hammermesh and Kimball, Ref. 7, give values of the cross section for several gamma energies:

$$\sigma(1.70 \text{ Mev}) = 1.0 \times 10^{-3} \text{ barns}$$
$$\sigma(1.81 \text{ Mev}) = 0.6 \times 10^{-3} \text{ barns}$$
$$\sigma(2.50 \text{ Mev}) = 0.5 \times 10^{-3} \text{ barns}$$

3. The neutron energy from this reaction is given by Wattenberg, Ref. 4, as 0.025 and 0.055 Mev.

4. Two alpha particles are emitted either simultaneously with the $Be^9(\gamma,n)$ reaction or in less than 10^{-14} sec from the disintegration of Be^{8*}, Ref. 8. The energy of the alpha particles is given as 0.039 Mev, Ref. 2. No neutrons will result from these alphas since the threshold energy for the $Be^9(\alpha,n)C^{12}$ reaction is about 0.2 to 0.3 Mev, Ref. 9.

DATA SUMMARY

Date	Source	Fct CTS/min	In A As cts/sec	In H As Cts/sec	$\phi=K_2A_s$ $K_2=4.21$ th neuts/Sec-cm^2	R = ϕ n/cm^{2}sec / F Cts/Min	ϕ' = Fct x R	($\phi' - \phi_o$) = ϕ (Sb)	Kw-h
12/27/54	Ra	71.6 ± 1.6					935		0
	Sb	90.6 ± 1.1					1183		0
	Sb & Ra	144.7 ± 1.7					1890		0
12/28/54	Sb	84.5 ± 2.5					1104	-79	~ 0
	Sb & Ra	149.5 ± 4.3					1952	+62	~ 0
1/3/55	Sb	78.4 ± 2.4					1024	-159	77
	Sb & Ra	133.5 ± 4.0					1744	-146	77
1/9/55	Sb	206.4 ± 3.0	558 ± 2.1		2349	11.38	2696	1513	768
	Sb & Ra	270.1 ± 4.5		710 ± 2.4	2989*	11.07	3528	1638	768
1/16/55	Sb	252.9 ± 4.0	771 ± 3.4		3246	12.84	3303	2120	1,363
	Sb & Ra	301.8 ± 4.3		881 ± 3.7	3709*	12.30	3942	2052	1,363
1/23/55	Sb	441.4 ± 4.4	1419 ± 5		5972	1353	5765	4582	2,464
	Sb & Ra	497.5 ± 5.0		1650 ± 5	6946*	13.96	6497	4607	2,464
2/20/55	Sb	649.5 ± 4.6	1982 ± 6		8344	12.85	8482	7299	5,393
	Sb			2007 ± 8	8449*	13.01			5,393
3/27/55	Sb	852.4 ± 4.9	2585 ± 9		10,883	12.77	11,132	9,949	8,921
	Sb			2725 ± 9	11,472*	13.46			8,921
6/12/55	Sb	1108.8 ± 5.9	3455 ± 10		14,546	13.12	14,481	13,298	18,624
				3525 ± 10	14,840*	13.38			18,624
8/8/55	Sb	1050.1 ± 4.8	3293 ± 10		13,864	13.20	13,714	12,531	23,696
				3216 ± 10	13,539*	12.89			23,696
9/5/55	Sb	1077.3 ± 4.4	3411 ± 10		14,360	13.33	14,070	12,887	27,022
				3495 ± 10	14,714*	13.66			27,022
9/26/55	Sb	1105.9 ± 7.4	3375 ± 10		14,209	12.85	14,443	13,260	28,688
			3416 ± 10	3416 ± 10	14,381*	13.00	4	51,976	
								12,994 = ϕ "equil".	

R av In a 13.06

R av In H 13.39

* Not accurate since 4.21 factor applies only to In A, although weights close

In A = 2.301 gms
In H = 2.310 gms

B. Antimony:

1. Natural antimony has an atomic weight of 121.76 and consists of two isotopes: 42.75 percent SB^{123} and 56.25 percent Sb^{121}. The thermal neutron absorption cross section for natural antimony is:

$\sigma a = 5.5 \pm 1.0$ barns, Ref. 7

2. $Sb^{123}(n,\gamma)Sb^{124}$ reaction: SB^{123} has the following cross sections for thermal neutrons, Ref. 10.

$\sigma abs = 3.9 \pm 0.3$ barns

$\sigma act(60\ day) = 2.5 \pm 0.5$ barns

The reactions of interest are:

$$Sb^{123} + n \rightarrow Sb^{124*} \rightarrow Sb^{124} + \gamma$$

The maximum capture gamma ray energy is about 8 Mev, the neutron binding energy. The gamma is emitted at the time of capture, i.e., during reactor operation.

$$SB^{124}\ (60\ day) \rightarrow Te^{124} + \beta + \gamma$$

Several gamma energies from 0.60 to 2.32 Mev for the 60 day decay are given in the literature. The percentage of gammas per beta decay above the Be(γ,n) threshold of 1.67 Mev is given as 28 percent by Asuma, Ref. 11; 29 percent by Moreau, Ref. 12; and 56 percent by Lazar, Ref. 13.

