

SHIELDING PROPERTIES OF THE  $UPb_3$   
INTERMETALLIC COMPOUND

by

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## I. INTRODUCTION

The rapid expansion of the nuclear industry in the past twenty years has resulted in a large increase in the volume of radioactive material. It is essential that both the general public and the personnel involved in working with these materials have sufficient protection against harmful amounts of radiation. However, the list of materials that can satisfactorily provide this protection is rather limited. These materials must not only serve as an adequate shield but also be available at a reasonable cost. Moreover, no single material can act as a satisfactory shield for all of the various forms of radiation. For example, substances such as water or paraffin which are composed of light elements are more efficient as neutron shields whereas heavy metals such as lead or uranium offer better protection from gamma radiation.

The most important shielding materials in use today include water, concrete, iron, and lead. For an installation such as a nuclear reactor, both neutron and gamma radiation must be considered; combinations of several of these materials are therefore required for adequate protection. Gamma radiation constitutes the primary hazard when handling or transporting such items as spent reactor fuel elements, radioactive wastes, or certain radioactive isotopes. Lead has been used almost exclusively as the major shielding material

in such instances.

When used as a shield, lead has two main disadvantages: its melting point is quite low ( $327^{\circ}$  C) and it has little structural strength. In the storage or transportation of radioactive substances, the containment vessel must provide the necessary protection not only under ordinary circumstances, but also in the event of an accident. If such an accident involves fire, loss of shielding due to the melting of the lead would result in local radioactive contamination. Some other form of accident could cause severe mechanical damage to an easily deformable lead container. In consideration of these possibilities, the AEC has introduced design criteria to serve as a standard for the construction of containment vessels.

Clearly there is a real need for metals other than lead that can be used in a satisfactory container. Several possible elements include tantalum, tungsten, or uranium; however, the cost of fabricating such a container would restrict its utility to special applications. One alternative to the use of elementary metals is the possible application of some alloy. An examination of the uranium-lead system indicates that certain alloys of these metals could perhaps be used as a satisfactory shield. Some of the properties of this system are discussed in the following section.

## II. REVIEW OF THE LITERATURE

In searching the literature for the background of this thesis, two different types of information were sought. First, it was necessary to determine the design standards and tests that the nuclear industry requires of containment casks in general. Second, the extent of investigations into the uranium-lead system was reviewed. The literature that has been cited in Part B appears to represent the major portion of the work that has been done with the uranium-lead system to date.

### A. General Requirements for Shipping Casks

The most stringent requirements for containers of radioactive material have been imposed on those which are to be transported. The overall problem of packaging and transportation was recently reviewed during an international symposium held at Albuquerque, New Mexico. The proceedings of the symposium (18) present a fairly complete picture of the latest testing methods and containers now in use.

In the design of shipping casks, the size, shape, and choice of materials are the option of the designer. In general, this is largely influenced by the class of radioactive material the cask is to contain. The AEC classification of these materials is determined by such factors as the amount and energy of the activity, and the type of radiation.

In addition, special consideration is given to the transportation of fissile materials. A detailed treatment of this and other federal regulations can be found in (17).

In order to evaluate the ability of the cask to withstand the demands which might reasonably be placed on it, the AEC has proposed a series of tests. A set of accident test conditions consist of "the sequence of a 30-foot drop onto a flat surface, followed by a 40-inch drop onto a 6-inch diameter plunger, followed by exposure to an 800° C thermal environment for 30 minutes, followed by immersion in water". The casks are not expected to survive, completely without damage, the effects of such severe testing. However, it is expected that they satisfy the following criteria:

1. Criticality is avoided if the cask contains fissile material.
2. The radioactive contents remain in containment.
3. Satisfactory shielding from these contents is still provided.

A large number of different types of containers were tested during this symposium. They were constructed from a variety of materials such as wood, steel, and lead. The performance of containers which utilized lead are of particular interest for this discussion; hence, some results of these tests will be considered.

