to

Hélène

ABSTRACT

It has been suggested that the Antimony-Beryllium neutron source may provide, via prompt gamma ray spectrometry, a solution to some practical elemental analysis problems. The analysis of the sulphur content of coal samples is an important problem of this type. The difficulty of adapting a photo-neutron source to this role is discussed along with experiments bearing on how a successful adaptation may be made. The construction of a prototype and its performance are also discussed. The observation of prompt gamma ray spectra of simulated iron ore samples and sulphated coal samples indicates that considerable success can be achieved through the use of this source.

ACKNOWLEDGEMENTS

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CONTENTS

LIST OF FIGURES

PHOTOGRAPHS

I. INTRODUCTION

Thermal neutron capture by almost every nuclide is followed by de-excitation of the resulting isotope by prompt gamma emission within 10^{-14} seconds or less. The few exceptions are either fission of heavy nuclei or particle emission by light nuclei. For example, Lithium^{6} and Boron^{10} emit alpha particles, and Sulphur ^{33} may emit either protons or alpha particles. Following prompt gamma emission, the nucleus resulting from thermal neutron capture is often stable. Hence the (n, γ) reaction is the dominant effect of thermal neutron capture.

Prompt gamma rays occur in cascades in which their energies and rate of emission per neutron capture are such that the total energy of the cascade is equal to the neutron binding energy in the product nucleus. The prompt gamma ray spectrum of a nuclide is the sum of several such cascades, and the following relationships is true for prompt gamma spectra:

$$\sum_{i=1}^{n} E_{\gamma i} \; I_{\gamma i} = B$$

where $\underline{n}$ is the number of gammas in the spectrum

$\underline{E}$ is the energy of a prompt gamma

I is the absolute intensity of the prompt gamma in photon per neutron capture.

$\underline{B}$ is the neutron binding energy of the product nucleus.

The range 5 Mev to 8 Mev includes the neutron binding energies for most nuclides. The above relationship has been used by L.V. Groshev et al.[1] to determine the absolute intensities of prompt gammas from a knowledge of the observed relative intensities and the neutron binding energies. This was done substituting the absolute intensities in the above expression by the relative intensities and normalising so that the sum equals the neutron binding energy. Since the neutron binding energy and energy level structure of each nuclide is unique, observation of prompt gamma spectra is a widely applicable technique for elemental analysis.

The discovery of prompt gamma rays was published by D. E. Lea in 1934[2], only two years after the discovery of the neutron. Systematic studies of prompt gamma rays began at several of the worlds reactor laboratories soon after the end of the war freed them for peaceful purposes. The three most prominent research teams were those of

Kinsey and Bartholomew at Chalk River[3], Hamermesh and Hummel at Argonne National Laboratory[4], and L. V. Groshev et al. in Russia[1]. In spite of this early interest, the practice of prompt gamma ray elemental analysis is still far less developed than the practice of activation analysis. This is largely due to the great difference in the level of neutron fluxes available to the two opposing techniques. In activation analysis, the highest available thermal neutron flux in the reactor core may be used, since the process of irradiation is isolated from the process observing the activation gamma rays. In prompt gamma ray work the simultaneity of the irradiation and the spectrometry requires that the thermal neutron flux be channeled out of the reactor before it can be used. The formation and propagation of a neutron beam through thick reactor shielding brings about an attenuation of the thermal flux by about six orders of magnitude. Most reactor-based work on prompt gamma rays has been done with thermal neutron beam fluxes on the order of 10^6 n/cm^2-sec. using reactors with an internal thermal neutron flux between 10^{12} and 10^{13} n/cm^2-sec.

A theoretical advantage, however, strongly counters this practical limitation. If one considers one of the nuclides which is radioactive after radiative capture, the rate of emission of prompt

gamma cascades at the instant of application of a neutron flux equals the rate of radioactive decay at saturation. This follows from the formula for radioactive decay where we use for the disintegration constant of prompt gamma emission a value of 10^{14} seconds^{-1}.

$$A = N F\sigma \ (1 - e^{-\lambda T_i}) \ (e^{-\lambda T_d})$$

where N = number of atoms in the target

F = thermal neutron flux

σ =thermal neutron capture cross section of target

A = number of disintegrations of prompt gamma cascades per second

λ =disintegration constant

T_i = irradiation time

T_d = decay time

Since prompt gamma rays are observed during irradiation, $T_d = 0$, and the second exponential is unity. One nanosecond after commencement of irradiation $T_i = 10^{-9}$ sec. and $\lambda T_i = 10^5$. The first exponential is essentially zero. Hence if a nanosecond is infinitesimal relative to the experimental period, then it can be said that the rate of prompt gamma rises instantaneously with the application of a neutron flux to the value NF_σ.

The application of this result to the case of analysis for iron yields remarkable results. Determination of iron by activation analysis is unsatisfactory because only 5.85% of natural iron consists the isotopes of mass numbers 54, 58 which are non-stable after radiative neutron capture. These isotopes have small cross-sections and form long-lived products. The calculations outlined below allow the comparison of the prompt gamma intensity with the levels of radioactivity achieved after an irradiation of one year. It is assumed that the sample is of naturally occuring iron and contains 10^{22} atoms, and that the neutron flux is 10^{6} n/cm^{2}-sec.

ACTIVATION PROCESS		PROMPT GAMMA EMISSION
Fe^{54} (5.82%)	Fe^{58} (0.3%)	Fe^{56} (91.66%)
No. of atoms = 5.82×10^{20} σ = 2.9 b (F^{55}) $T_{\frac{1}{2}}$ = 2.7 y	No. = 3×10^{19} σ = 1.1 b (F^{59}) $T_{\frac{1}{2}}$ = 49 d	No. of atoms = 9.166×10^{21} σ = 2.7 b
λ = .257 y^{-1} $e^{-\lambda t} = e^{-.257y^{-1}} \times 1$ year = .773	Saturated well approximated in one year.	Saturation achieved in a matter of microseconds.
$A = N\sigma F(1 - e^{-\lambda t})$ $= 5.82 \times 10^{20} \times 2.9 \times 10^{-24} \times 10^{6} \times .227$ = 376 disintegrations/s.	$A = N\sigma F$ $= 3 \times 10^{19} \times 1.1 \times 10^{-24} \times 10^{6}$ = 33 dis./sec.	$A = N \times F$ $= 9.166 \times 10^{21} \times 2.7 \times 10^{-24} \times 10^{6}$ = 24800 cascades/sec.

The above comparison shows that the rate of prompt gamma emission by an iron sample from the moment irradiation starts is sixty times the rate of radioactive decay after one year of irradiation. T. L. Isenhour[7] has shown that if a more practical irradiation period of one hour is considered, the prompt gamma intensity from an iron sample in the externally obtainable neutron flux of 10^6 n/cm^2-sec. will equal the activation gamma counting rate of the same sample after it has been irradiated in a reactor core at a flux level 10^{12} n/cm^2-sec. Moreover, cadmium, hydrogen and gadolinium are actually more sensitively determined in the weak flux by their prompt gamma rays than in the millionfold increased flux by their activation gamma rays. Isenhour also concludes that if the flux available for prompt gamma ray work could be increased to one thousanth of the flux obtainable in activation analysis, prompt gamma ray analysis would be equally effective or more effective than activation analysis for 29 elements. These include S, Hg, Cu, Cl, K, Ni, Cr, Mo, Zr, Ti.

Use of radioisotope neutron sources in prompt gamma analysis could make possible very important applications in industry where the need for continuously available analytic facilities preclude the borrowing of often distant reactor facilities. Perhaps the most

significant of these possible applications is the continuous flow monitoring of the output quality of mining operations.
In a private communication with the Commercial Products Division of Atomic Energy of Canada Ltd. (circa 1960), the U.S. Bureau of mines predicted possible annual savings to the coal industry on the order of 20 million dollars if a rapid method of analysing the quality of coal were found. Sulphur content, which may be as high as 10%, is one of the essential indexes of coal quality, but is not rapidly determinable by present methods. One of the purposes of the present work is to determine whether or not prompt gamma ray analysis using a radioisotope neutron source is applicable to this problem.

Besides being readily available, radioisotope neutron sources have other important advantages. Since there are no geometric constraints, such as are imposed by beam formation, the sample to be analysed may occupy a relatively large irradiation volume. Large sample volumes, in the litre range, would be required by a continuous flow process, in which the material being analysed may be transported through the irradiation volume on a conveyer belt.

II. MEANS AND OBJECTIVES

The purpose of the work reported here was to obtain experimental data bearing on the adaptability of Sb-Be neutron sources to prompt gamma ray analysis. This is a photoneutron source and has some outstanding features which would make it appear very attractive for this application. An unsuccessful attempt at Atomic Energy of Canada to detect sulphur in coal by prompt gamma rays using Alpha-neutron sources motivated the present work[8]. It is hoped that the data presented here will establish the practicability of a system based on the Sb-Be source for the rapid analysis of coal, and for other analyses of practical importance.

The advantages of the Sb-Be source become apparent when one considers the distribution of the Sb gamma rays which are energetic enough to excite neutrons from beryllium. Since the (γ, n) reaction utilized has a negative Q value equal to the binding energy of a neutron in beryllium, only gamma rays with energy in excess of 1.668 Mev will excite neutrons. These gammas and their intensities are shown in the following table.

E_γ	N_γ	E-Q
1.692 Mev	.48j/dis	.023 Mev
1.90 Mev	.004j/dis	.23 Mev
2.09 Mev	.063j/dis	.42 Mev
2.26 Mev	.006j/dis	.59 Mev

Though there are neutrons formed with energies up to .59 Mev, the gammas producing neutrons with energies between 200 kev and 600 kev represent only 13% of the total intensity of the gamma rays listed above. Hence the characteristic neutron energy is only 24 kev. Since fast neutron reaction cross-section curves have a very limited area of overlapping with such a neutron spectrum, an important source of background radiation in the thermal neutron capture prompt gamma energy range is strongly reduced by the choice of Sb-Be as a source.

Another advantage of the Sb-Be source is the relatively small thickness of moderator required. A two inch of paraffin yields

a neutron spectrum which is about 95% thermal. Neutrons from an α-n source are characteristically some 200 times more energetic than Sb-Be neutrons and a greater thickness of moderator is required to obtain a good thermal-to-fast flux ratio. At the same time the slowing down of fast neutrons by inelastic scattering in various materials around the source creates troublesome background gammas. If H^1 is used as a moderator, the greater thickness required to thermalize the faster neutrons would cause a larger percentage of the neutrons produced to be exchanged for troublesome 2.2 Mev hydrogen prompt gamma rays.

Advantages shared by all photo-neutron sources are ease of fabrication, unlimited size and controllability. Because of the high penetrating power of the radiation causing neutron emission, an efficient neutron source is obtained by simply placing a beryllium block close to a capsule of irradiated antimony. Mixing is not required and the antimony is irradiated separately. Antimony sources of 1000 curie strength are produced on a routine basis at Chalk River Nuclear Laboratories and when surrounded in every direction by a 10 cm thickness of beryllium would provide a yield of 1.7×10^{10} neutrons per second.[9] The neutron production may be terminated at will by

withdrawing the antimony source into a lead container of sufficient thickness.

The disadvantage of the Sb-Be source is the large number of penetrating gamma rays emitted by the antimony. The gamma field level is similar to that of the Ra-Be source. The source described in the previous paragraph would have a field of 1100 r/hr at 1 m. A 1400 curie Ra-Be source would have about the same yield and would have a field of 1200 r/hr at 1 m. The number of gamma rays emitted per neutron produced is 4.1×10^3 and a lead shield[10] 1.8 inches thick is required to produce an attenuation by a factor of 10. These gamma rays could render low energy prompt gamma rays inobservable, and especially when scattering by the surroundings is considered, they could constitute a serious high background counting rate problem. However this problem is basically more easily solved than that of elimination of background from fast neutron reactions while using an alpha-neutron source.

III. BASIC GEOMETRICAL CONSIDERATIONS

There are countless possible arrangements in which a counter can be exposed to prompt gamma rays from a sample while being shielded from the gamma rays from the antimony source. It is necessary to start with some basic design considerations which will secure the greatest possible prompt gamma intensity with the least interference. Four such considerations explain the basic features of the Sb-Be prompt gamma facility which was built at the University of Ottawa.

The Sb source and the counter must be separated by a thick column of lead. A three inch scintillation detector 50 cm from a 10 curie source would be exposed to 1.01×10^9 gammas per second. An attenuation factor of 10^6 would be desireable to control this potential high background problem, and this would require 10.8" of lead, assuming that each 1.8" layer produces an attenuation in number of gamma rays by a factor of 10.

The intensity of prompt gammas recieved by the counter is adversely affected by the squares of three distances: the antimony source to beryllium target distance, the beryllium to sample distance, and the sample to counter distance (see figure 1). The product of these three

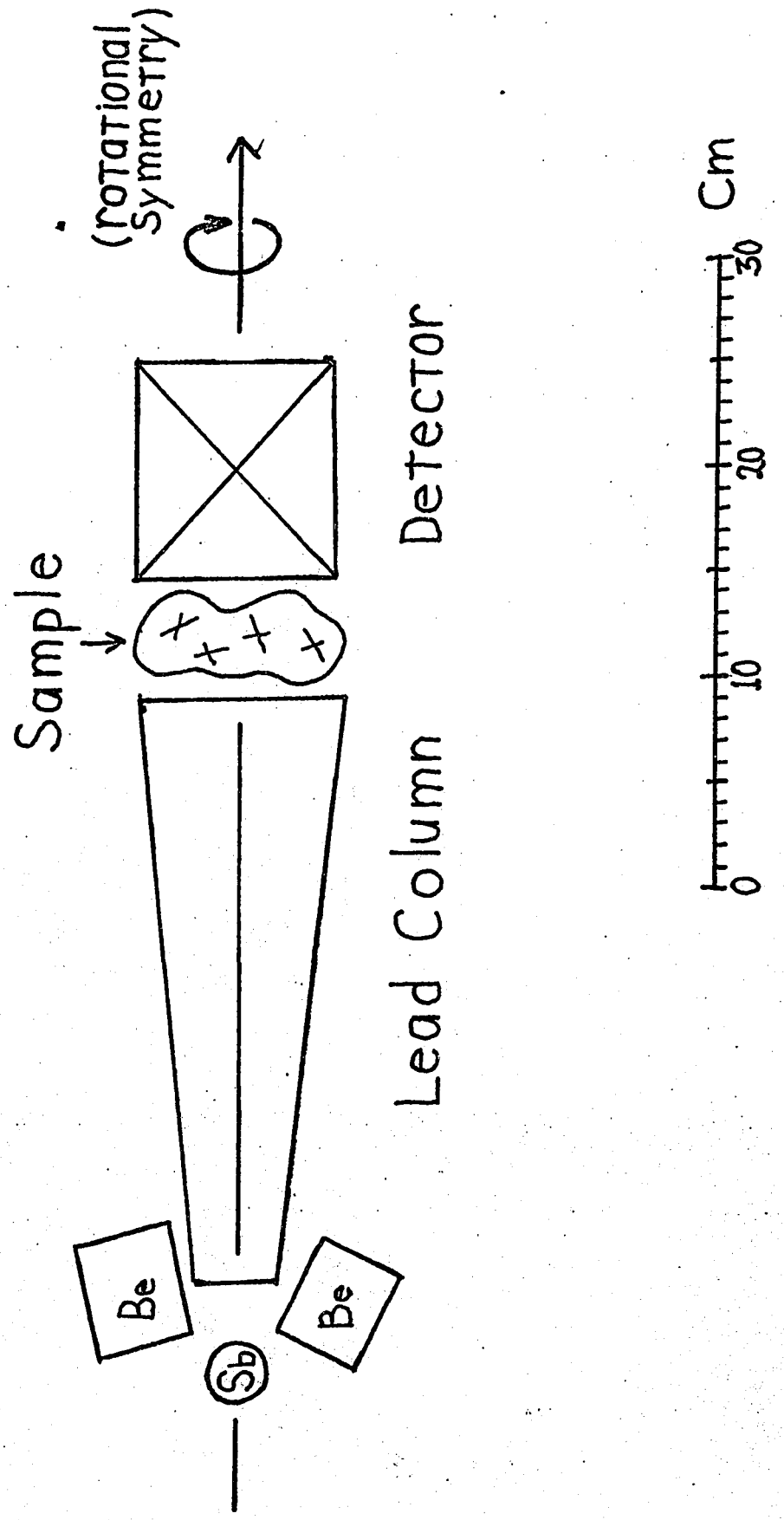


Figure 1. THE BASIC CONFIGURATION

inverse square factors can be minimised by lengthening one distance to allow the others to be short. For example, if the source is 50 cm from the counter and this distance is divided into thirds by the beryllium and the sample, then the aggregate inverse square factor is 5.14×10^{-8} cm^{-6}. However if the distances are 5 cm, 30 cm, and 15 cm then this factor becomes 1.95×10^{-7}, an improvement by a factor of four. The long distance should be the target to sample distance since neutrons, unlike gamma rays, can be channeled to a certain extent and the neutron flux at the sample will not be penalized to the full extent of the inverse square law.

The sample should be shielded from the source since it could otherwise scatter antimony gamma rays into the counter. Since at the same time it is desired to minimise the sample to counter distance, the sample should be located between the counter and the lead column. This may also carry the advantage of partial shielding of the counter from neutrons by the sample, since large sample sizes are envisaged.

With the above three considerations in mind one can easily envisage an arrangement having rotational symmetry about a source to counter axis (as shown in figure 1). Ideally this rotational symmetry should be preserved in the design of the apparatus so the path for

gamma rays from the source to the beryllium and the channel for the neutrons from the beryllium to the sample will be as broad as possible. There is nothing to be gained by restricting the antimony gamma rays or the neutrons to a beam. By avoiding a beam configuration the isotropy of gamma emission from the source and neutron emission from the beryllium can be exploited to provide a greater neutron flux at the sample.

While the above four considerations provide the basic rationale for the design of a prototype antimony-beryllium prompt gamma facility, they do not touch on a number of potential problems for which solutions must be arranged. The counter must be shielded from neutrons without seriously depressing the neutron flux in the sample. The counter should be isolated from neutron capture gamma rays originating in components of the apparatus. The entire apparatus must be surrounded by moderator, neutron absorber, and biological shield against the antimony gamma rays. These surrounding materials, having considerable bulk, will scatter a large number of antimony gamma rays towards the counter, which must be shielded against them. Even the beryllium poses problems as a potential scatterer.