3. $Sb^{121}(n,\gamma)Sb^{122}$ reaction: The thermal neutron cross sections are:

$\sigma abs = 5.7 \pm 0.5$ barns, Ref. 7

$\sigma act(2.75\ day) = 6.8 \pm 1.5$ barns

The reactions are:

$$Sb^{121} + n \rightarrow Sb^{122*} \rightarrow Sb^{122} + \gamma$$

$$Sb^{122}(2.75\ day) \rightarrow Te^{122} + \beta + \gamma$$

Gamma energies, for the 2.75 day half-life, from 0.095 to 1.9 Mev are given in the literature. The percentage of gammas per beta decay with energies above 1.67 is reported to be 21 percent by Moreau, Ref. 12; 25 percent by Glaubman, Ref. 14; 30 percent by Farrelly, et al., Ref. 15; and 36 percent by Cork, et al., Ref. 16.

4. Resonance cross sections; The neutron absorption cross section for Sb^{121} has several resonances. The main ones are: 1400 barns at 6.2 ev, 800 barns at 15 ev, and 140 barns at 30 ev. The Sb^{123} cross section has a resonance cross section of 1200 barns at 23 ev, Ref. 10.

The Sb source is located in graphite near the sphere where the measured cadmium ratio for 5 mil thick In foils is 4. Hence, a considerable fraction of the neutrons is in the Sb resonance range. These neutrons will contribute to the Sb activation. Resonance absorption will not be considered in this paper, although rough estimates indicate that it adds about 30 percent to the activity calculated from thermal absorption.

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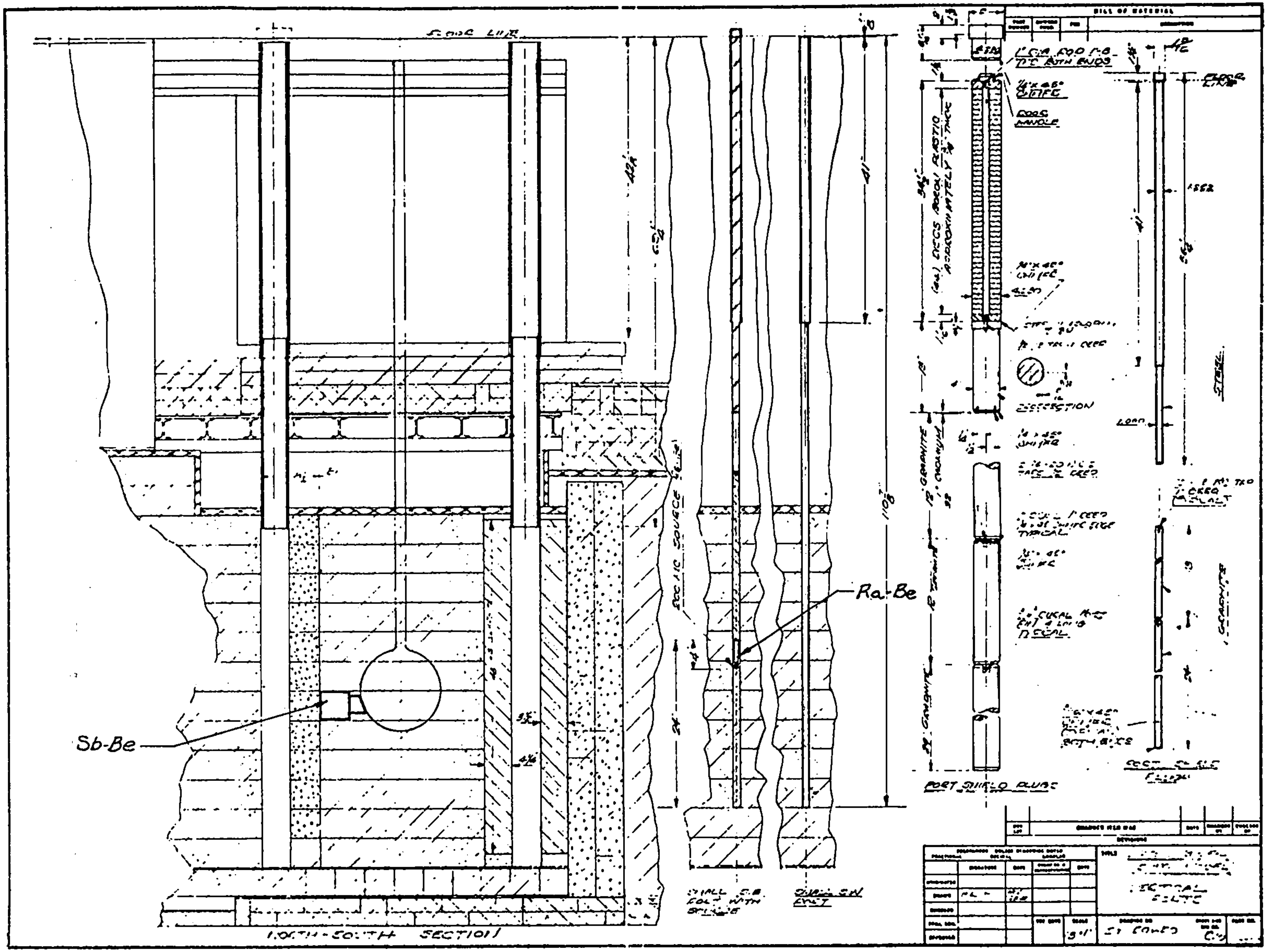