For the most part, these containers withstood the testing in a satisfactory manner. The successful performance of

many of the containers reflect the improvements made as a result of previous testing programs. The information which was shared and the results that were obtained during the proceedings at Germantown (21) and John Hopkins University (20) in 1962 were utilized in constructing many of the present models. The difficulties that were encountered arose during the fire test, and could usually be traced to damages suffered during drop testing. As a result, some containers lost a portion of the lead shielding due to melting; others suffered structural damages due to expansion of molten lead and to gas pressures.

In order to utilize the shielding abilities of lead, a number of rather expensive modifications are often required, especially if the cask is to contain large activities such as a spent fuel element. A container of this type was recently built by Knapp Mills Inc. (12). Steel liners provided the necessary structural strength. Heat generated by the radioactivity was removed by a circulating water system and cooling fins. Since lead has a tendency to shrink away from steel surfaces upon cooling and thus hindering heat transfer, metallurgical bonding of the lead to the steel liners was required. Obviously, these problems could be simplified if the shielding material itself had greater structural integrity and a higher melting point.

## B. The Uranium-lead System

The bulk of the initial research into the uranium-lead system appears to have been conducted by two groups. R. J. Teitel (15) first published his findings in 1952; this was soon followed by the report of Frost and Maskrey (5). Both groups investigated the system over its entire composition range and in general, their findings were very similar. Two intermetallic compounds, UPb and UPb<sub>3</sub>, are formed in this system. Their melting points are 1280 and 1220° C, respectively. Both groups assigned a cubic structure to UPb<sub>3</sub> which was later confirmed by Brown (3). UPb was found to be very pyrophoric; UPb<sub>3</sub> was also pyrophoric but to a lesser degree.

A common approach to fabrication of these alloys is to place the constituents in a graphite container and heat to a temperature of about 1250° C in an inert atmosphere. Hampel (9) reports that a good vacuum or inert atmosphere which prevents oxidation of the uranium is a normal requirement in the fabrication of all types of uranium alloys. An alternate method suggested by Teitel<sup>1</sup> would be to "form a thick dispersion of UPb<sub>3</sub> particles in a lead matrix by reacting uranium chips with lead at around 500 to 600° C, allow the particles to settle at around 400° C, cool and cut off any clear lead. The lead completely surrounds each particle and protects it

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<sup>1</sup>Teitel, R. J., Douglas Aircraft Co., Inc., Santa Monica, California. Comments on alloy preparation. Private communication. 1966.



from oxidation".

A limited amount of research on some of the properties of the system has been reported. The Bolla Metallurgy Research Center conducted an investigation of various uranium alloys for potential use as a bearing metal (22). Among those tested were uranium-lead alloys containing from 2 to 20% uranium. It was reported that successful methods for fabricating and casting had been developed. In regard to corrosion damage, Barton and Greenwood (1) tested an alloy of 5% uranium in lead. They observed that water, in either liquid or vapor form, produced corrosion damage which resulted in cracks and a subsequent embrittlement of the matrix. Corrosion damage was negligible in dry air. Reference (11) contains information on the thermodynamic properties of some uranium-lead alloys. Warren and Price (23) investigated thermoelectric properties of  $UPb_3$ .

From the rather limited amount of information that is available on the uranium-lead alloys, it seems apparent that practical methods of fabricating shields up to the uranium composition of  $UPb_3$  are available or can be developed. Higher compositions appear difficult to attain and will probably not be practical. As a shielding material  $UPb_3$  offers some definite advantages over lead, the most obvious being its high melting point. From its composition, it seems quite logical that  $UPb_3$  would be an effective shield against gamma radiation. Hence, this compound has been selected as being the

most desirable composition of the uranium-lead system for potential use as a shielding material. It is the purpose of this thesis to determine just how effective  $UPb_3$  is as a shield against gamma radiation.