IV. EXPERIMENTS ON THE SCATTERING OF ANTIMONY GAMMA RAYS

It is desired to limit the numbers and mean energy of scattered antimony gamma rays in the region of the counter to a degree compatible with counting rate limitations and detection of low energy prompt gamma rays. An experimental determination of the dependence of counting rate and mean energy of detected gammas on the scattering angle was seen as a necessary step in this direction. Accordingly, the gamma rays from an antimony source scattered from a 5 x 5 cm graphite block were observed through a collimator (figure 2) at various scattering angles. Carbon was chosen since its density and atomic number approximate that of beryllium. The total counting rate and mean energy were determined for scattering angles ranging from 20^{o} to 170^{o} in 10^{o} steps.

The aperture of the collimator was a 2.5 cm hole in a tungsten block placed 113 cm from the center of the 3 x 3" NaI(Tl) scintillation crystal. This gave a narrow angle of acceptance of 3^{o} which was necessary for measurements at low scattering angles. Wider collimators were tried in conjunction with larger carbon blocks, but these did not yield interpretable results because of increased acceptance of gammas back-scattered by the concrete wall of the cell

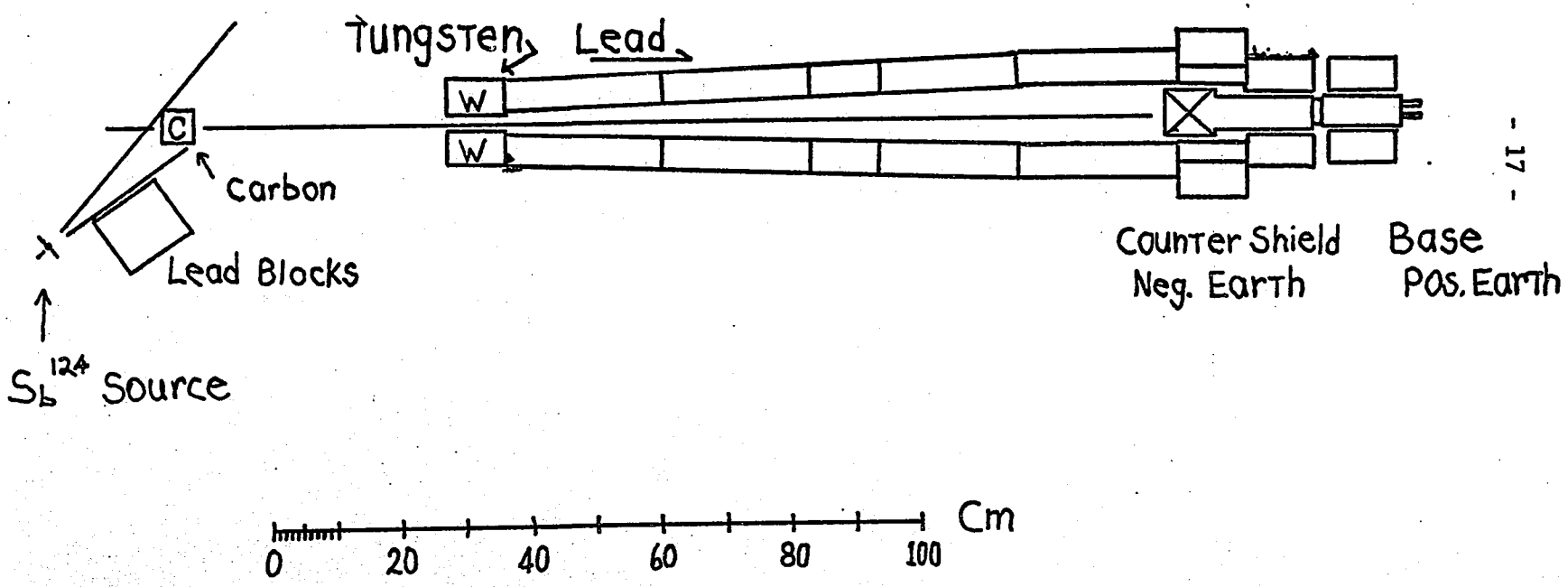


Figure 2. THE COLLIMATOR USED FOR STUDYING THE EFFECT OF SCATTERING ANGLE ON ANTIMONY GAMMA RAYS

approximately 8 feet from the collimator aperture. It was also found that when a wide aperature (5 to 7 cm) was formed using lead blocks the inside walls of the aperature were scattering gamma rays directly from the source to the counter. The narrow collimator, in addition to limiting interference, provides a more precise specification of scattering angles and allows the use of a smaller carbon block in which self-shielding is kept to a minimum.

For each scattering angle the complete spectrum of the radiation scattered by the carbon block was recorded during a 2 min. counting period with an 18.45 mc Sb^{124} source 28 cm from the center of the carbon block. Then the carbon block was removed and the multi-channel analyser was placed in subtract mode and the background count was taken for an equal period of time. The resulting spectrum is believed to contain only a very small percentage of extraneous gammas. Sample spectra are shown in figure 3. Since the Tullamore ST-200D analyser used did not have a functioning automatic output the number of counts in the peak channel, the peak channel number, and the channel number representing the highest energy of scattered gammas received were the only items of digital information recorded. The spectrum was recorded using sheet-film camera mounted at a fixed point in front of the visual output of the analyser. The 840 kev peak provided by an

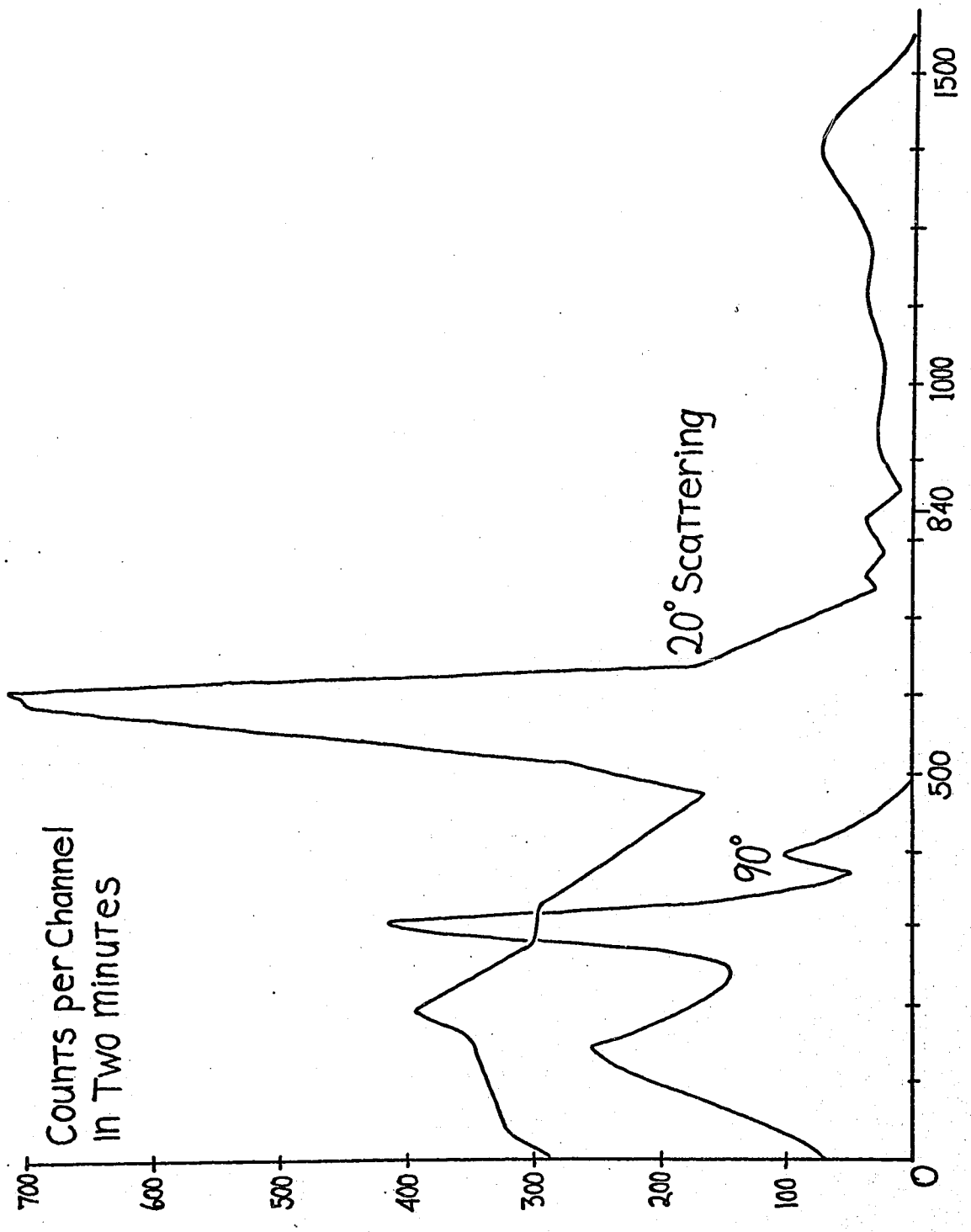


Figure 3. SPECTRA OF ANTIMONY GAMMA RAYS SCATTERED THROUGH 20° AND 90°.

Mn^{54} source was always observed along with the carbon block spectrum to insure energy calibration against possible drifting. Possible uncertainty in energy assignments due to non linearity was found to be about 2% by testing the counter with Co^{57}, Ba^{135}, Am^{237} and Sb^{124} sources.

The integrals involved in finding the total number of counts analysed and their main energy were solved by the expedient of "weighing the photograph" of the spectrum. The photographs of the spectra were printed on 8 x 10" photographic paper and the print was cut out along the spectrum curve and along the zero-count line. The resulting cut-out spectrum was then weighed to one ten-thousanth of a gram. It was then necessary only to know the number of counts corresponding to one gram of paper, which depends on the area density of the paper and the total number of counts represented by one cm^2 of paper. The latter figure is the vertical scale of the photograph in counts per centimeter, easily found from the number of counts in the peak channel, times the number of channels per cm on the horizontal scale. The mean energy of the spectrum was found by exploiting the equivalence between the vertical line representing the channel of mean energy and the vertical line passing through the center of gravity of the cut-off. Since the photographic paper used was quite stiff it was a simple matter to accurately locate the center of gravity of the cut-out by balancing

it on a knife edge. The validity of this method is verified by the similarity between the integrals for mean energy and center of gravity of a curve on a flat sheet of constant area density.

$$\overline{E} = \frac{N(E)dE}{N} \qquad \chi_{c.g.} = \frac{\sigma \int h(x)dx}{M}$$

It was desired to express the counting rate results as the number of counts per sec. received at a given scattering angle by a 3 x 3" crystal per cc of carbon for a 1 curie source with the distance as described. It is thus necessary to apply normalizing factors for counting time, dead time, sample volume, and source strength. The normalization for sample volume is complicated by the fact that the narrow cone of full acceptance (inside which a small source sees the whole counter) does not include whole lateral extent of the carbon block. In order to find the effective volume of scatterer seen by the counter the carbon block was considered to consist of cylindrical annuli of radius r measured from the collimator axis, the volume of which were weighted by the percentage acceptance at distance r from the axis. The percentage acceptance as a function of r was found by placing a 100 μc Sb source at stations $\frac{1}{2}$ cm apart along a line perpendicular to the collimator axis. (see figure 4) The total counting rate was measured at each point.

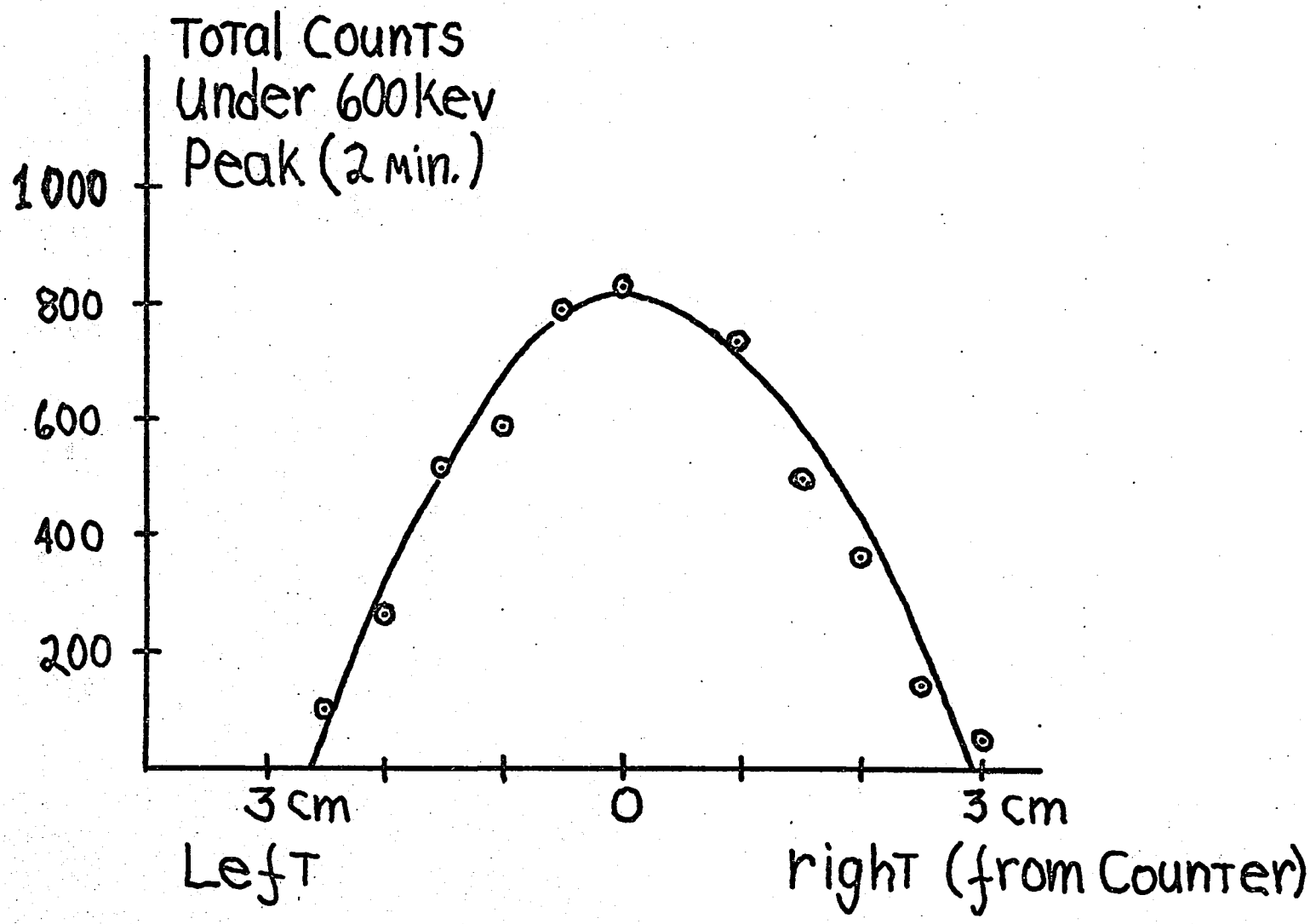


Figure 4. THE ACCEPTANCE PROFILE OF THE COLLIMATOR

Table - Normalization of Results

Strength of source	18.45 mc	
Counting time	120 sec	
Effective scattering volume	59.4 cm^3	
Normalization factor for the above	131.6	(divide total counts by this to get normalized counts per second)
Dead time	125 u sec	
Maximum counting rate (20^o)	296 cps	
Live time (%) at maximum rate	96.3%	

Results for Scattering from a Carbon Block

Angle	Total Counts	Normalized Counting Rate	$\bar{E}$ Mean Energy	Cut-off Energy	$\bar{E}$ C Mev. cps
20^o	3.55×10^4	280 cps	463 kev	1490 kev	130
30^o	2.90×10^4	227 cps	408 kev	1268 kev	92.6
40^o	2.36×10^4	184 cps	367 kev	1084 kev	67.5
50^o	1.89×10^4	147 cps	321 kev	870 kev	47.2
60^o	1.55×10^4	121 cps	298 kev	764 kev	36.0
70^o	1.29×10^4	99.0 cps	257 kev	635 kev	25.4
80^o	1.19×10^4	91.8 cps	244 kev	504 kev	22.4

Angle	Total Counts	Normalized Counting Rate	$\overline{E}$ Mean Energy	Cut-off Energy	$\overline{E}$ C Mev. cps.
90^{o}	1.08×10^{4}	83.5 cps	214 kev	482 kev	17.8
100^{o}	1.04×10^{4}	81.1 cps.	199 kev	404 kev	16.1
120^{o}	1.26×10^{4}	97.2 cps	191 kev	390 kev	18.6
140^{o}	1.20×10^{4}	91.3 cps	174 kev	318 kev	15.4
150^{o}	1.18×10^{4}	89.8 cps	168 kev	303 kev	15.6
160^{o}	1.16×10^{4}	100 cps	161 kev	288 kev	16.1
170^{o}	1.30×10^{4}	94.5 cps	148 kev	265 kev	14.0

The normalized counting rate is the number of gamma rays detected by a 3 x 3" NaI(Tl) counter originating from a 1 curie Sb source and scattered into the counter by 1 cc of carbon located 28 cm from the source and 155 cm from the center of the counter.

The random uncertainty in the total counts values is due mainly to non-uniformity of the photographic paper and imprecise location of the antimony source. By weighing several portions of the paper smaller in area than the spectrum "cut-offs" the uncertainty due to non-uniformity was assessed at approximately 3%. The antimony source was contained in a capsule which was easily handled with simple equipment

only if unopened, and the source was free to move approximately $\frac{1}{2}$ cm from the center of the capsule. Since this uncertainty exists in a square law term, the contribution to the overall uncertainty is on the order of 4%. Besides the 7% random uncertainty accounted for here, there is a systematic uncertainty of perhaps 10% in the calculated "weighted" volume of the scatterer.

The random uncertainty in mean energy values is due to paper non-uniformity, counter non-linearity, and uncertainty in the balance point determination. The first two have been previously quoted as 3% and 2%. The balance points were determined with approximately 2% accuracy. Again the total random uncertainty accounted for is 7%.

The last two columns in the table of results of the scattering experiment are the most useful in drawing conclusions bearing on the design of a prompt gamma spectrometer. The cut-off energy is the energy corresponding to the highest channel in the background-free spectrum to show some counts. It represents the minimum energy which a prompt gamma would need in order to be observed separately from antimony background, assuming that the antimony background would be reaching the counter scattered through a specified angle. The last

column gives the product of mean energy and normalized counting rate. This is a figure of merit bearing on the amount of counter shielding required to reduce the background counting rate to within acceptable limits. It will thus be seen from the table that the gamma rays scattered through a small angle will necessitate much more shielding than will those scattered through large angles. The dependence of shielding requirements on scattering angle is probably much stronger than the dependence of $\overline{E}$ C on the same, because the predominent shielding mechanism in the energy range involved, for heavy-element shields, is the photoelectric effect. Photoelectric absorption varies as the -7/2 power of energy.