Fig. 1. Vertical Section of Water Boiler with Sb-Be and Ra-Be Sources

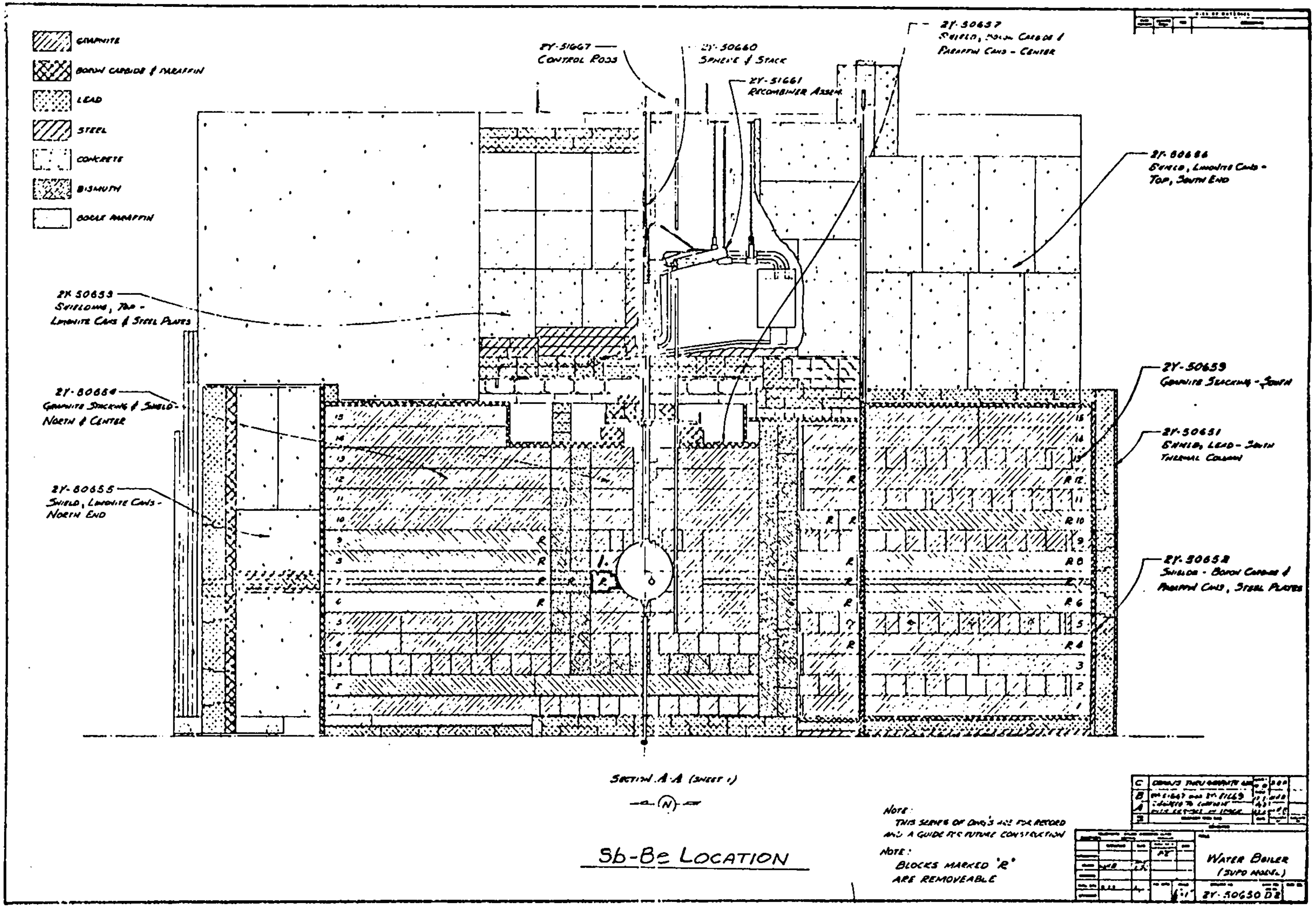


Fig. 2. Vertical Section of Water Boiler with Sb-Be source.