## III. ANALYSIS

In the investigation of the shielding properties of the  $UPb_3$  compound, an analytical expression will be derived which describes the attenuation of gamma radiation in an absorbing medium. This expression will then be modified to furnish a particular solution which will accurately predict the attenuation that can be expected in  $UPb_3$ .

The common unit for radiation exposure is the roentgen r, which is defined as follows:

"The roentgen shall be the quantity of X or gamma radiation such that the associated corpuscular emission per 0.001293 gram of air (one  $cm^3$  at S.T.P.) produces, in air, ions containing one electrostatic unit of quantity of electricity of either sign."

It should be noted from this definition that any radiation dose expressed in roentgens is independent of the time during which it is received. The total dose will be the integrated product of absorption rate, in roentgens per unit time, and the exposure time. The appropriate expression for the dose rate can serve as a measure of the radiation intensity in a particular region.

The basic unit of electrical charge carried by either member of a singly-ionized ion pair is  $4.8 \times 10^{-10}$  esu. Therefore,  $1/(4.8 \times 10^{-10})$  or  $2.083 \times 10^9$  ion-pairs are necessary

to yield a total charge of one esu of either sign. Since about 34 ev of energy are required to produce one ion-pair in air, the total energy absorbed per  $\text{cm}^3$  per roentgen is  $(34)(2.083 \times 10^9)$  ev, or  $7.082 \times 10^4$  Mev.

Assume that at some particular location there exists a flux of  $\phi$  photons/ $\text{cm}^2$  sec of energy  $E$  Mev/photon. If the energy absorption coefficient of air for these photons is  $\mu_e \text{ cm}^{-1}$ , then the rate at which energy is absorbed in the medium will be  $\phi E \mu_e$  Mev/ $\text{cm}^3$  sec. Since one roentgen is equivalent to the absorption of  $7.082 \times 10^4$  Mev per  $\text{cm}^3$  of air, then the photon flux in terms of roentgens per sec will be

$$\begin{aligned} \phi \frac{\text{photons}}{\text{cm}^2 \text{ sec}} &= \frac{\phi \frac{\text{photons}}{\text{cm}^2 \text{ sec}} \times E \frac{\text{Mev}}{\text{photon}} \times \mu_e \text{ cm}^{-1}}{7.082 \times 10^4 \frac{\text{Mev}}{\text{cm}^3 \text{ r}}} \\ &= \frac{\phi E \mu_e}{7.082 \times 10^4} \frac{\text{r}}{\text{sec}} \end{aligned}$$

$$\text{or} \quad 1 \frac{\text{r}}{\text{sec}} = \frac{7.082 \times 10^4}{E \mu_e} \frac{\text{photons}}{\text{cm}^2 \text{ sec}} \quad (1)$$

In calculations involving dose rates, the hour and the milliroentgen are more convenient units to use; thus Equation 1 can be expressed as

$$1 \frac{\text{mr}}{\text{hr}} = \frac{1.967 \times 10^{-2}}{E \mu_e} \frac{\text{photons}}{\text{cm}^2 \text{ sec}} \quad (2)$$

Equation 2 is the general expression for converting a photon flux of  $\phi$  photons/ $\text{cm}^2$  sec to a dose rate in mr/hr.

Normally the photon flux from a radioactive source can be measured directly by some type of a detecting device such as a scintillation detector. The amount of radiation actually detected will be some fraction of the total radiation emitted. This fraction is dependent upon the distance of the detector from the source as well as the medium through which the radiation passes. Any medium will scatter or absorb photons to some extent; however, the attenuation in air is so slight that this may be neglected for air thicknesses up to several meters. If a heavier medium such as lead is placed between the source and the detector, the attenuation will be much greater.

The mechanisms by which gamma (or photon) radiation interacts with matter falls into three main categories. These are the photoelectric process, Compton scattering, and pair production. A number of other processes also occur, but their effects are either small enough to be neglected or concern photon energies outside the range of reactor shielding interest. Hence, only the three primary processes will be considered here.