Consideration of the cut-off energy and $\overline{E}$ C curves (see figure 5) reveals a very useful rate of drop-off in these values as the angle increases up to about 90^{o}. After this point the rate of decrease in both cases is considerably less. From the practical viewpoint of controlling the background of scattered gamma rays reaching the counter by eliminating small scattering angle paths to the counter, a point of diminishing return seems to be reached at about 90^{o}. It is important not to go beyond this point since "elimination of scattering" paths refered to requires the use of massive lead or bismuth shielding.

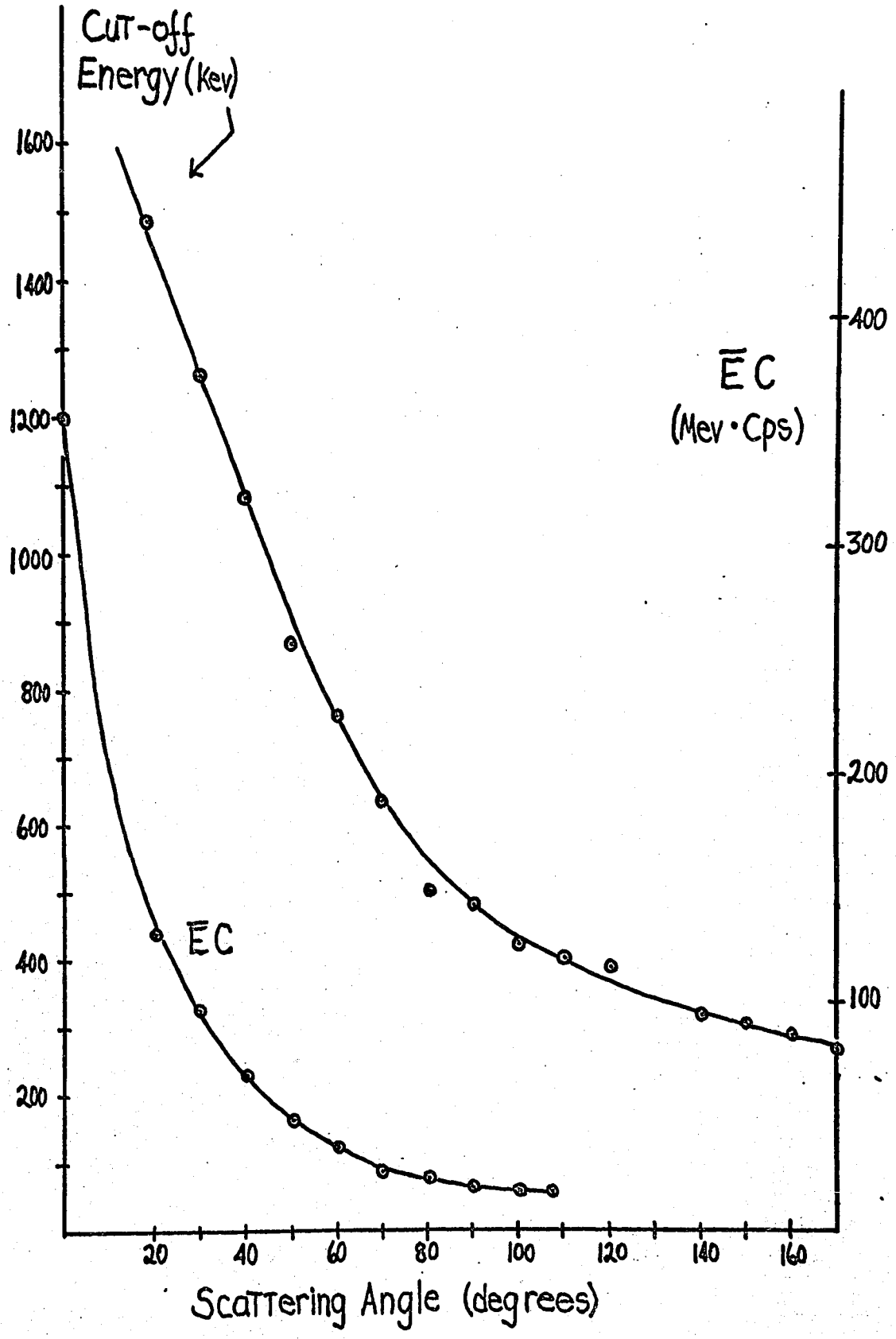


Figure 5. DEPENDENCE OF ANTIMONY GAMMA SPECTRA CUT-OFF ENERGY AND MEAN ENERGY-COUNTING RATE PRODUCT ON SCATTERING ANGLE.

Also the accessibility of Be blocks to the direct gamma rays must not be limited. Hence the practical conclusion of the scattering experiment with respect to the design of a prompt-gamma spectrometer may be stated as follows: Any material located as that it will scatter gamma rays from the source into the counter at an angle of less than 90^{o} should be separated from the counter by adequate shielding.

It has been earlier mentioned that a 10.8" lead column, or equivalent, would be needed between the course and the counter to control the exposure of the latter to direct gamma rays. If this shield were widened at the end towards the counter to form a broad cone, then all gamma rays scattered towards the counter through less than 90^{o} would be attenuated by a thickness of lead which decreases as the scattering angle increases. Such a cone shield would seem to provide an effective means of achieving the aim set out at the end of the last paragraph. In the diagram depicting this arrangement, a circle having the source-counter axis as a diameter has been drawn in. Any points inside this circle automatically forms an angle of less than 90^{o} with respect to the counter and the source. The circle therefore helps to indicate the required width of the cone shield.

The scattering experiment results indicate that if a shield such as described above is provided the counter will be exposed to a gamma ray spectrum reduced in amplitude, and having a mean energy of between 170 kev and 220 kev. To shield against this, the counter will have to be surrounded by a light shield. The rate of attenuation of gamma rays of this spectrum in lead is determined in the experiment which follows.

Using approximately twenty 6 x 12 x 24 cm lead blocks a pyramid shaped shield approximately the desired wide-cone shield was constructed immediately adjacent to one of the thirty inch thick concrete walls of the radiation cell in which the scattering experiment was done. The source and counter were 60 cm apart and separated by a thickness of 30 cm of lead. The counter was enclosed in a 1/8 inch copper tube on which five layers of 1/8 inch thick lead sheet were rolled. This arrangement is shown in figure 6. With this much shielding around the counter and radiation back-scattered from the cell wall behind the counter dominates the spectrum seen by the counter unless the "window" at the back end of the copper tube is blocked off with adequate shielding.

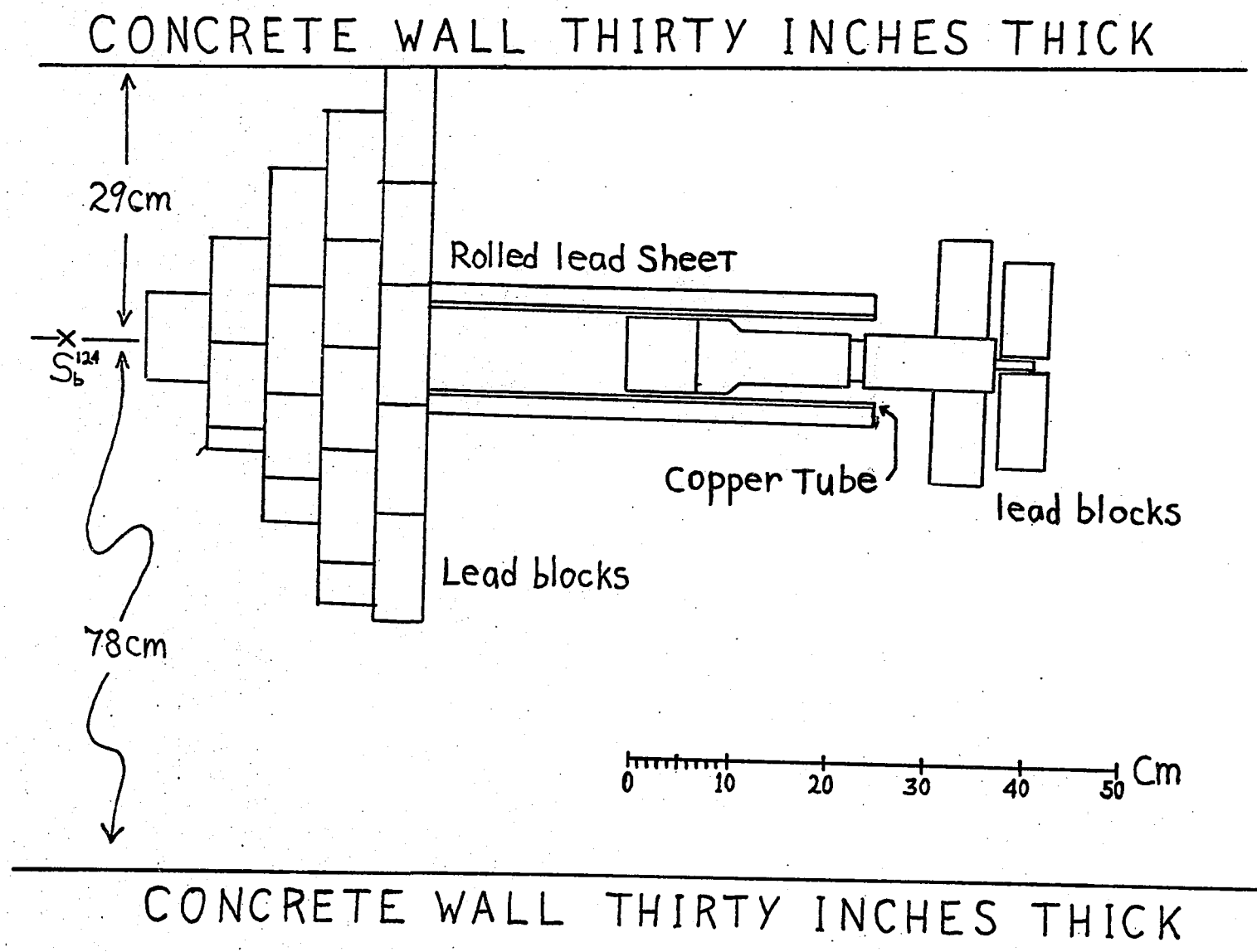


Figure 6. THE EXPERIMENT ON SECONDARY SHIELD REQUIREMENTS.

The spectra received by the shielding counter were observed with the number of lead sheets on the copper tube varying from five to one.

An Mn^{54} source was again included to verify the energy scale, and its spectrum removed by counting in subtract mode. With no lead sheet on the copper tube the spectrum was distorted and the Mn^{54} peak had only approximately one thousanth of its normal amplitude. The full spectrum of gamma rays scattered into the area behind the pyramid shield could thus not be directly observed since the analyser appeared to become overloaded at a fairly modest counting-rate. The strength of the antimony source was 4.93 mc.

The counting rates for the five different thicknesses of shielding have been normalised to the values that would have been obtained with a ten curie course and the logarithms of the normalised values have been plotted (figure 7) as a function of number of 1/8 inch lead sheets. If the plot is extropolated to zero thickness of lead, the counting rate which was observed to block the analyser is indicated as 9×10^6 cps for 10 curies or 4×10^3 for the source actually used. At this rate live time in the Tullamore ST 200-D analyser is only 50%

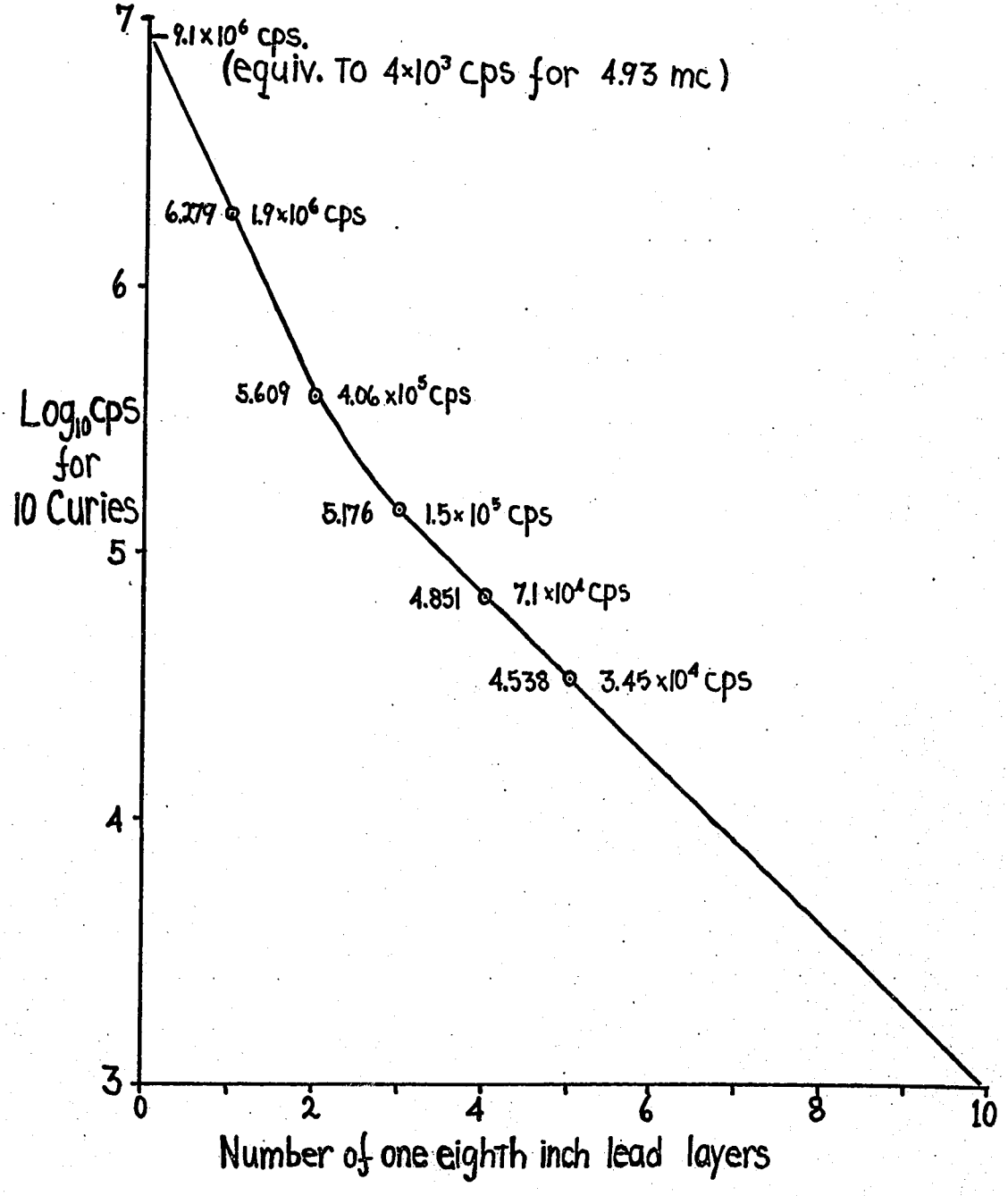


Figure 7. ATTENUATION IN THE SECONDARY SHIELD.

of clock time (dead time constant at 125 micro-seconds). On the basis of this evidence it was decided to design the two shields (primary or pyramid shield and secondary or counter enclosure) for a total background from direct and scattered antimony gamma rays of approximately 2000 cps for a 10 curie source. This was envisaged as being 50% direct penetration through the axis of the pyramid shield and 50% penetration of scattered gamma rays through the secondary "light" shield surrounding the counter.

The average cut-off energy of the spectra seen through the lead sheets is 545 kev, which corresponds on the plot of cut-off energy as a function of scattering angle to an angle of 85%. This is in good agreement with the intended 90^{o} minimum scattering angle. The mean energy is 220 ± 20 kev, which agrees well with the 214 kev mean energy observed for 90^{o} scattering.

An estimate of the thickness of the secondary shield surrounding the counter was obtained using the attenation plot derived from the present experiment. It is necessary to assume that the scattering from the concrete walls of the cell is similar to the scattering that will occur is the antimony-beryllium prompt gamma facility. If the attenation

plot is extropolated down to the desired counting rate or 1000 cps, the corresponding thickness of lead is 1.25".

The design features considered so far are summarized in the following diagram. This conceptual diagram (figure 8) of an antimony-beryllium prompt gamma facility is an extension of the previous one in the light of the two antimony gamma ray scattering experiments. The sample is placed inside the secondary shield so that it also is shielded from scattered antimony gammas (double scattering could occur) without the counter being shielded from the sample. The secondary shield should thus be made of bismuth so that it will not reduce the neutron flux at the sample. The advantage of making the secondary shield a closed box around the sample and counter springs from the choice of bismuth as the shielding material. In effect part of the lead primary shield is thus replaced by bismuth which produces far fewer prompt gammas and which will reduce the lead prompt gamma intensity both by its shielding value and causing the counter to be farther from the lead.

The large volume enclosed by the secondary shield not only allows the use of large samples, but also serves to attenuate the field of gammas due to the action of the neutrons on the bismuth.