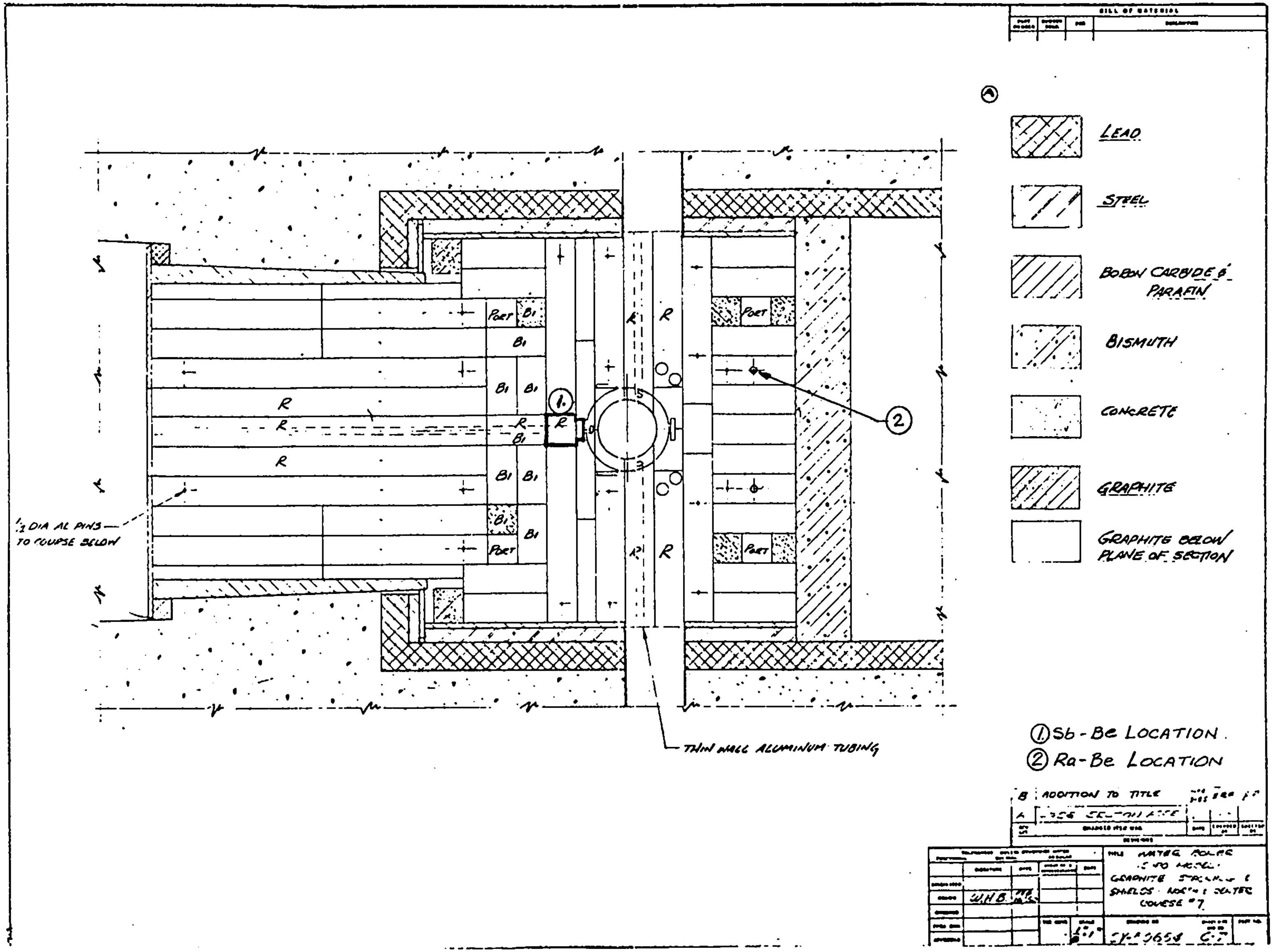


Fig. 3. Horizontal Section of Water Boiler with Sb-Be and Ra-Be Sources

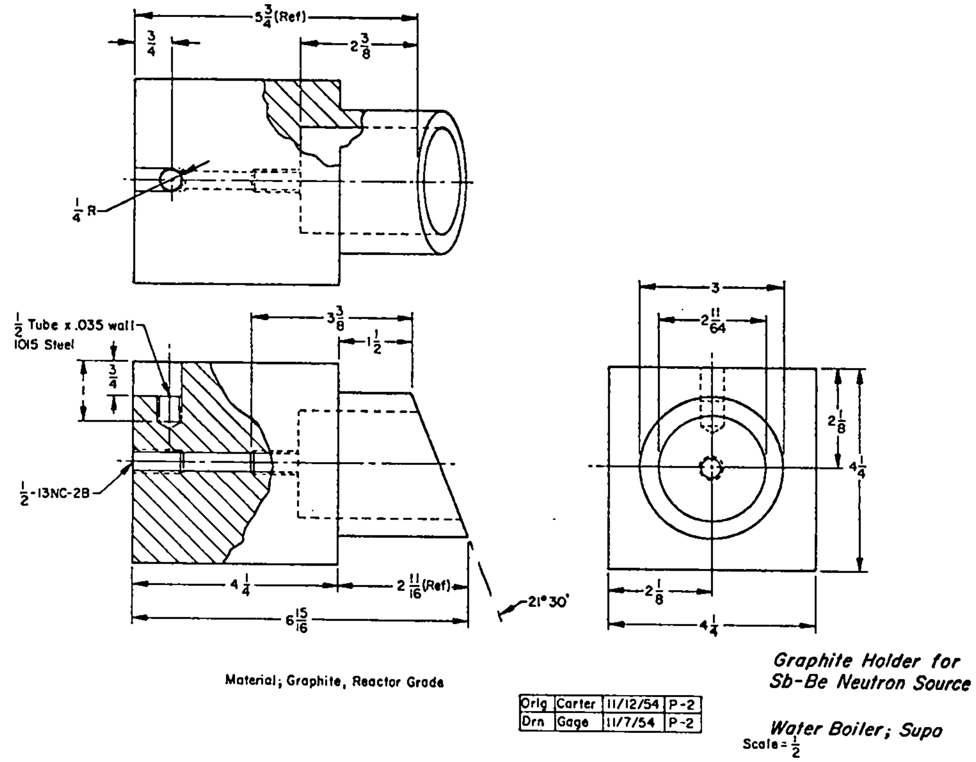