In both the photoelectric and pair production interactions, the photon is essentially absorbed by the medium through which it passes. Thus, for a given narrow beam of photons, the number which succeed in passing through an absorbing medium will be just those which have not suffered a collision. The change in uncollided photon flux then will be proportional to the thickness of a thin absorber. There-

fore, the flux change  $\Delta\phi$  resulting from passage through a thickness  $\Delta x$  of an absorbing medium will be

$$\Delta\phi = -\mu\phi\Delta x \quad (3)$$

where  $\mu$  is the proportionality constant. This constant, called the absorption coefficient, is characteristic of the absorber, and is independent of the absorber thickness if the photon beam is monoenergetic. In this case, integration of Equation 3 yields

$$\phi(x) = \phi_0 e^{-\mu x} \frac{\text{photons}}{\text{cm}^2 \text{ sec}} \quad (4)$$

where  $\phi_0$  is the original photon flux.

Equation 4 accurately describes the uncollided photon flux passing through the absorber; however, it does not account for the effects of Compton scattering. By this process photons suffer a scattering collision and hence still pass through the absorber. The contributions due to Compton scattering vary with photon energies as well as with the type and thickness of the absorber. The effect is generally very significant over a wide range of photon energies, especially in connection with thick shields. As a result of the numerous scattering collisions, the Compton effect produces a buildup of photon flux in the shielding material. Therefore, Equation 4 must be modified by an appropriate buildup factor  $B(\mu x)$  to account for the effects of the scattered radiation. Equation 4 then becomes

$$\phi(x) = B(\mu x) \phi_0 e^{-\mu x} \frac{\text{photons}}{\text{cm}^2 \text{ sec}} \quad (5)$$

The buildup factor is generally expressed as a function of the absorption coefficient  $\mu$  and the thickness  $x$  of the absorber. The value of  $\mu$  will depend upon the material of the absorber and the energy of the gamma radiation. Equation 5 can now be combined with Equation 2 to yield the corresponding expression for the dose rate  $D(x)$ .

$$D(x) = \frac{E \mu_e}{1.967 \times 10^{-2}} \phi_0 B(\mu x) e^{-\mu x} \frac{\text{mr}}{\text{hr}} \quad (6)$$

where

$$\frac{E \mu_e}{1.967 \times 10^{-2}} = \text{ratio for converting flux in photons/cm}^2 \text{ sec to dose rate in mr/hr.}$$

$$\phi_0 = \text{radiation flux in photons/cm}^2 \text{ sec before passage through a thickness } x \text{ of absorber.}$$

$$B(\mu x) e^{-\mu x} = \text{a dimensionless quantity which is that fraction of the original flux, both collided and uncollided, which succeeds in passing through the absorber.}$$

Equation 6 is the general expression for the dose rate due to the fraction  $B(\mu x) e^{-\mu x}$  of the original gamma flux  $\phi_0$  that has passed through a thickness  $x$  of an absorbing medium. In applying this equation, the shielding material and the geometry of the source must first be specified. For these considerations the shielding material will be  $\text{UPb}_3$ . The remaining required quantities, such as absorber thickness and photon energy, will be considered later when particular solutions of Equation 6 are found.

One of the most common geometric considerations used in shielding calculations is that of a point source. Assuming that this point source emits its radiation uniformly in all directions, then at some distance  $R$  away from the source, the radiation can be considered to pass evenly through the surface of a sphere of radius  $R$ . Thus, for a point source emitting  $S$  photons/sec, the flux at  $R$  will be  $S/4\pi R^2$  if there is no flux attenuation.