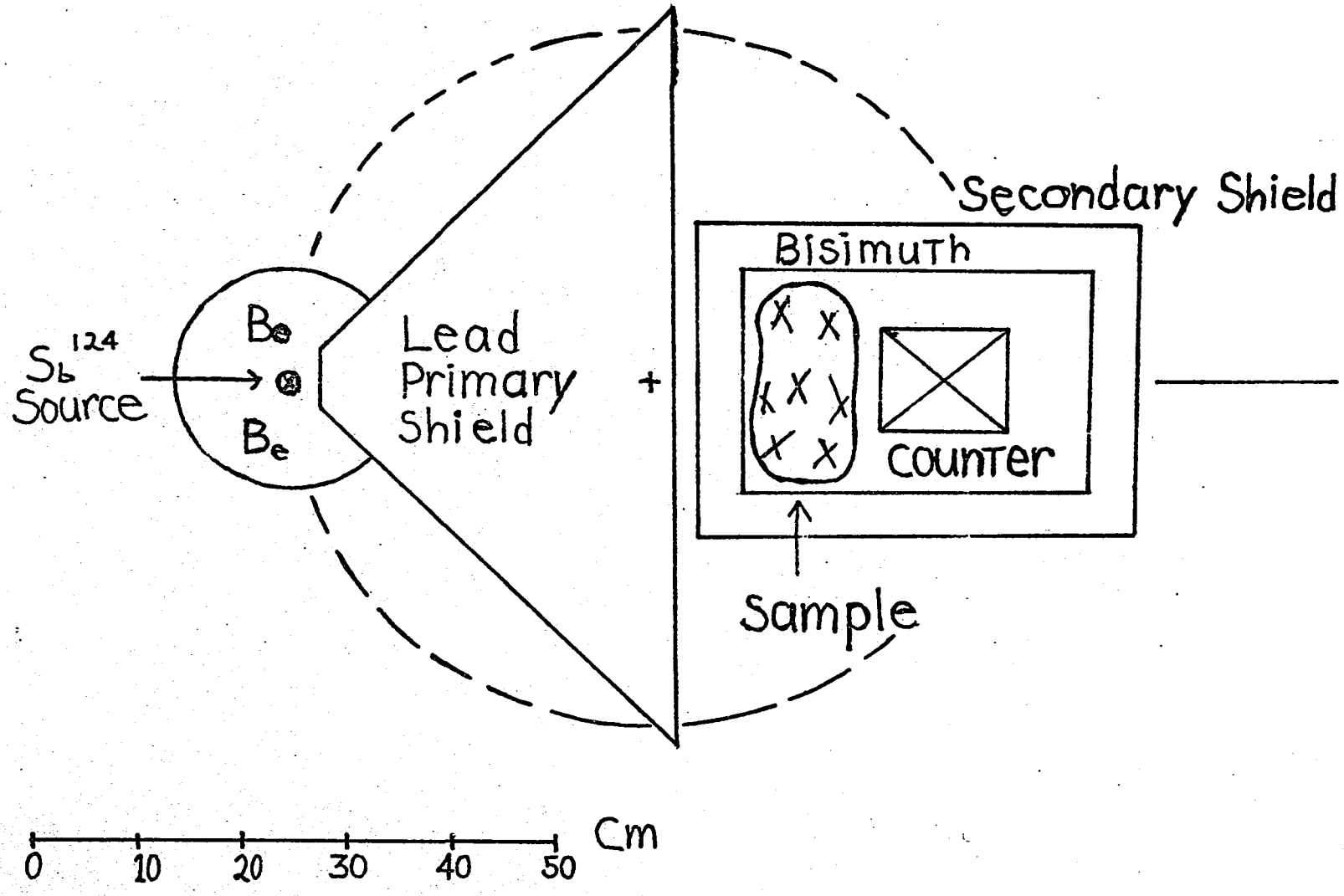


Figure 8. "CONCEPTUAL DIAGRAM" OF AN ANTIMONY BERYLLIUM PROMPT GAMMA FACILITY.

V. DESIGN OF AN "IDEAL" PROTOTYPE

The handling of neutrons has so far been discussed only with reference to the bismuth secondary shield and to desireability of lengthening the neutron path in favour of short gamma ray paths. The neutron must be thermalized and confined. Stray (n, γ) reactions must be kept to a minimum, especially near the counter. Excessive activation of the surroundings and health hazards must be avoided.

Ordinary hydrogen was chosen as the moderating element, since this element will allow the greatest flexibility for alterations and at the same time require the least thickness of moderating layer. Since the hydrogen has a relatively large absorption cross-section for thermal neutrons (332 mb), the 2.22 Mev prompt gamma emitted by a large quantity of hydrogen can be expected to be a major background feature. A compromise must be made between the flux advantage of having an uninterupted moderator envelope and the background advantage of eliminating hydrogen from the area of the counter.

Since a scintillation crystal is readily activated, the counter enclosure will have to be shielded against neutrons. Since the isotope

Li^6 combines a very high alpha emission cross-section (945 b) with a very small radiative capture cross section, the separated isotope is ideal for compact non-background-producing counter shielding. Eliminating the more abundent (91.7%) Li^7 reduces the required thickness by a factor of 13.3 and eliminates the beta emitted during decay of Li^8 (33 mb). Mounting the Li^6 on brass plates provides shielding from the alpha particles, and also from 90 kev bismuth x-rays.

The entire apparatus must be surrounded by an efficient neutron absorber. Since this will be far from the counter, boron can be used as the absorbing element in spite of its prominent prompt gamma rays. A boron compound suspended in a hydrogen rich matrix would be an ideal neutron barrier.

The "ideal" prototype is an application of the above considerations to the design concepts previously discussed and is depicted in figure 9. This drawing is an extension of the previous "conceptual diagram".

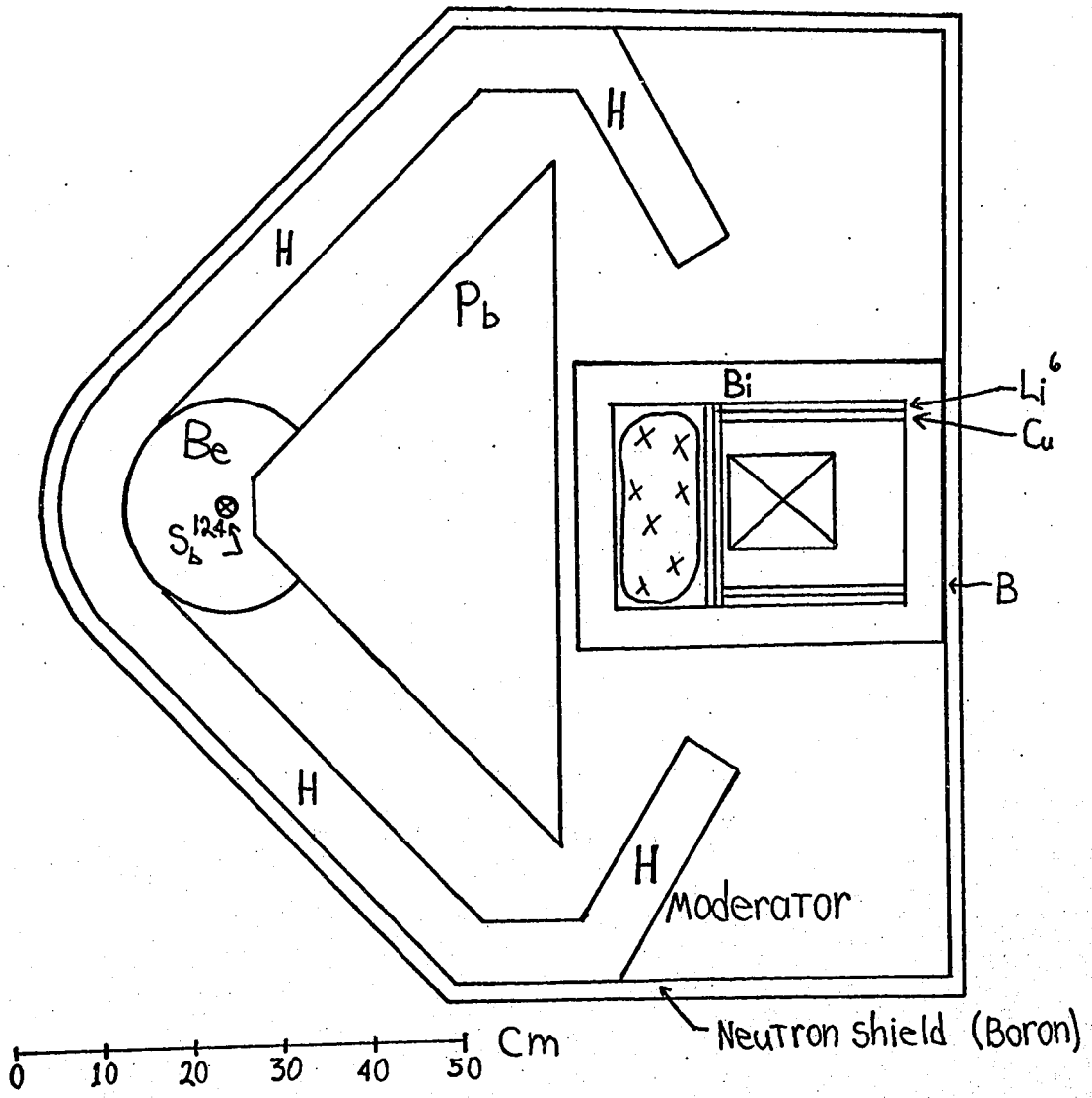
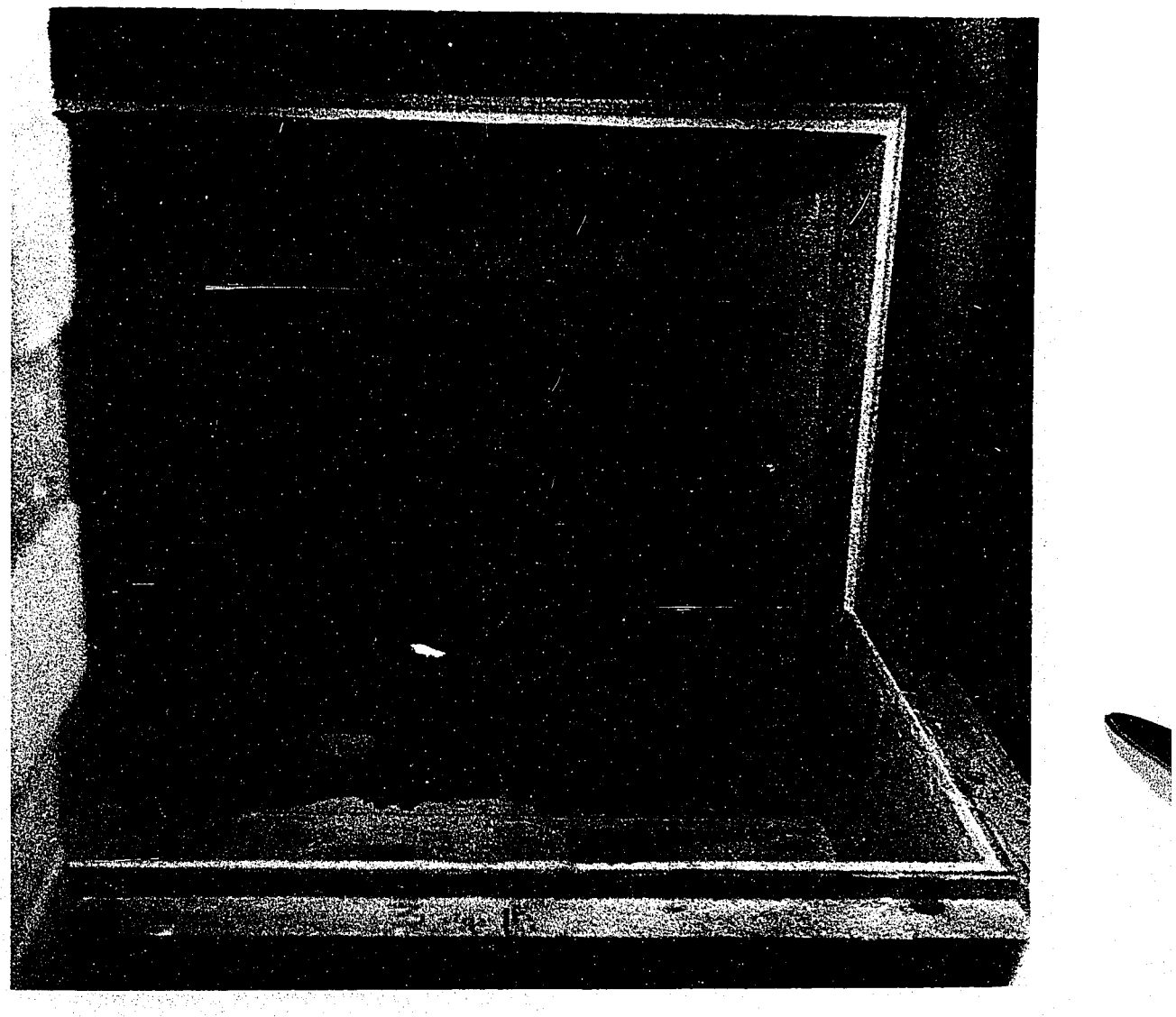


Figure 9. AN "IDEAL" PROTOTYPE.

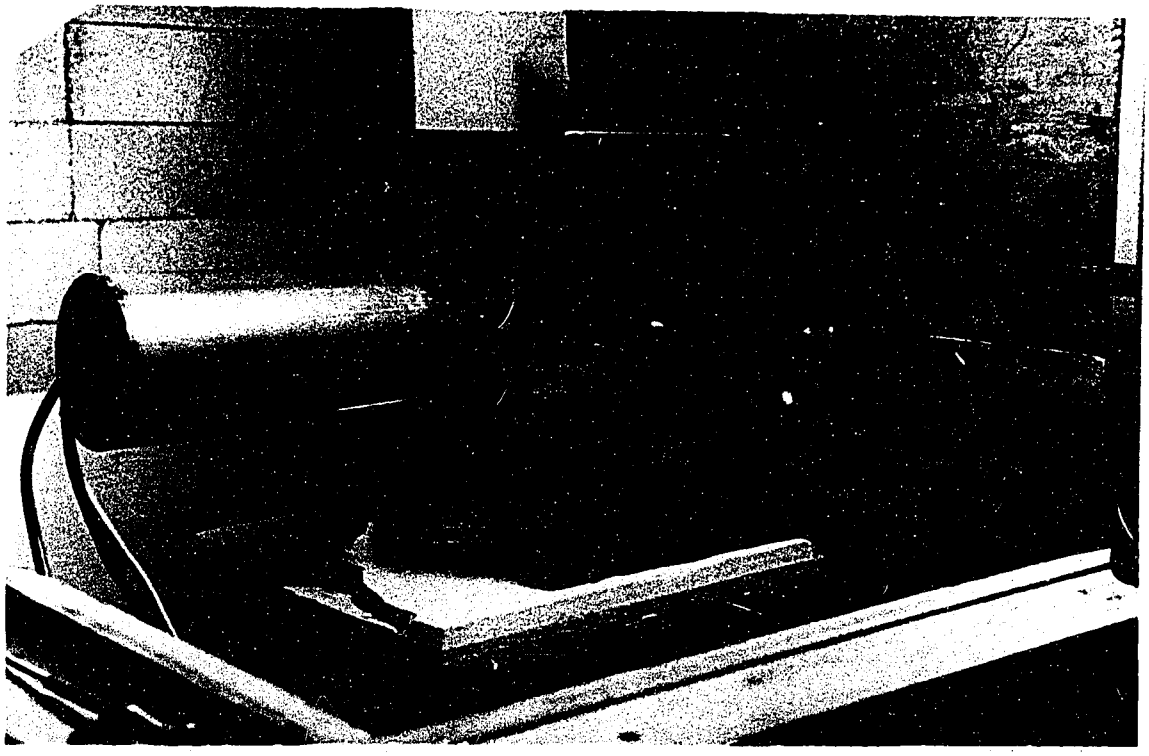
A PRACTICAL PROTOTYPE

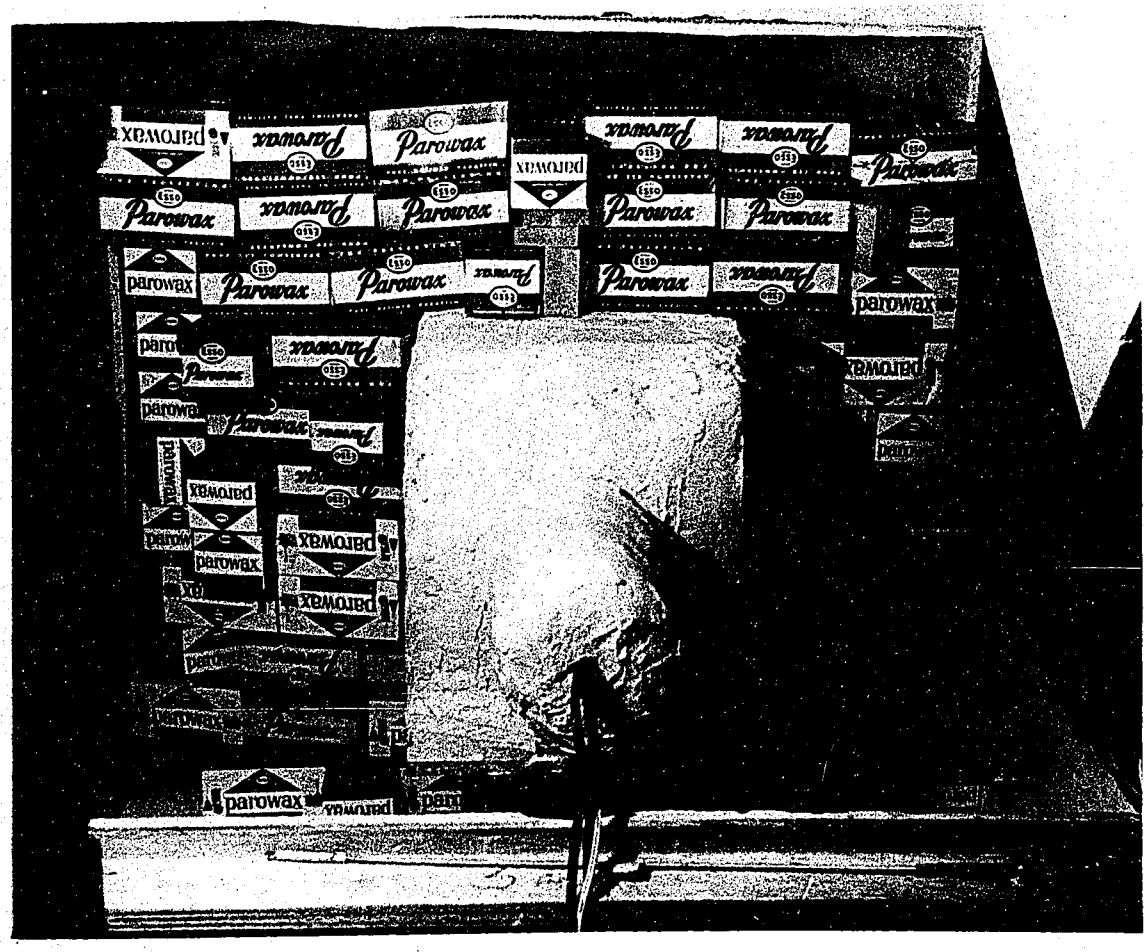
Since the secondary shielding as shown in the "ideal prototype" drawing would weigh on the order of 180 kg, it was not found practical to use bismuth throughout. The neutron transparency of bismuth is most advantageous around the sample. Hence the secondary shield was divided into two parts: a bismuth sample container and a lead counter enclosure (see photograph 2 and figure 10). The lead enclosure was covered on the outside by a thick layer of boron-based neutron absorber, as shown in photograph 3).