Fig. 4. Graphite Holder for Sb-Be Neutron Source

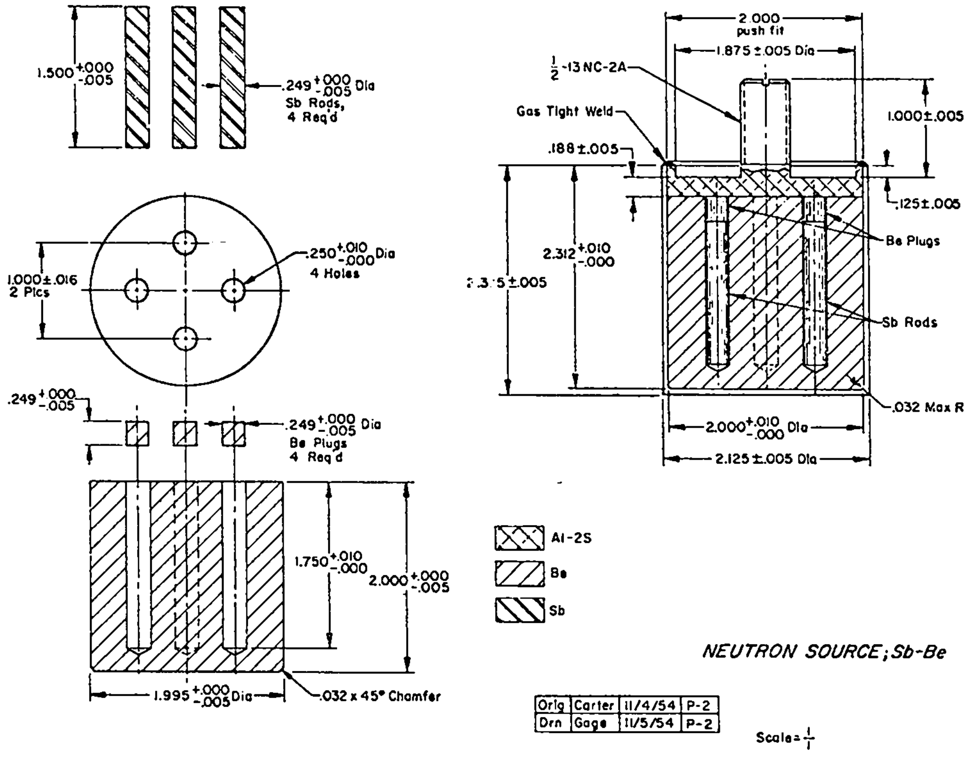
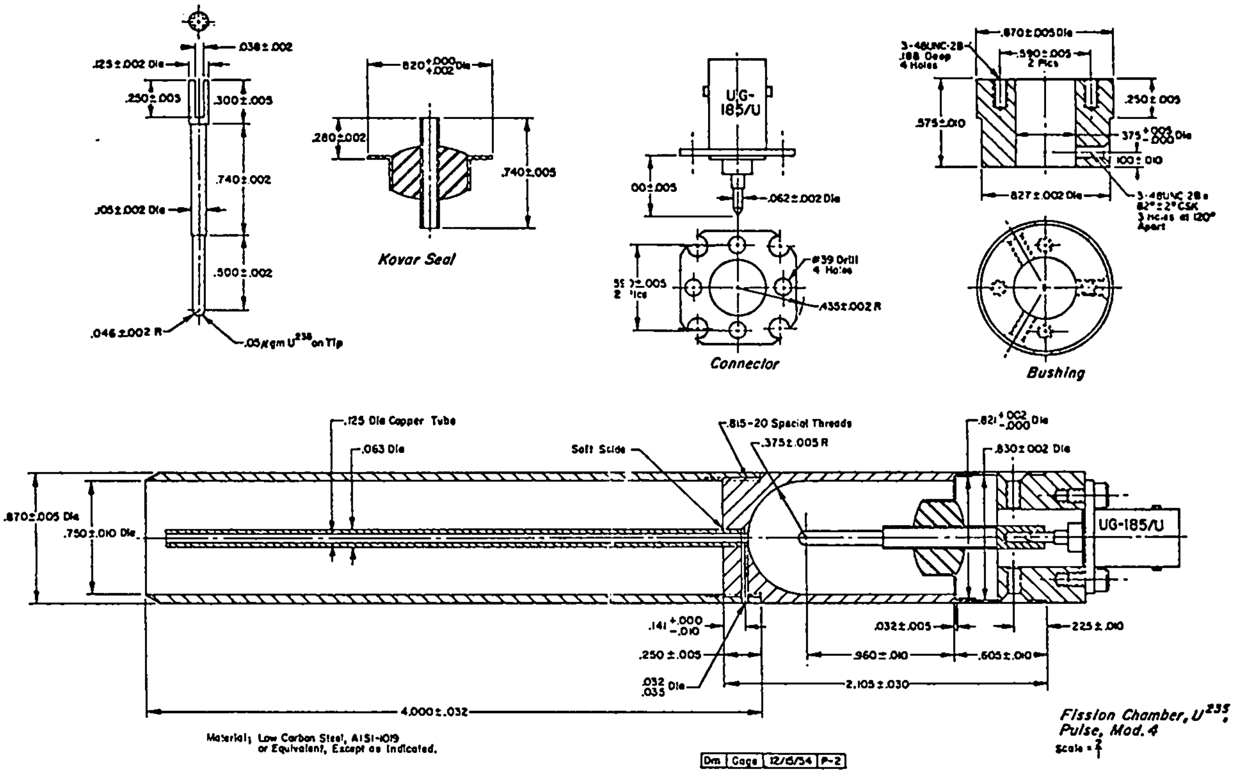