The source term  $S$  can be conveniently expressed in units of curies. A curie is defined as the quantity of any radioactive nuclide in which the number of disintegrations per second is  $3.7 \times 10^{10}$ . For the following treatment, it will be postulated that one gamma ray is emitted for each nuclear disintegration; therefore  $S$  can be written as

$$S = 3.7 \times 10^{10} \frac{\text{disintegrations}}{\text{sec curie}} \times C \text{ curies}$$

and the flux term  $\phi_0$  in Equation 6 becomes

$$\phi_0 = \frac{3.7 \times 10^{10} \text{ C} \frac{\text{disintegrations}}{\text{sec}} \times 1 \frac{\text{photon}}{\text{disintegration}}}{4\pi R^2 \text{ cm}^2}$$

or

$$\phi_0 = \frac{3.7 \times 10^{10} \text{ C}}{4\pi R^2} \frac{\text{photons}}{\text{cm}^2 \text{ sec}}$$

Inserting this expression for  $\phi_0$  into Equation 6, one has

$$D = \frac{E \mu_e}{1.967 \times 10^{-2}} \times \frac{3.7 \times 10^{10} \text{ C}}{4\pi R^2} \times B(\mu r) e^{-\mu r} \frac{\text{mR}}{\text{hr}}$$

Combining constants,



$$D = 1.5 \times 10^{11} \frac{E \mu_e C}{R^2} B(\mu r) e^{-\mu r} \frac{m r}{h r} \quad (7)$$

where  $r$  is the radius in cm of the shield and  $R$  is the distance in cm from the point source to the detecting device as indicated in Figure 1 below.

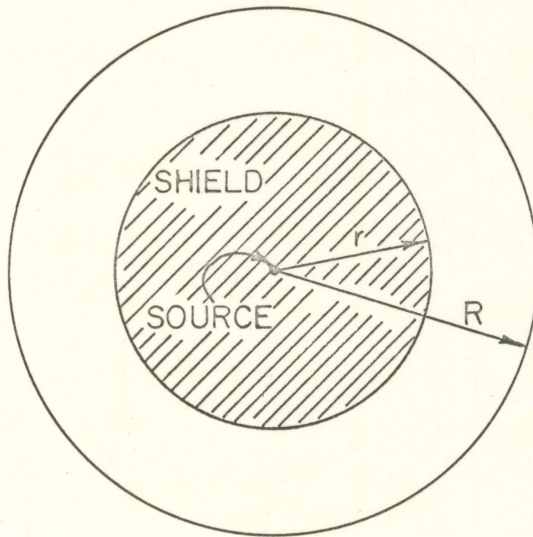


Figure 1. Point source

Equation 7 represents the general equation that will be used in analyzing the shielding properties of the  $UPb_3$  compound. In order to find an appropriate solution to this equation, consideration must be given to some of the individual terms.

Values for the energy absorption coefficient of air have been tabulated in Reference (19) for selected gamma-ray energies between 0.1 and 10.0 Mev. However, after gamma radiation

of some initial energy  $E_0$  has passed through a heavy shielding material, the gamma rays which emerge will no longer be monoenergetic, but will be represented by a spectrum of energies as a result of multiple scatterings. The exact composition of this spectrum is very difficult to predict, and the use of some particular absorption coefficient of air has little meaning. However, this coefficient is fairly constant for gamma-ray energies between 0.07 and 2.0 Mev, and a constant value of  $3.5 \times 10^{-5} \text{ cm}^{-1}$  is often used within this range which is accurate to within  $\pm 10$  percent (6). Using this value for  $\mu_e$ , Equation 7 becomes

$$D = 5.24 \times 10^6 \frac{EC}{R^2} B(\mu r) e^{-\mu r} \text{ mr/hr} \quad (8)$$