The primary shield has been conceived in relation to a compact round source. In practice the available antimony sources are elongated, measuring 1" in diameter by 5" in length. The primary shield should thus not be a perfect rotational form, but should be widened in the direction of the source axis, which in the present prototype is horizontal. Other concessions to practicality are motivated by a need to simplify the casting of the lead and the physical support of the resulting shield, which in final form weighed in at 412 kg. Thus the primary shield of the prototype is a trapezoidal prism. In order to obtain maximum use of the laboratory floor for support, the source-counter axis was placed horizontally and one of the slanted faces of the primary shield was redesigned to form a horizontal base (see figure 11). The major part of the shield was then cast as a series of flat trapezoid plates, the slanted upper portion being cast as a separate prism form (photograph 1).



Photograph 1





Top: Photograph 2, Bottom: Photograph 3.

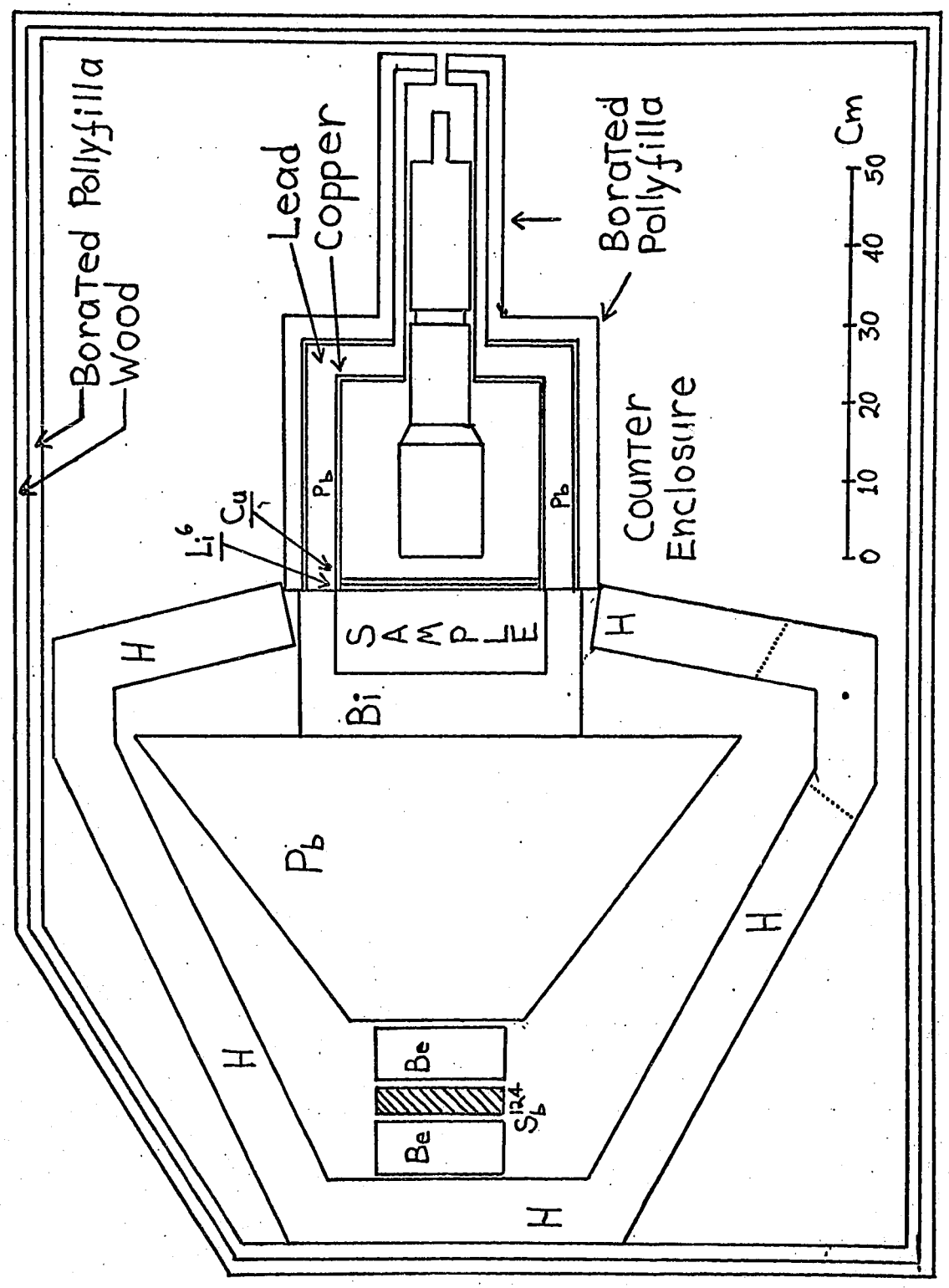


Figure 10.. PLAN VIEW OF THE PROTOTYPE AS CONSTRUCTED.

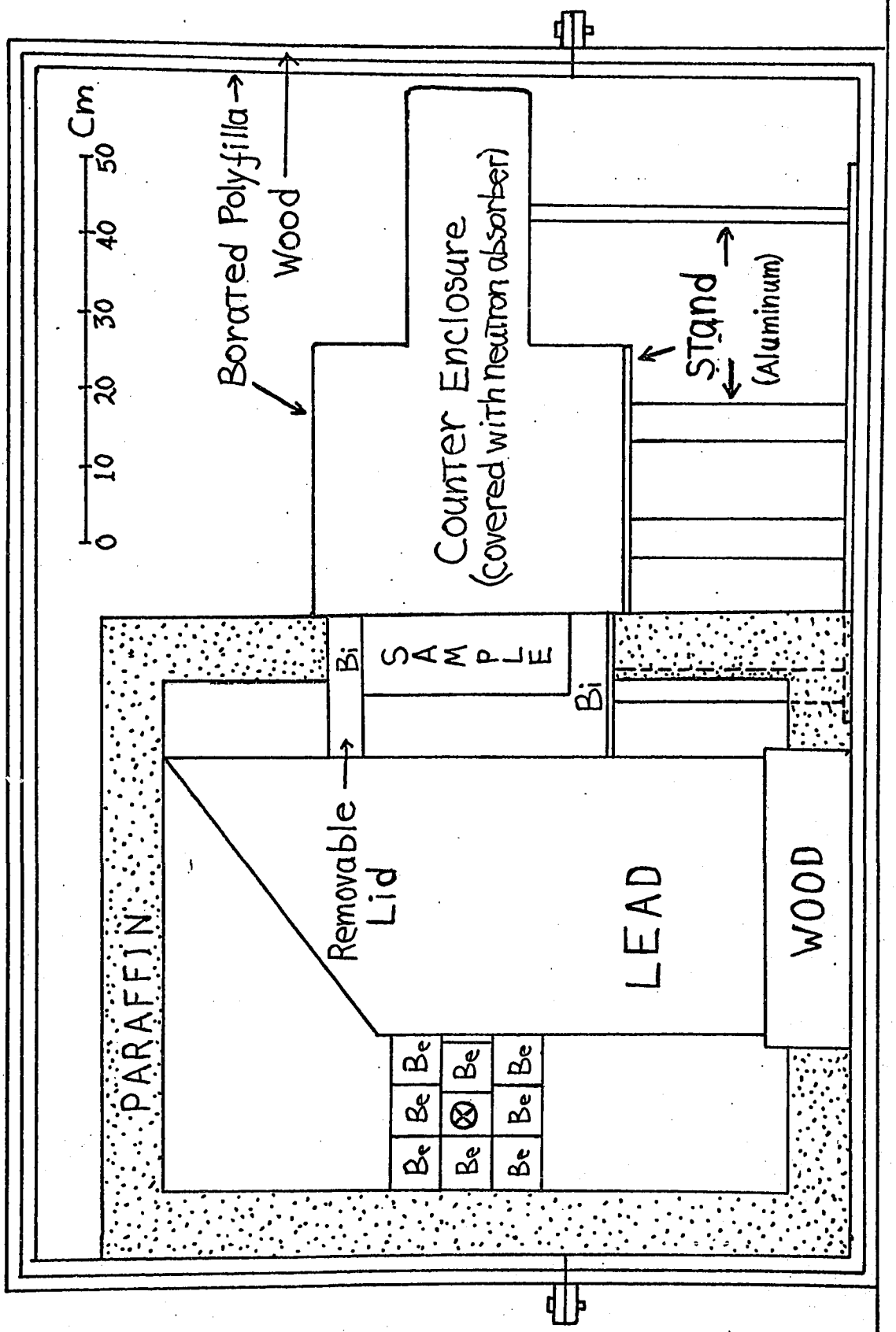


Figure 11. VERTICAL SECTION OF PROTOTYPE.

Beryllium was available in the form of rectangular blocks 2" x 2" x 5" in size. These were packed as compactly as possible around the antimony source position. If the characteristic thickness of the beryllium assembly as a target for antimony gamma rays is taken as five cm. the neutron yield could be expected to be 10^7 n sec/curie[9].

The most convenient form of hydrogen-based moderator is paraffin in the small consumer-type boxes. These can be readily stacked in any arrangement desired. To assemble the moderator as shown in the diagram (figure 10) 270 lbs of paraffin were used.

The neutron absorber was prepared as a plaster-like mixture of boric acid, and polyfilla, a wall filling compound. A one-half inch thick coating of this was applied to the inside of the large plywood box designed to contain the whole experiment. Approximately 60 lbs of boric acid and 90 lbs of polyfilla were used giving a B^{10} area density of 14.6 mg cm^2.

THE RADIOLOGICAL FACILITY

The prototype constructed as described in the previous section is complete and operable in all respects except for the safe handling of a strong antimony source. Provision must be made for safe storage of the antimony away from the beryllium, its rapid transfer to and from the operational position, and the reduction of the gamma ray fields in the laboratory to safe levels.

Safe storage of the source was secured by means of a 1343 kg "Junior J-Rod" shipping container on loan from Atomic Energy of Canada. The end plug of this lead flask was modified in order to allow passage of the source through the plug and into a guide tube which attaches unto the plug. The guide tube mates with a 1" diameter hole bored through the 9" of lead of the plug and with the hole forms a channel through which the source is drawn to the operational position. The flask was laid on its side, as shown in figure 12, so that this channel would be horizontal and close to the floor (16.5"). A carriage was designed to hold the flask in this position and allow it to be rolled away on rails during shut-down for alterations to the apparatus.

When the flask was initially loaded at Atomic Energy of Canada

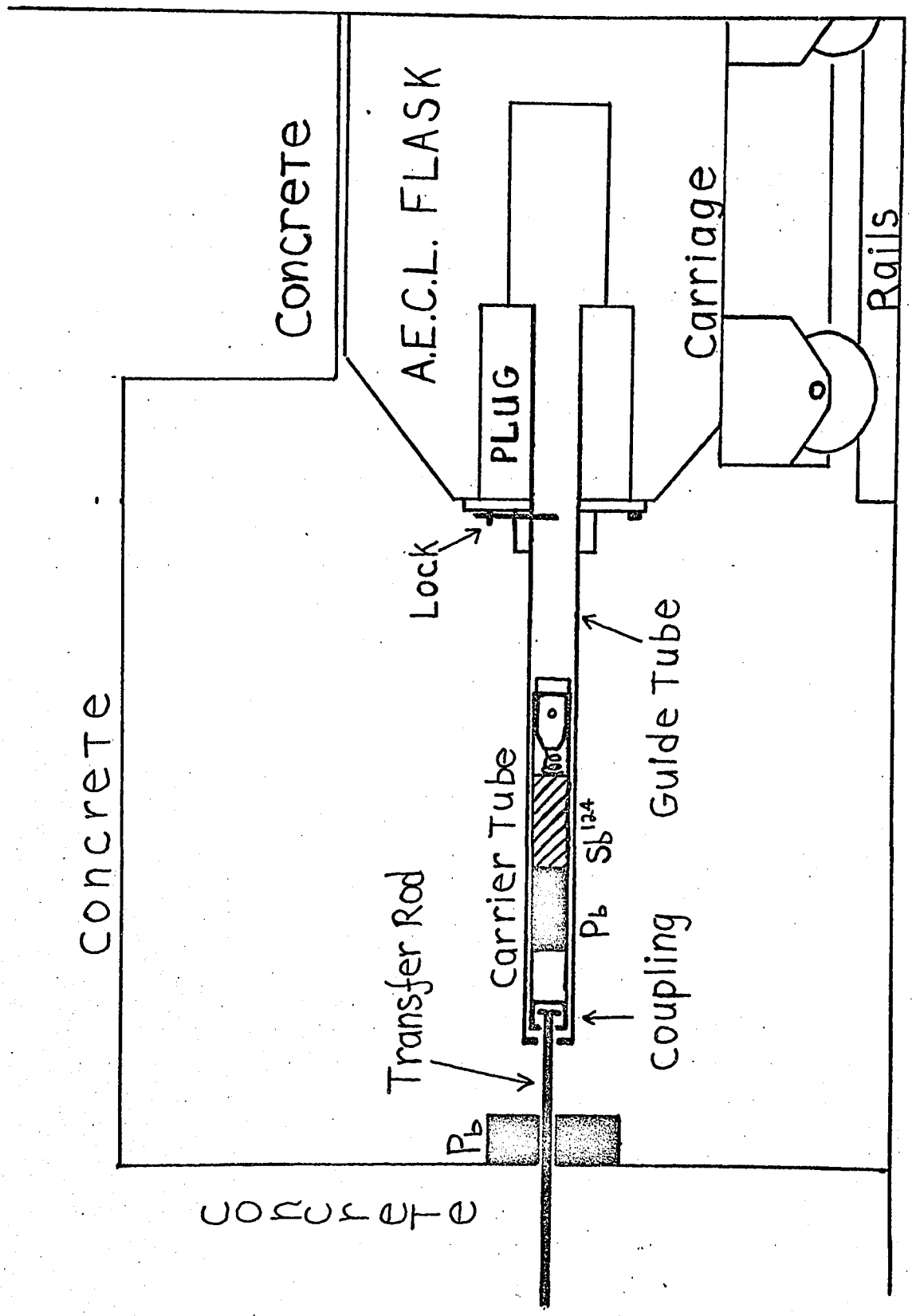


Figure 12. SECTION DETAIL OF SOURCE HANDLING ARRANGEMENT

the source was not placed directly into the flask. If so it would have been impossible to withdraw the source from the flask at the site of the experiments. Instead the source was slipped into an 18" long carrier tube designed to slide freely through the guide tube. The open side of the carrier tube was then closed with a spring loaded bayonnet type plug designed to be handled by remote manipulators. The other end of the carrier tube had been fitted with a coupling designed to engage with a "T" on the end of an 11 foot long transfer rod. With the flask in operational position at the experiment site the source could be withdraw from the flask by inserting the transfer rod into the guide tube and causing the end of the transfer rod to engage the coupling on the carrier tube. The position of the source can then be quickly controlled by pushing or pulling on the transfer rod, (figure 13).

The "biological" shielding was provided by housing the experiment and access area in a concrete structure with walls 31" thick (figure 14). Two thousand six hundred bricks were used, the total mass of concrete being 41,800 kg. The results of a gamma ray dose rate survey with a 4.5 curie source in operational position are shown on the diagram of the biological shielding.

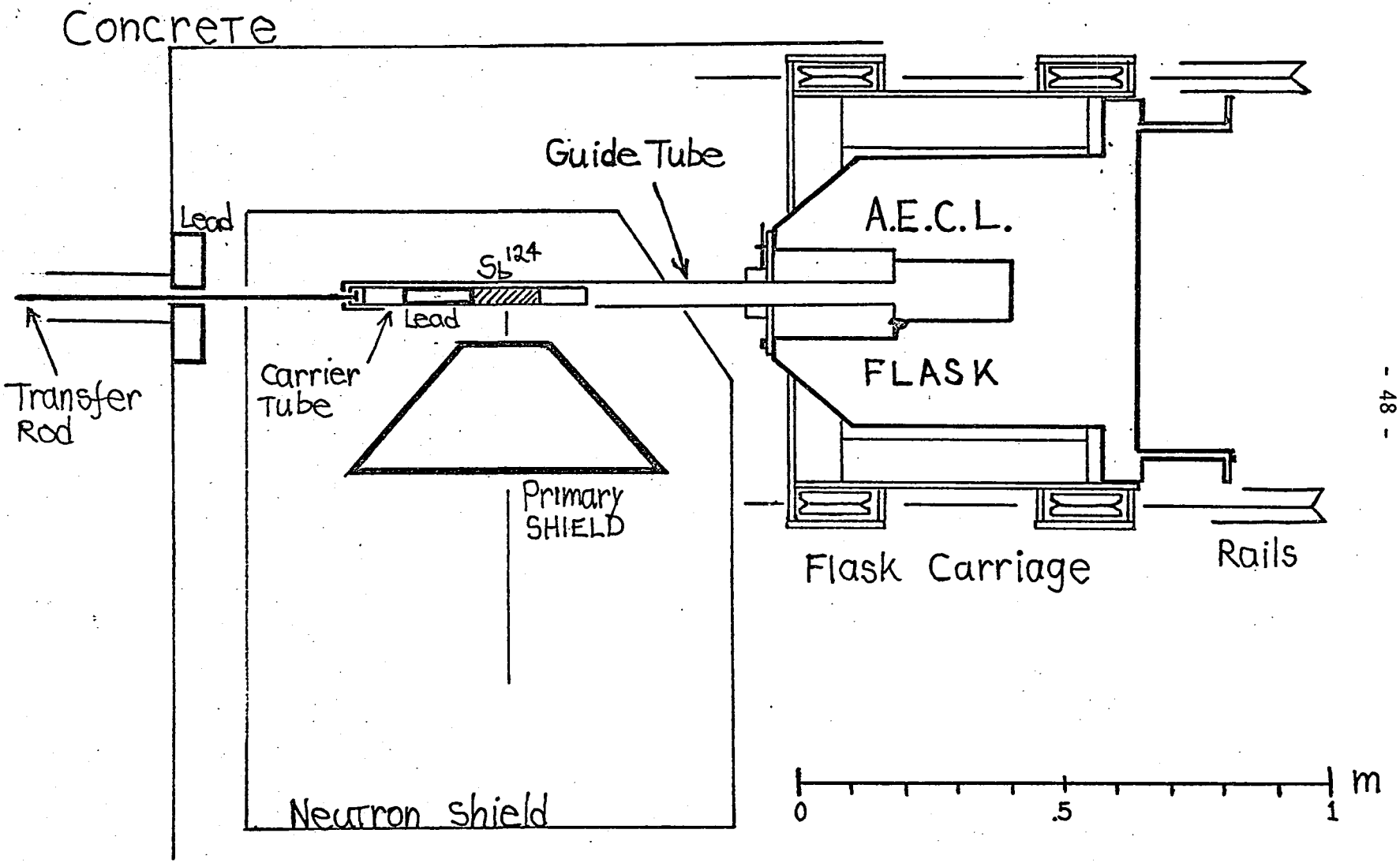


Figure 13. PLAN VIEW OF THE EXPERIMENT AREA.