Fig. 5. Neutron Source; Sb-Be

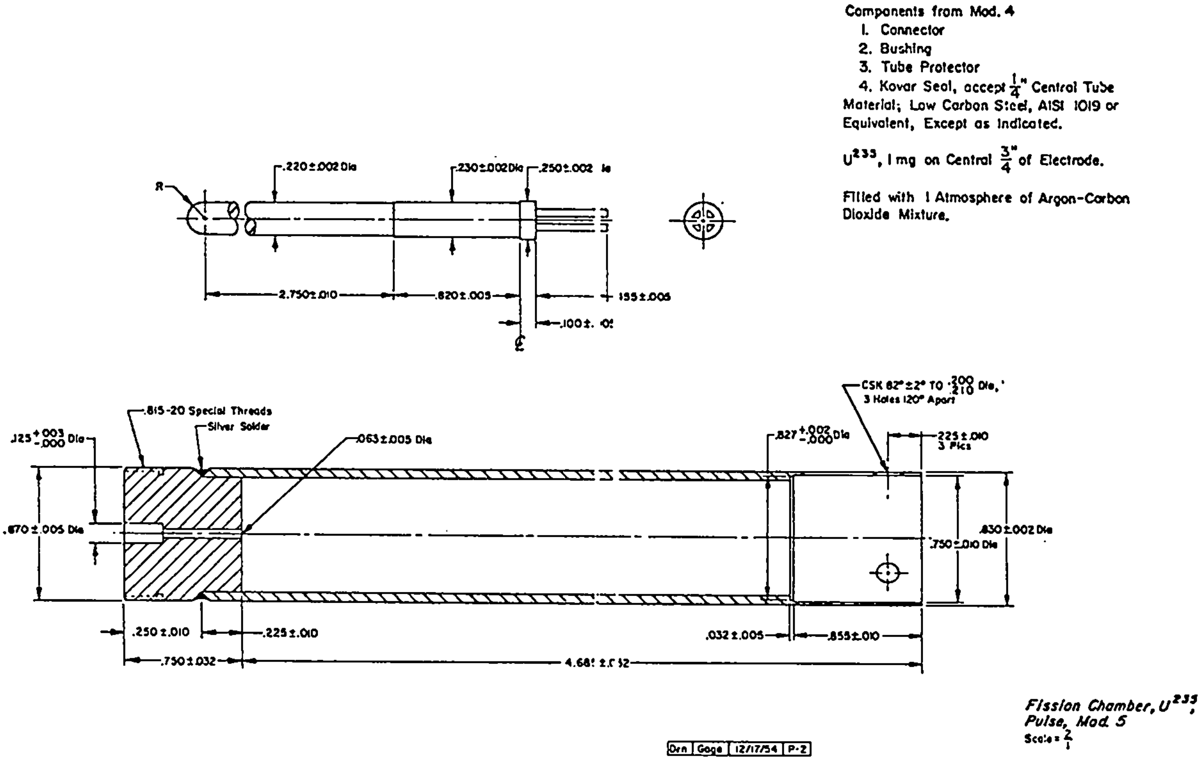


Material; Low Carbon Steel, AISI-1019 or Equivalent, Except as Indicated.

Drn	Gage	12/6/54	P-2

Fission Chamber, U²³⁵, Pulse, Mod. 4
Scale = 2/1

Fig. 6. Fission Chamber, U^{235}, Pulse Mod. 4



Components from Mod. 4
1. Connector
2. Bushing
3. Tube Protector
4. Kovar Seal, accept $\frac{1}{4}$" Central Tube

Material; Low Carbon Steel, AISI 1019 or Equivalent, Except as Indicated.

U^{235}, 1 mg on Central $\frac{3}{4}$" of Electrode.

Filled with 1 Atmosphere of Argon-Carbon Dioxide Mixture.