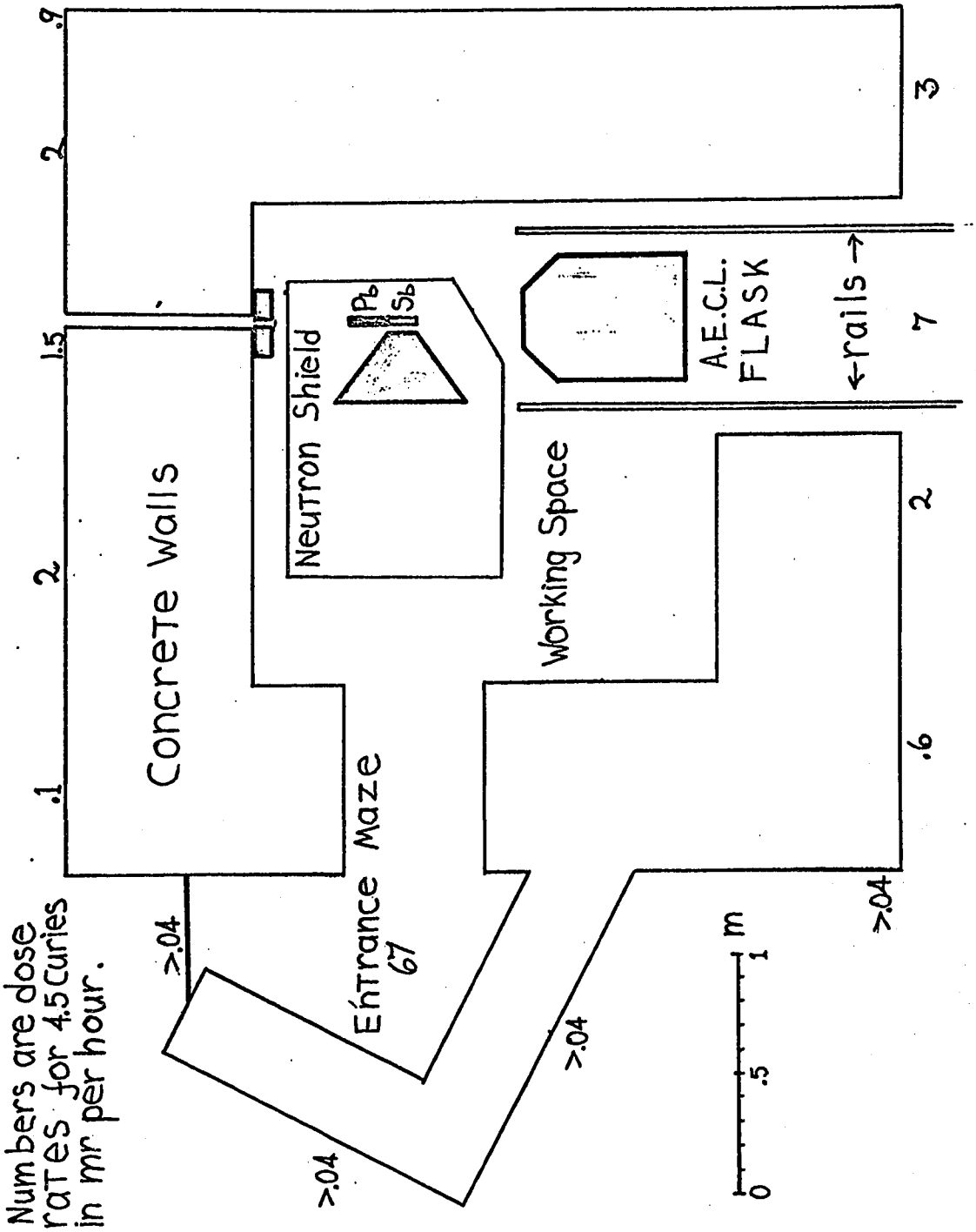


Figure 14. PLAN VIEW OF EXPERIMENT AREA AND CONCRETE RADIATION CELL.

VI. BACKGROUND RADIATIONS OBSERVED IN THE PROTOTYPE

The backgrounds due to the antimony source, neutron reactions in the apparatus, and activation were each studied.

The antimony background was studied in the absence of neutrons by removing the beryllium blocks, once with the paraffin absent and once with the paraffin in place. The difference between these two should be a measure of the rate of scattering from the paraffin to the counter. The results of the first experiment on scattering can be used to estimate this rate, since only a narrow zone of the moderator containing about 13.6 kg of paraffin is exposed to the source and visible from the counter. The center of this zone is 44 cm from the source and 39 cm from the counter and the angle between these two distances is 110^{o}. Since compton scattering cross-sections for light elements are proportional to the number of electrons present, and hence nearly proportional to the mass of scattering material, the 13.6 kg of paraffin will scatter 6450 times as many gammas as 1 cc of carbon. Applying the distance factors using the experimental results for scattering through 110^{o} gives an estimate of 3×10^{7} cps for a 10 curie source. The second experiment on scattering indicated that when the concrete cell walls surrounding the apparatus were the only scattering material present the counter would show 9×10^{6}

cps. Thus projections based on previous experiments indicate that when the paraffin is added the total counting rate due to antimony gamma rays via all paths should increase by a factor of 4.3.

With source strength at 4.04 curies, 900 cps were observed with neither beryllium or paraffin in place and 3400 cps were observed with the paraffin added. The addition of paraffin increased the counting rate by a factor of 4.8

The main features of the background arising from the antimony source are a strong lead or Bismuth x-ray peak, a broad hump centered at around 174 kev, and a broad peak at 515 kev. When the bismuth sample container was pulled away from the lead counter enclosure opening up a 1/8" gap the 174 kev peak grew many times stronger and became the dominant feature of the spectrum, while the 512 kev peak did not change. This suggests that the 512 kev peak is a residual effect of the antimony radiation coming directly through the lead. In any case its energy is too high to allow explaining it in terms of scattering since the smallest scattering angle possible is about 90^{o}, and the cut-off energy for the 90^{o} scattering spectrum is 482 kev. The scattering spectrum corresponding to the 174 kev hump is for 140^{o} scattering.

Before observations on background during neutron generation reported below were made, some modifications had been carried out. The inside of the lead counter enclosures was coated with borated polyfilla, the lithium plate was separated from the sample area by a ½" layer of paraffin. Applying these changes to the "practical prototype" specifications give the apparatus in its final form. The reasons for these changes will be discussed under the heading optimisations.

The two landmarks in the background during neutron generation are the 2.2 Mev hydrogen prompt gamma and the 480 kev boron prompt gamma. Accordingly prompt gamma spectra and background will be divided into three ranges - the low energy range (0 - 500 kev), the medium energy range (.5 - 2.2 Mev), and the high energy range (over 2.2 Mev).

The high energy background is devoid of peaks, but its extinction at about 7.5 Mev points to the large mass of moderately high cross-section lead as a source. Lead has very prominent prompt gammas at 7.38 and 6.85 Mev. This radiation contributes 576 counts per second to the background, essentially equal to the high energy counting rate due to a 1.2 kg iron plate in the sample enclosure. Clearly the high energy background will be greater than the high energy spectra of typical

samples, and this will be an inherent limitation on resolution in the present apparatus. Use of more bismuth is called for in future designs.

The medium energy range includes the energies of direct antimony gamma rays, though very few of these reach the counter. The medium energy background thus consists largely of gamma rays from neutron induced reactions, especially counts belonging to the compton tail of the hydrogen (n, γ) reaction. The background counting rate in this range is 1700 cps, about 6 times the counting rate due to the iron sample.

The low energy background consists of the boron prompt gamma, the scattered antimony gammas, and the lead x-ray. Activation also makes its largest contribution in this range. The background counting rate in this range is 4.05×10^3 cps, thirteen times the counting rate due to the low energy iron spectrum and 1.2 times the counting rate observed in the absence of beryllium.

When the activation background was investigated there was no neutron absorber lining the inside of the counter enclosure. Instead, the lithium plate was placed closer to the counter, $\frac{1}{4}$" from its face. After a three hour irradiation period, the activation observed by removing the antimony source amounted to 150 counts per second and extended 2.1 Mev,

tapering off towards its high energy limit. After 1 hour, the counting rate had decreased to approximataly 1/3 of its initital rate. This points towards the iodine of the counter as the main activation source, since iodine 128 has a strong (79 per 100 disintegrations) beta at 2.12 Mev with a half life of 25 minutes.

The total background for a long irradiation with a 4.04 curie source is about 6400 cps. Such a high counting rate was not actually observed because the source had weakened considerably since the first measurements were made at 4.04 curies. All other background rates were measured with less antimony activity and were normalized to the initial value. The neutron flux measured at various points in the sample container with indium foils varied from 1.9×10^3 to 6.1×10^3 neutrons per cm^2 per sec. Assuming as before that the neutron production is 10^7 n/sec and assuming no reflection of neutrons, a geometric calculation predicts a flux of 1.5×10^3 /sec/cm^2 for 4.04 curies. Apparently the paraffin is serving a useful function as a reflector as well as moderator.

VII. OBSERVATION OF THE IRON PROMPT GAMMA SPECTRUM

A sodium-24 source was used to calibrate the counter in the high energy range, and study its resolution performance before serious observation of the iron spectrum. Sodium-24 has strong gammas at 1.37 Mev, 2.75 Mev and 4.12 Mev (Sum Peak). These peaks are depicted as observed in figure 15. The source used also contained copper-64, which gives a strong 511 kev gamma. Each peak has been analysed to give the resolution of the counter over the energy range available. Resolution is much more easily studied this way than with prompt gammas, since the background with a radioactive source is minimal. Figure 16 shows how the resolution of the present counter compares with that used by Greenwood in preparing his atlas of prompt gammas[11]. It can be seen that Greenwood's 3 x 3 NaI(Te) spectrometer can resolve from 25% to 40% finer detail than the present spectrometer. This together with the large background must be taken into account when comparing the iron spectrum presented here to the spectrum found in the atlas.

Several facts should be noted in connection with the very poor resolution of the Cu-64 peak. The calibration spectrum was also observed with a Ge(Li) detector, and a second gamma ray was seen very close to the

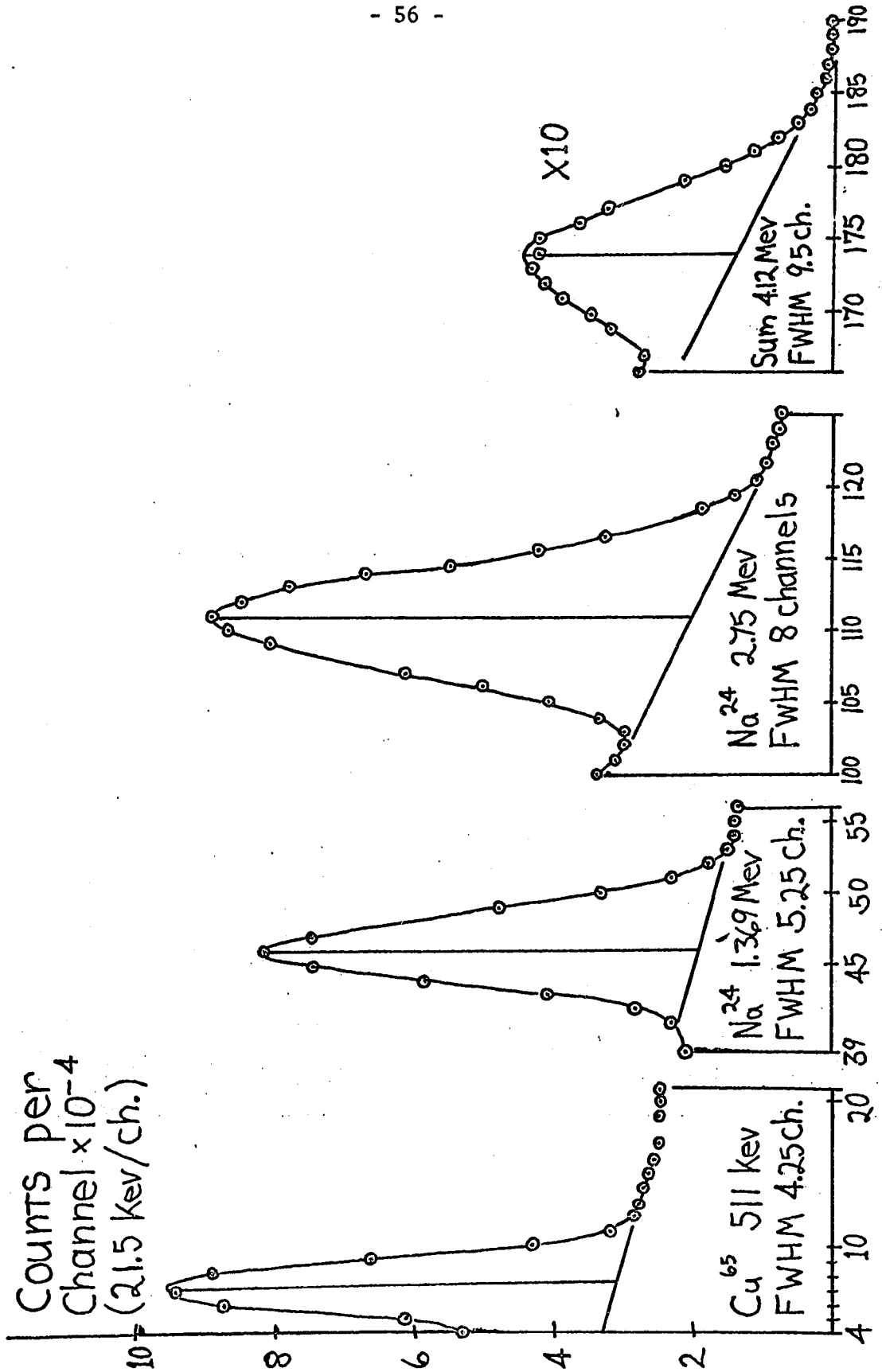


Figure 15. RESOLUTION TEST USING SODIUM-24 SOURCE.

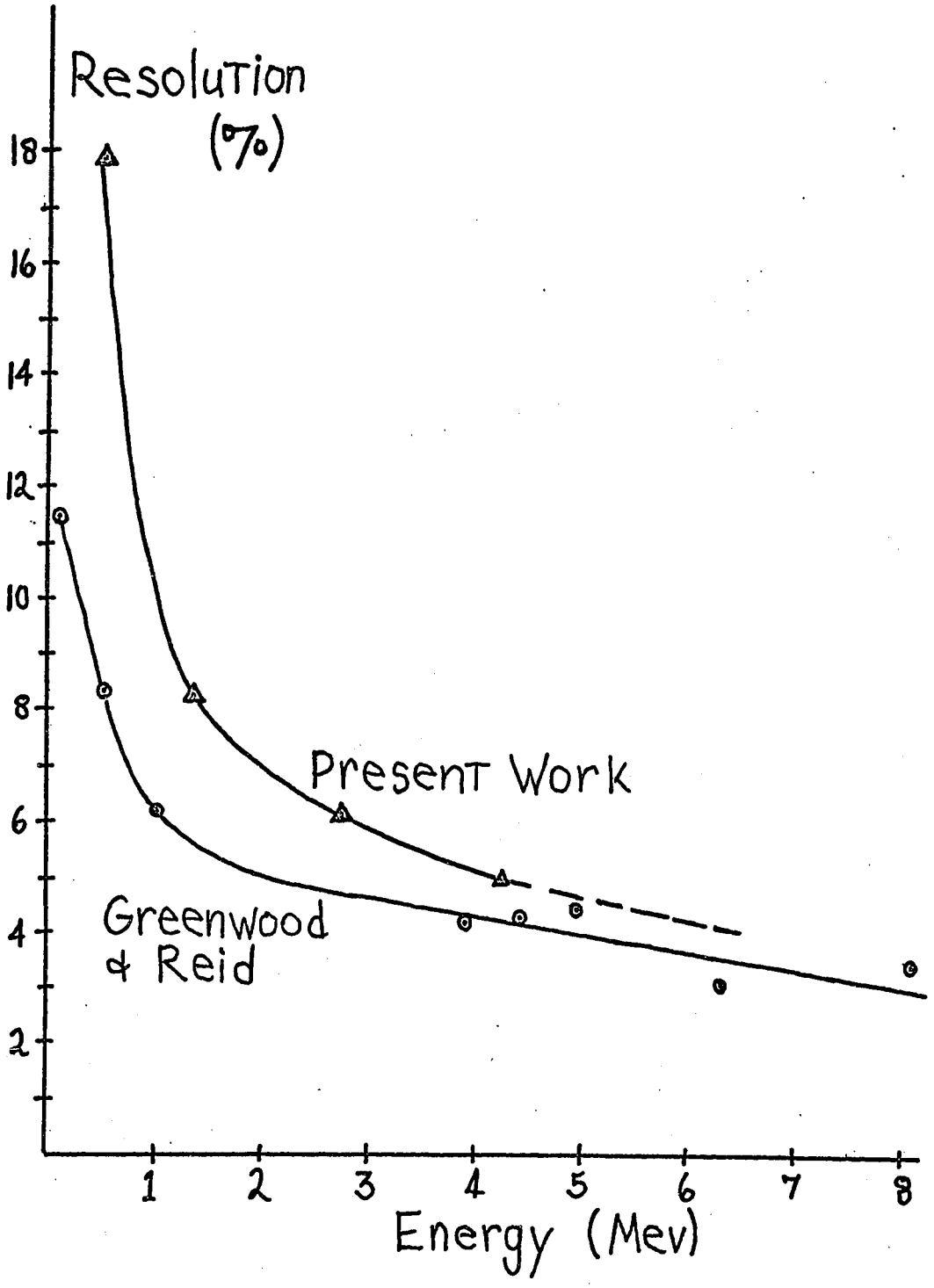


Figure 16. RESOLUTION CURVE OF THE 3 x 3 INCH NaI(Tl) COUNTER

Cu-64 peak. This was identified as the As-76 559 kev gamma. The resolution of the Cu-64 peak was poor for a Ge(Li) detector- approximately 4.5% as opposed to 1.95% for the 1.37 Mev peak (2.2 times poorer resolution comparing the two peaks). The NaI detector, as shown on the resolution curve, also gives a resolution ratio of 2.2 for the same two peaks. This suggests that the poor resolution may be based on the mechanism of detection of the Cu-64 radiation, which is actually a low energy positron. Perhaps this mechanism involves small additions to or subtractions from the energy of annihilation received by the counter, in such a way as to broaden the peak. The resolution problem for this peak was not investigated because the resolution in the high energy range, the range of greatest present interest, appeared quite acceptable.