Drn	Gage	12/17/54	P-2

Fission Chamber, U²³⁵, Pulse, Mod. 5
Scale = 2/1

Fig. 7. Fission Chamber, U^{235}, Pulse Mod. 5

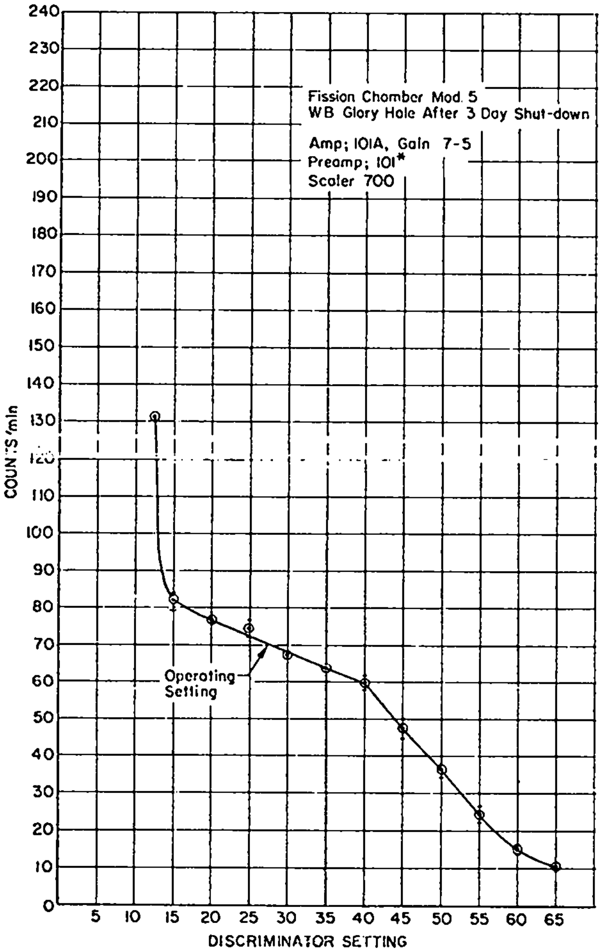


Fig. 8. Discriminator Setting
Fission Chamber Mod. 5

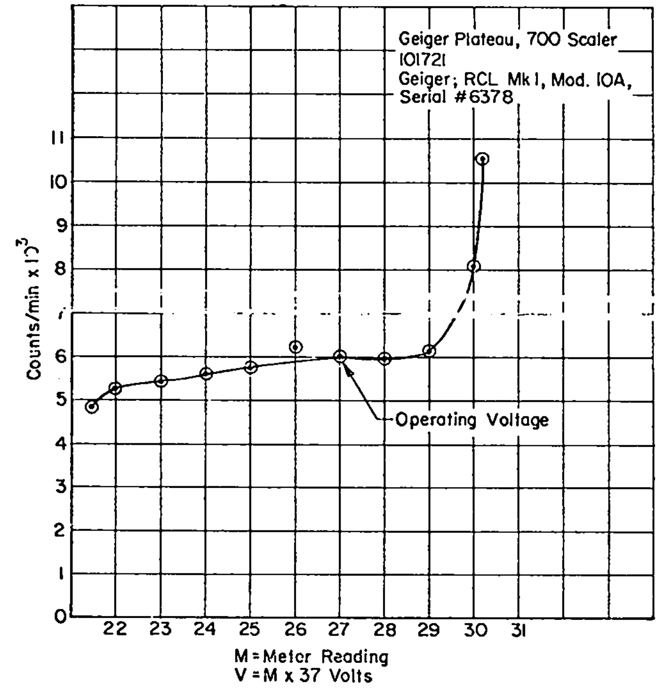
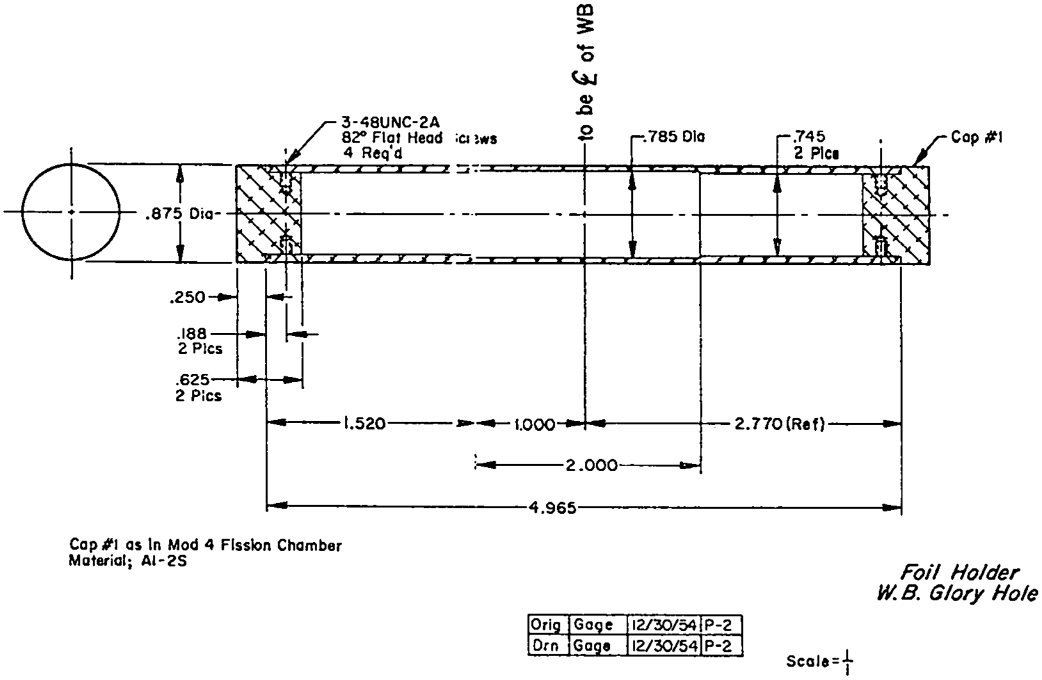