Following calibration with the sodium source, several iron prompt gammas were readily identified. The channel numbers and energy identification of these peaks have been plotted along the sodium peaks on a calibration graph (figure 17).

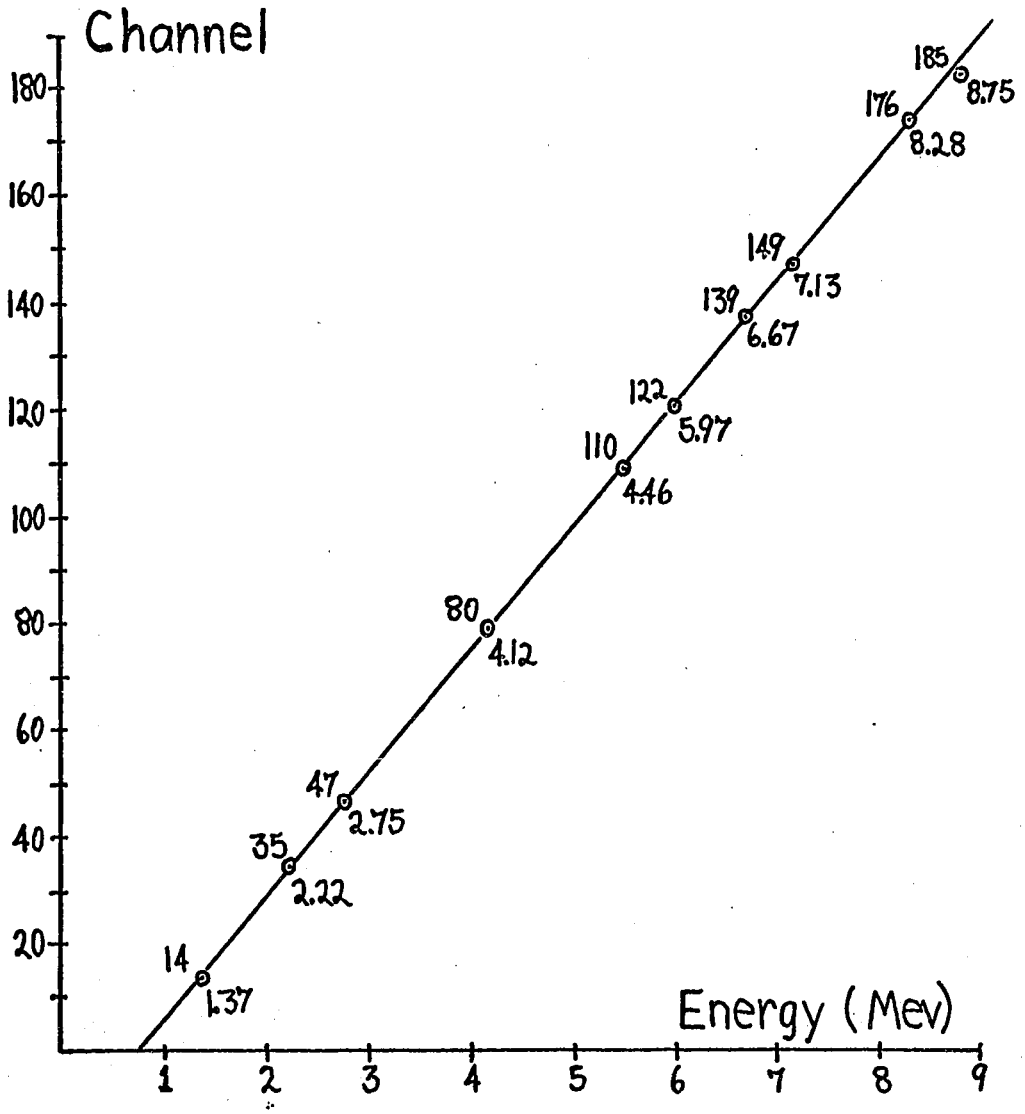


Figure 17. HIGH ENERGY CALIBRATION USING Na^{24} ACTIVATION GAMMA PEAKS AND IRON PROMPT GAMMA PEAKS.

The data are as follows:

Peak Channel	Identification
14	1.37 Mev (Na)
35	2.22 Mev (H)
47	2.75 Mev (Na)
80	4.12 Mev (Na)
110	5.46 Mev (Fe)
122	5.97 Mev (Fe)
139	6.67 Mev (Fe)
149	7.13 Mev (Fe)
176	8.28 Mev (Fe)
185	8.75 Mev (Fe)

Figure 18 shows the iron spectrum obtained and compares it with the Greenwood and Reid atlas spectrum and with the spectrum obtained by Christell and Ljunggren using 4.4 x 5 cm NaI(Te) counter with a Pu-Be source[12]. Above and below the reactor-generated Greenwood and Reid Atlas Spectrum are the spectra obtained with the radioisotope neutron sources. Both of the latter show greatly decreased resolution of the iron prompt gamma peaks. Both show a very high background level,

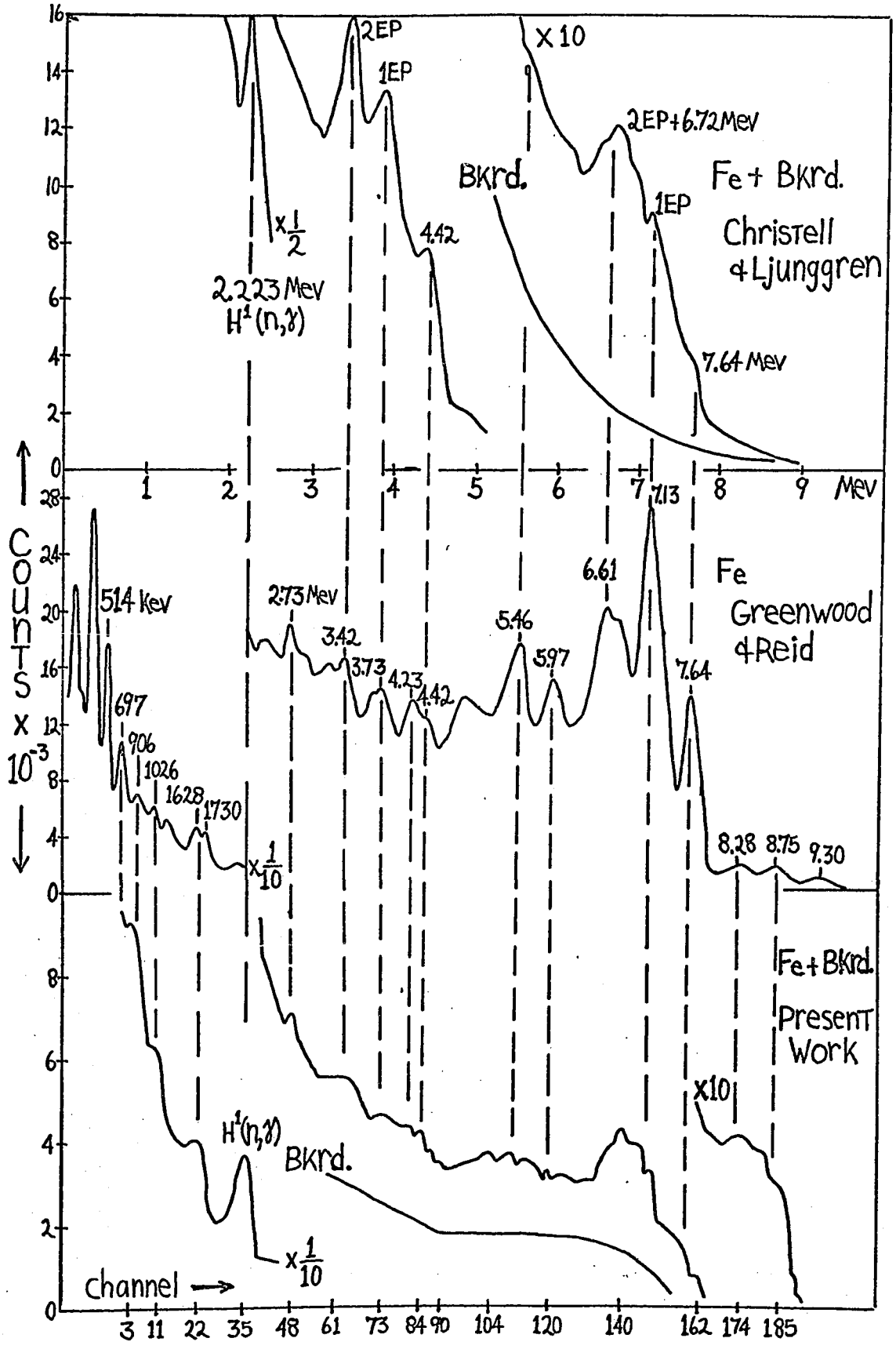


Figure 18. IRON PROMPT GAMMA SPECTRA.

which is probably the chief cause of the poor resolution. It is also probable that Greenwood used a very carefully prepared NaI(Tl) crystal giving a level of resolution superior to that of the detectors used in the other two experiments.

In viewing high energy gamma spectra it must be remembered that the nature of the interaction of high energy gammas with matter cause full energy peaks to be almost insignificant relative to escape peaks and the compton tails. Compton scatter photon and annihilation photon escape increases strongly with energy. The compton edge may become so nearly placed on the energy scale to the full energy peak as to be unresolved from it. For back scattered photons the compton electron energy as a percentage of incident photon energy is $2\alpha \quad x\ 100\%/1 + 2\alpha$, where α is the incident photon energy in units of electron rest mass. Near the upper end of the iron spectrum the energy is 15 mc^2 and higher. Substituting 15 for α gives a compton edge energy equal to 30/31 or 97% of the full energy peak. Projecting the resolution curve obtained for the present counter into that energy range suggests that resolution much better than 4% cannot be obtained, so the compton tail would be expected to be fused onto the peak. For the sodium sum peak $\alpha = 8$ and the separation between the peak and the edge is 6%. The measured

resolution is 5%, consistent with the fact that the peak is just barely resolved and has a low energy side height only about 1/3 of its high energy side height.

VIII. OPTIMISATION

The number of factors which could be varied in order to find the most efficient configuration were limited by the massive nature of the shielding and the length of time required to gain access to the shut-down apparatus. Changes made were evaluated by analysis of the iron spectrum subsequently obtained. Three figures of merit were computed from each spectrum. The ratio of the counts in the channel at the 4.6 Mev minimum to the counts in the peak channel at 8.28 Mev is a measure of the relative strength of the background. The effect of the high energy background at 4.4 Mev is maximal and at 8 Mev essentially non-existent. The ratio of counts in the 2.2 Mev peak channel to counts in the 8.28 Mev peak channel is a measure of the interference due to hydrogen. The total number of counts over 8 Mev is a measure of the rate of neutron capture in the iron. In final configuration, the minimum to 8.28 Mev ratio is 15.5, the hydrogen to 8.28 Mev ratio is 83, and the over 8 Mev counts were 5240 in 10 minutes.

Before the boron coating was applied to the inside of the counter enclosure, the lithium plate was moved temporarily 3 cm away from the sample enclosure towards the counter. The aim was to see if increased

separation between the sample and lithium would minimize any depression of the neutron flux at the sample due to the lithium. The iron plate was located at the middle of the sample container with the $\frac{1}{2}$" paraffin sheet next to it on the side facing the lithium sheet. The total counts over 8 Mev decreased by about 10%. Since no explanation is apparent this experiment is perhaps best termed inconclusive. The "hydrogen ratio" however did increase by about 10%, perhaps indicating that the expected flux depression has a short range of action, (less than the thickness of the intervening paraffin).

Lining the counter enclosure with boron improved the "minimum" ratio by a factor of 1.8. This would appear to identify the lead of the counter enclosure as a major contributor to the high energy background. Evidently an appreciable number of neutrons get into the lead in spite of the boron coating on the outside of the enclosure, probably through the face contacting the bismuth.

A $\frac{1}{2}$" layer of paraffin was placed on the large flat face of the sample container next to the pyramid shield in order to thermalize neutrons which followed a direct path through the lead. This affected only one of the figures of merit. A 15% improvement in the minimum ratio resulted. To see if this benefit could be enlarged, the paraffin layer

was extended over the entire vertical face of the pyramid shield. This did not result in a further improvement of the minimum ratio. On the other hand, it did increase the hydrogen ratio by 12%. Obviously again the paraffin is useful in limited quantities.

The size of the moderator envelope around the pyramid shield was reduced temporarily with the removal of 10% of the paraffin. It was thought that confining the neutrons to a smaller volume would result in a flux improvement. At the same time an attempt was made to minimize the sharpness of corners in the moderator envelope. Contrary to expectations the available flux, as indicated by the counts over 8 Mev decreased by 30%. Evidently, reducing the width of the paths between the paraffin and the lead forced more of the neutrons to go into the lead and be absorbed. This view is supported by an 8% rise in the minimum ratio, corresponding to an increase of lead capture gamma background.

Finally, an indication of the influence of the source to sample distance was obtained by moving the entire secondary shield assembly with counter backwards 5 cm away from the counter. This resulted in a 14% reduction in neutron flux at the sample as indicated by the counts over 8 Mev.

IX. SENSITIVITY CURVES

Since the purpose of the present work is to determine the effectiveness of the antimony-beryllium source for prompt gamma analysis of objects such as ore samples, the spectra of "artificial ores" of various known concentrations were observed. Iron chips mixed with sand, and sulphur mixed in powdered coal were used. In each case an attempt was made to find an energy range in which the total counts including background was sensitive to the sulphur or iron content. For iron the total counts from 6.55 to 7.41 Mev including background were observed as a function of concentration during 10 minute live-time counts. The results are tabulated below and displayed graphically on a sensitivity curve (figure 19).

Sample volume	Iron Density	Iron % by Wt.	Counts
2.05 litres	——————(sand only)——————		20306
	.169 gm/cc	10.1%	38261
	.356 gm/cc	19.8%	53585
	.488 gm/cc	26.9%	58875
	.584 gm/cc	33.2%	63249
	.741 gm/cc	41.2%	73461
	.930 gm/cc	49.1%	81959

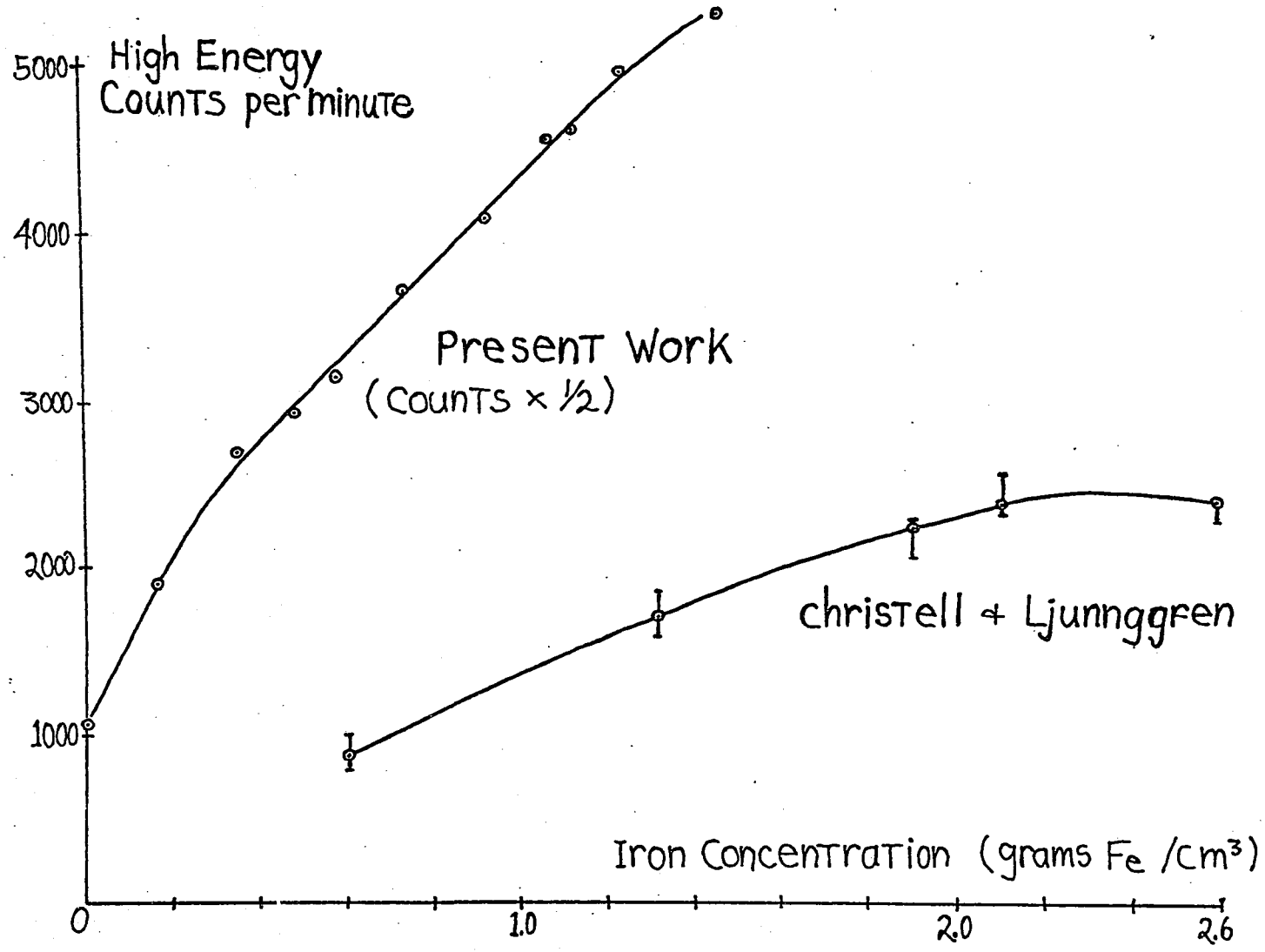


Figure 19. PROMPT GAMMA SENSITIVITY CURVES FOR IRON ORE METALS.