Fig. 10. Geiger Plateau



Cap #1 as in Mod 4 Fission Chamber
Material; Al-2S

Fig. 9. Foil Holder

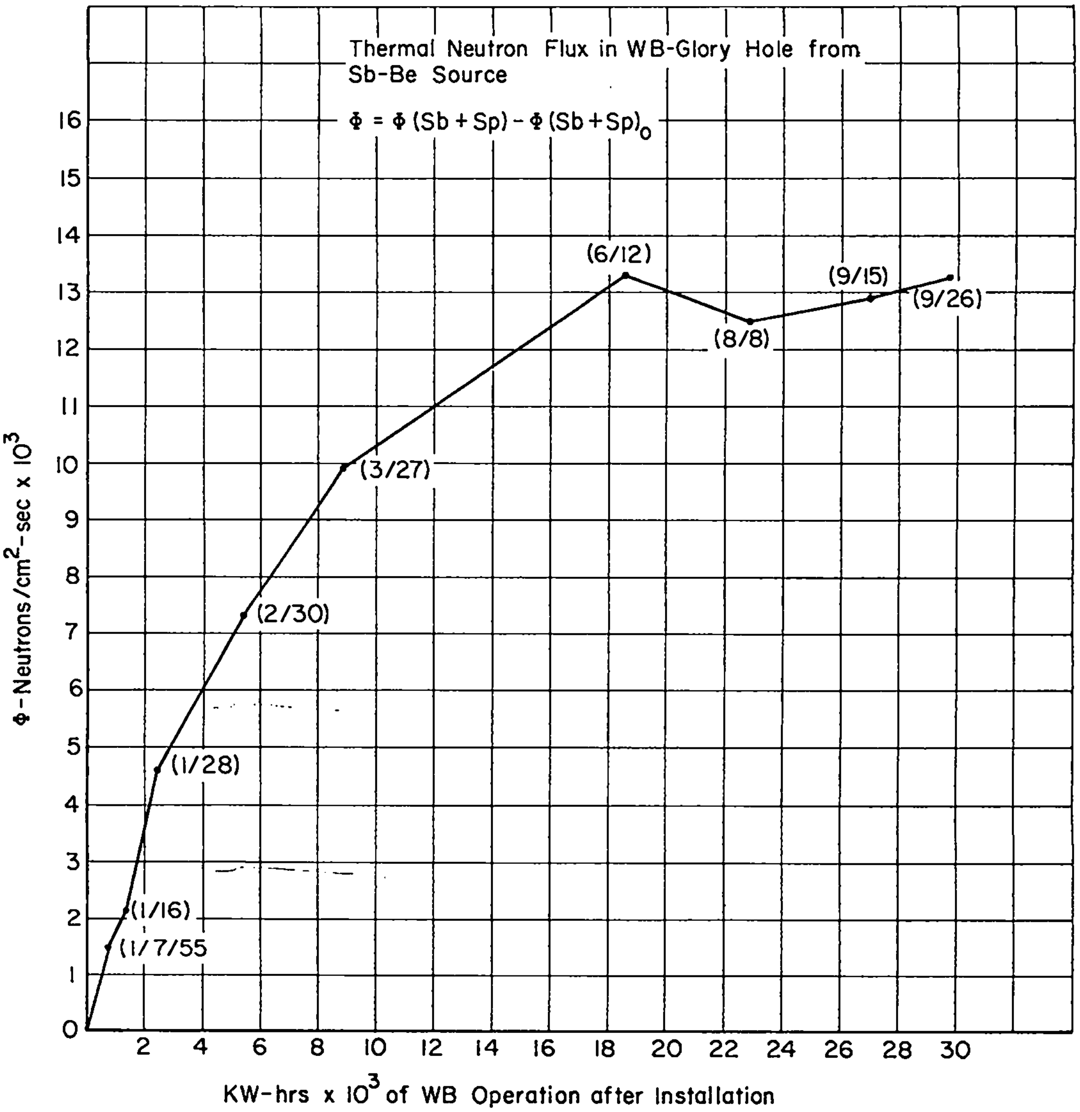


Fig. 11. Thermal Neutron Flux in WB-Glory from Sb-Be Source

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