Sample volume	Iron Density	Iron % by Wt.	Counts
	1.07 gm/cc	56.3%	91348
	1.13 gm/cc	64.6%	92286
	1.24 gm/cc	73.0%	99232
	1.46 gm/cc	93.9% (no sand)	106097

Study of the sensitivity curve indicates that the present technique is capable of measuring iron concentrations at least as low as 0.2 gm/cc with a precision of .04 gm/cc. Both the precision and the range minimum appear to be very readily improvable. For a given concentration the uncertainty in the counting rate indicated by the graph is 2200 counts. The threshold concentration for the detection of iron with the present arrangement would be the concentration at which the counting rate is elevated above background (on the graph line) by just this amount. The threshold concentration thus determined is only 0.02 gm/cc.

Sulphur presents a different problem since its neutron capture cross-section is small (.512b) compared with that of iron (2.62b) and sulphur has no prominent gammas higher in energy than the background cut-off energy. Also the sulphur concentrations in samples of interest are

much less than those in iron ores. Coals contain at most a few percent of sulphur. A good sulphur spectrum was obtained by filling the sample container with 1.9 kg of amorphous sulphur. This is shown in figure 20 with the Greenwood spectrum for comparison.

As evident of the Greenwood sulphur spectrum, the 3.9 to 5.3 Mev region stands out most clearly against the background. In fact the 4.91 Mev gamma and the escape peaks of the strong (82% intensity) 5.42 Mev peak provide a more outstanding landmark in the middle of the high energy range than is available in the case of iron, where the greatest intensity under 5.6 Mev is 4%. Before measuring the total number of counts in the 3.9 Mev to 5.3 Mev interval, the background using a relatively sulphur free coal sample was subtracted during an equal live time counting period. This was done four times with different amounts of sulphur mixed in the original coal. No sulphur gammas were evident in the background coal sample.

Source 2.82 curies	Sulphur density	Sulphur % by Wt.	Counts
100 min. Live time	nil	————	16500
Vol. 2.05 l.	.022 gm/cc	2.74%	5126
	.044 gm/cc	5.47%	9207
	.084 gm/cc	10.5%	27316

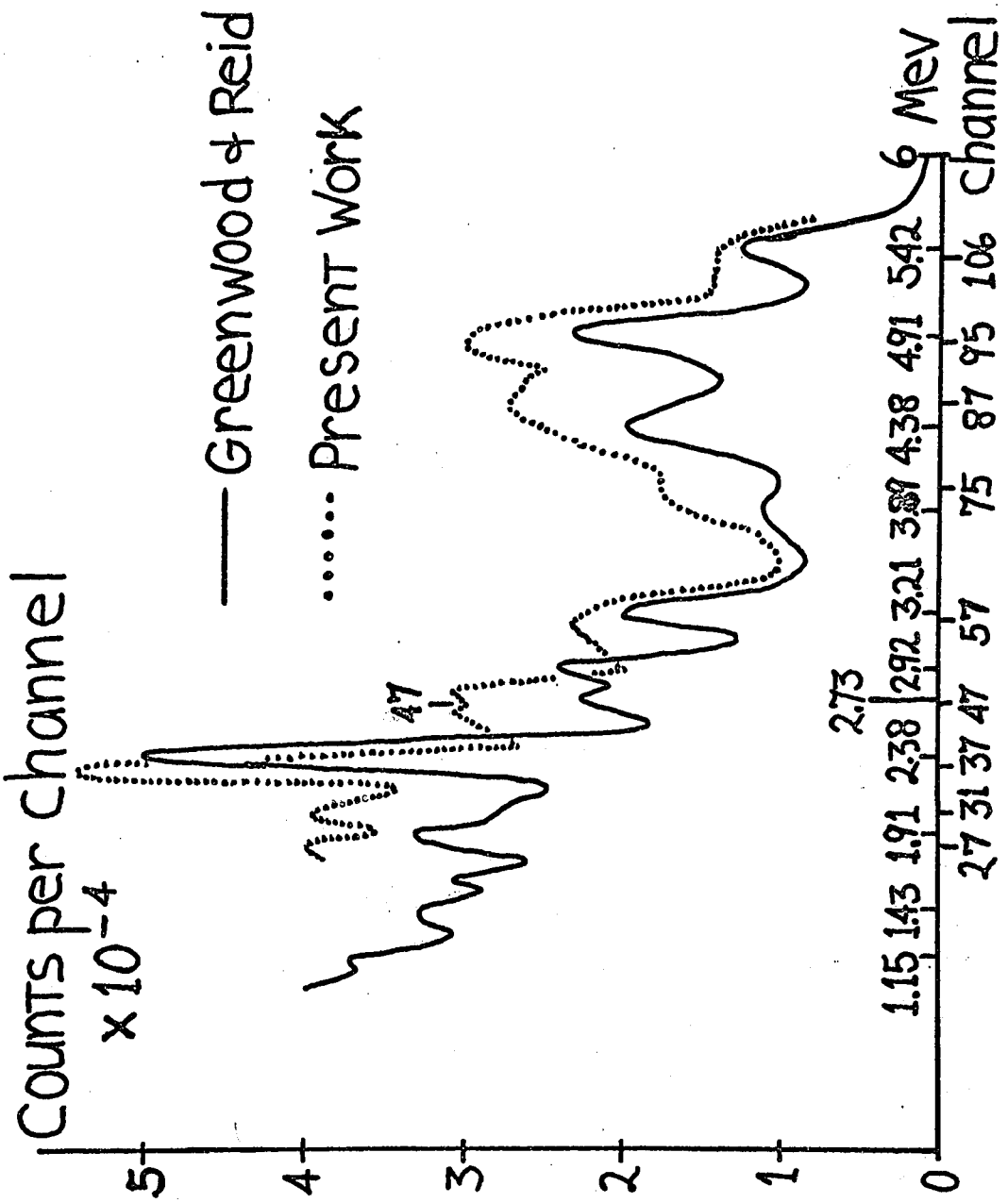


Figure 20. SULPHUR PROMPT GAMMA SPECTRA.

Sulphur Density	Sulphur % by Wt.	Counts
.164 gm/cc	20.5%	78310
.925 gm/cc	100.0%	8×10^5

Background due to unsulphured coal 26134

The sensitivity curve resulting from the data is shown in figure 21. Below a sulphur concentration of 0.02 gm/cc, the present apparatus would require several hours of counting time in order to overcome the poor statistics inherent in a background level several times larger than the effect due to the sulphur. However the data clearly supports the feasibility of rapid analysis of coal with as little as .002 gm/cc of sulphur. At such a low sulphur level one could still receive ten times as many counts from sulphur in a one minute counting period using a "full strength" 3000 curie antimony source.

When viewing the above table of data it should also be kept in mind that in real coal samples (unpowdered) the sulphur density figure will correspond to a much smaller weight percent figure, as a chunk of coal. A hard anthracite coal with .02 gm/cc of sulphur may have as little as 1.35% of sulphur.

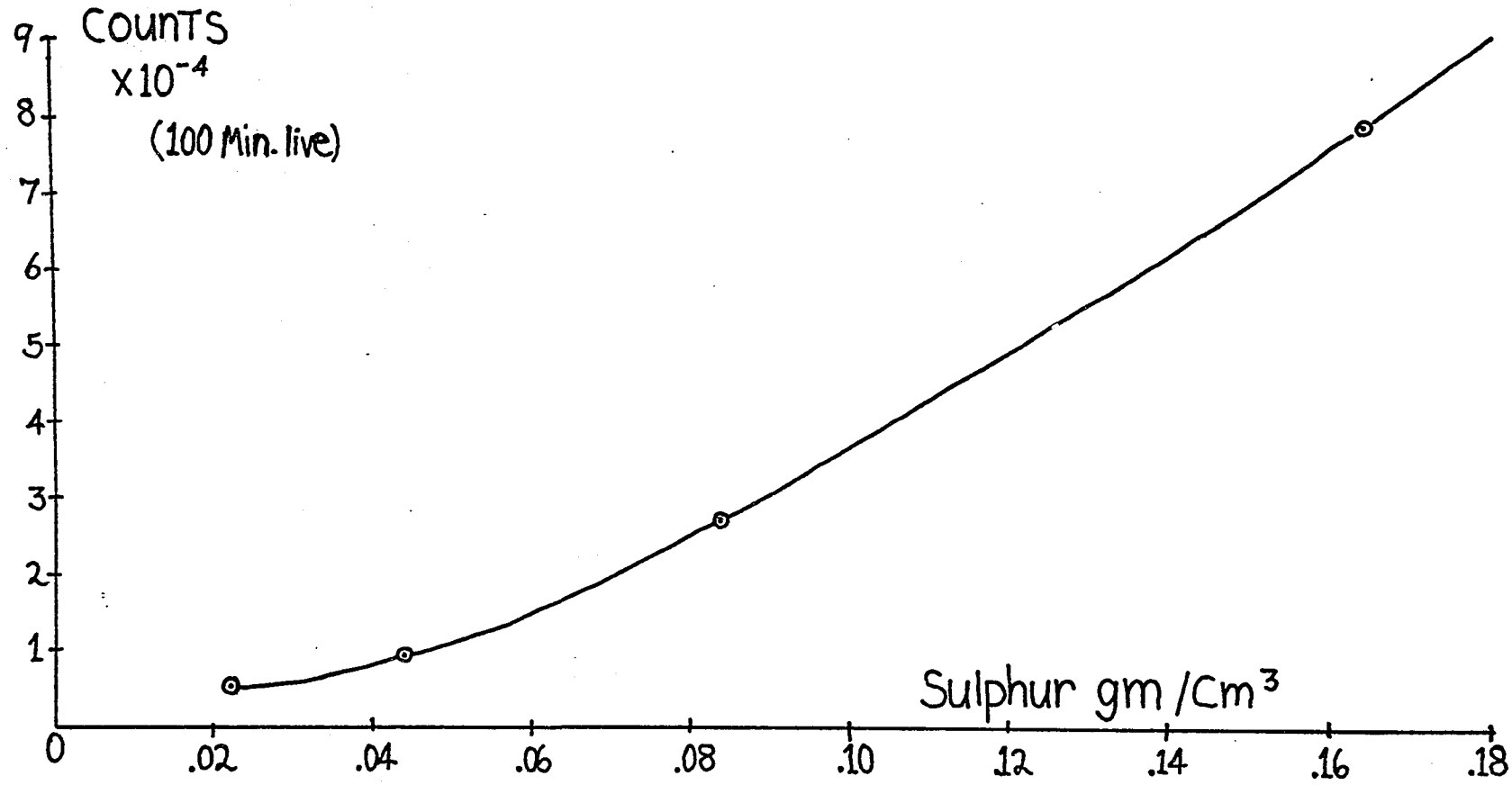


Figure 21. SENSITIVITY CURVE FOR SULPHUR IN COAL.

X. FUTURE WORK

Using the experimental evidence reported here, a new apparatus can be made which incorporates 10 modifications and which will make possible very efficient and rapid observation of prompt gamma rays from a wide range of samples up to 2 litres in volume. The modifications are shown in figure 22, and are discussed below:

1. Six beryllium blocks may be added to the nine already present, given approximately 50% more neutron production.
2. The moderator envelope should be drawn in more closely to the source-counter axis and is made with rotational symmetry as originally desired. The neutron volume will be about two-thirds of the present volume, and this should increase the flux by 1.5.
3. The lead in the counter enclosure and part of the lead in the pyramid shield should be replaced by bismuth. This should cut the high energy background down to about 20% of its previous level (i.e. to 120 cps)

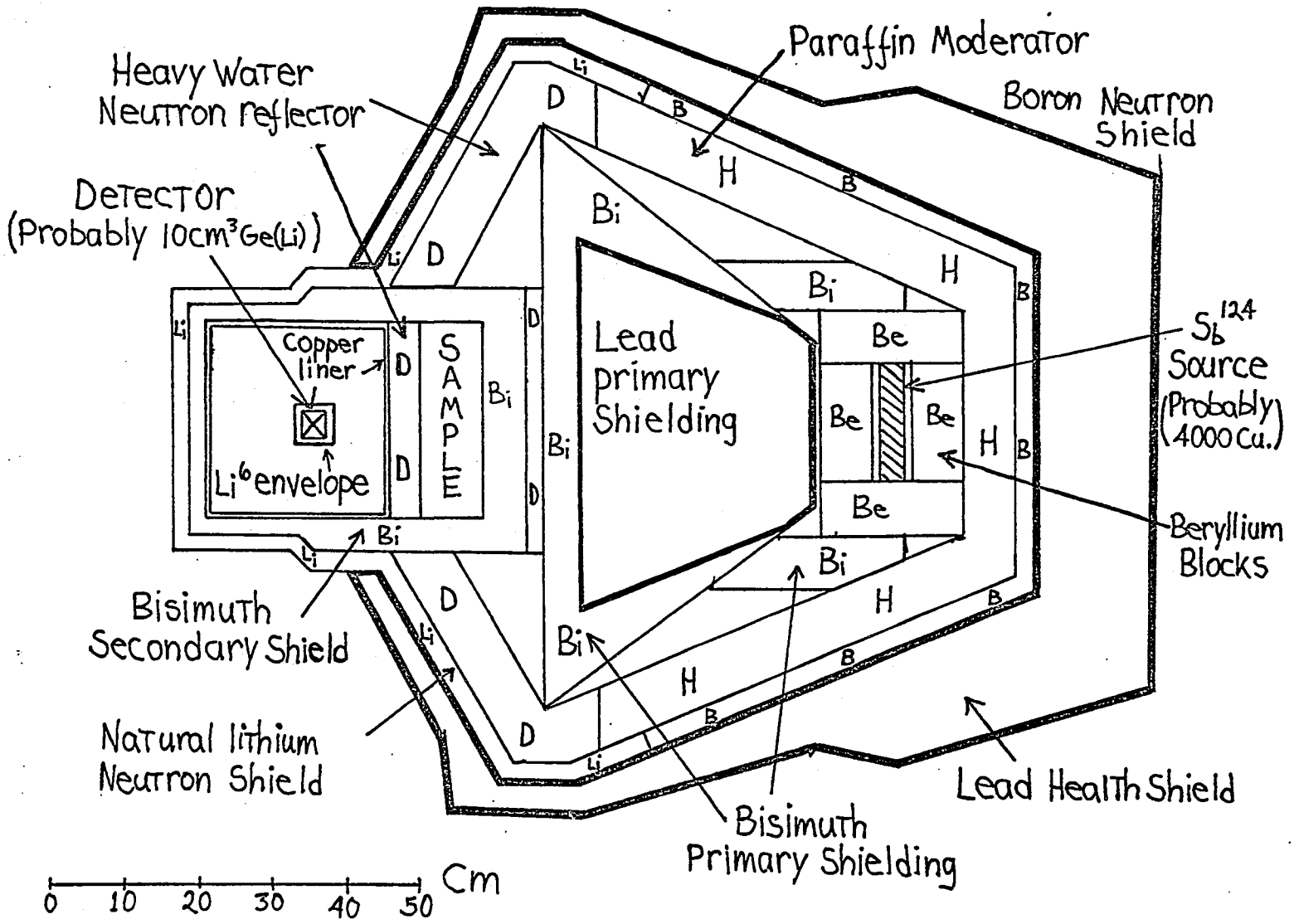


Figure 22. AN IMPROVED Sb-Be PROMPT GAMMA FACILITY.

and provide the same channel for the neutrons between the lead pyramid and the moderator as before.

4. The paraffin near the counter should be replaced by heavy water. This will virtually eliminate the hydrogen prompt gamma (approximately 1400 cps removed from medium energy background). The lead background of 300 cps in this range will be reduced to 20%, so the medium energy background becomes only 60 cps. It should also produce a substantial increase in neutron flux, perhaps 50%. The total benefit to the neutron flux given by modifications 1, 2 and 3 will be approximately 1.5 x 1.5 x 1.5 or 3.4 times increased.

5. The very bulky concrete health shield should be replaced by a lead shield having a maximum thickness of 10". Aside from providing a vastly more compact facility this will do away with the 500 cps of low energy background scattered by the concrete. Approximately 400 cps of the low energy background is attributed to direct penetration and will not be affected.

6. The boron around the counter enclosure should be replaced by about 1" of natural lithium and the boron component of the counter shield should be kept far from the counter. This will, together with the removal of the compton tail due to hydrogen gammas, remove about 600 cps of background.

7. The low energy background is now reduced from 4050 cps for 4.04 curies to 2950 cps. About 2500 cps of this is due to scattering from the paraffin, the rest being due to direct penetration and x-rays formed by the secondary shielding. This 2500 cps component can be easily decimated by placing a 2" thick bismuth collar around the beryllium. The low energy background estimate would then be 250 + 450 cps.

   The total background will thus be roughly as follows:

Low Energy	700 cps
Medium Energy	60 cps
High Energy	120 cps
	880 cps

8. The lithium-6 plate is replaced by a close fitting Li^6-

sheath surrounding the counter. This should assure that crystal activation will be negligible.

9. A 4000 curie source should be used instead of the 4 curie source used in the present work. The following specifications then apply:

Background seen by 3 x 3" NaI(Te) 9.3×10^5 cps

Minimum neutron flux in sample container

$= 3.4 \times 2 \times 10^3 = 7 \times 10^6$ n/sec/cm^2

Average neutron flux 10^7 n/sec/cm^2

Total Iron rate for 3 x 3/ NaI(Te) = 1630 cps x 3.4×10^3

$= 5.55 \times 10^6$ cps

10. It can be seen that the Sb-Be source may be used to obtain prompt gamma and background counting rates similar to those obtained in current reactor-based experiments. In these circumstances the use of a Ge(Li) spectrometer is indicated, since the lower efficiency will no longer be a penalty. A 10 cm^3 crystal will produce peaks 4.2 times higher than those produced by a 3 x 3/ NaI(Te) crystal in the

same flux by virtue of the greatly increased resolution[13]. This will allow easy analysis of an element by observing the peak counting rate for a single gamma characteristic of the element. Analysis of many elements in a single sample will be straightforward, even when they all have similar neutron cross-sections.

Given the great selectivity of the Ge(Li) detector, the only serious analytical problem should be the inhomogeneity of samples, and neutron flux. When these problems are solved, the monitoring of a continuously flowing sample should be investigated. For many elements present in the products of continuous flow processes a prompt gamma peak will be available whose height will be sufficiently sensitive to concentration to allow fluctuations to be located with a short time constant. That is, with sample flowing through the sample container at a rate of several inches per second the peak counting rate should still reflect the instantaneous contents of the sample chamber fairly accurately.

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