The next term to be considered is the linear absorption coefficient  $\mu$  of the  $\text{UPb}_3$  compound. The general method of finding  $\mu$  for a homogeneous mixture has been described by Goldstein (7). In general, the mass absorption coefficient  $\mu_m$  is written as

$$\mu_m \text{ cm}^2/\text{g} = \sum_i w_i \mu_i \quad (9)$$

where  $w_i$  is the corresponding weight fraction per molecule of the *i*th element. The gram-molecular weight of  $\text{UPb}_3$  is  $238.07 + 3(207.21)$  or 859.7 grams. The mass absorption coefficient of  $\text{UPb}_3$  is

$$\mu_m \text{ cm}^2/\text{g} = \frac{621.63 \text{ g Pb}}{859.7 \text{ g total}} \mu_m(\text{Pb}) + \frac{238.07 \text{ g U}}{859.7 \text{ g total}} \mu_m(\text{U})$$

$$= 0.7231 \mu_m(\text{Pb}) + 0.2769 \mu_m(\text{U}) \quad (10)$$

The linear absorption coefficient,  $\mu \text{ cm}^{-1}$ , can be obtained by multiplying the results of Equation 10 by the density of  $\text{UPb}_3$ . This density was reported by Teitel (16) as  $12.98 \text{ g/cm}^3$ . Values of both the mass and linear absorption coefficients are tabulated in Table 1. Reference (19) was used to obtain the appropriate values for the mass absorption coefficients of lead and uranium.

Table 1. Absorption coefficients

Gamma-ray energy, Mev	Lead <sup>a</sup> $\mu_m \text{ cm}^2/\text{g}$	Uranium <sup>a</sup> $\mu_m \text{ cm}^2/\text{g}$	$\text{UPb}_3$ $\mu_m \text{ cm}^2/\text{g}$	$\text{UPb}_3$ $\mu \text{ cm}^{-1}$
0.1	5.29	1.06	4.12	53.5
0.15	1.84	2.42	2.00	26.0
0.2	.896	1.17	.972	12.6
0.3	.356	.452	.383	4.97
0.4	.208	.259	.222	2.88
0.5	.145	.176	.154	1.99
0.6	.114	.136	.120	1.56
0.8	.0836	.0952	.0869	1.13
1.0	.0684	.0757	.0705	.915
1.25	.0569	.0615	.0581	.754
1.5	.0512	.0548	.0522	.678
2.0	.0457	.0484	.0464	.602

<sup>a</sup>Values of  $\mu_m$  for lead and uranium were taken from Reference ANL-5800.

Table 1. (Continued)

Gamma-ray energy, Mev	Lead <sup>a</sup>	Uranium <sup>a</sup>	UPb <sub>3</sub>	UPb <sub>3</sub>
	$\mu_m \text{ cm}^2/\text{g}$	$\mu_m \text{ cm}^2/\text{g}$	$\mu_m \text{ cm}^2/\text{g}$	$\mu \text{ cm}^{-1}$
3.0	.0421	.0445	.0427	.554
4.0	.0420	.0440	.0426	.553
5.0	.0426	.0446	.0431	.559
6.0	.0436	.0455	.0441	.572
8.0	.0459	.0479	.0464	.602
10.0	.0489	.0511	.0495	.643

The coefficients  $\mu_m$  of lead, uranium, and UPb<sub>3</sub> were compared by plotting  $\mu_m/\mu_0$  vs E as shown in Figure 2. Each of the curves was normalized to the  $\mu_0$  corresponding to some particular energy. For example, the lower curve has been normalized to the value of the absorption coefficient for 0.5 Mev photons; hence,  $\mu_m/\mu_0$  will be a ratio of the coefficients at other energies to that of the normalizing coefficient. A comparison of these curves indicates that the shape of the  $\mu_m$  curve for UPb<sub>3</sub> matches that of the elements uranium and lead. Therefore, in calculating the buildup factor, the UPb<sub>3</sub> compound can be considered as some element with an equivalent atomic number Z somewhere between 82 (lead) and 92 (uranium).

The equivalent atomic number for UPb<sub>3</sub> was found by plotting  $\mu_m/\mu_0$  vs Z for particular photon energies. In Figure 3, the  $\mu_m/\mu_0$  ratios were those normalized to 0.5 Mev. From

Figure 2. Normalized mass absorption coefficients of lead,  $UPb_3$ , and uranium

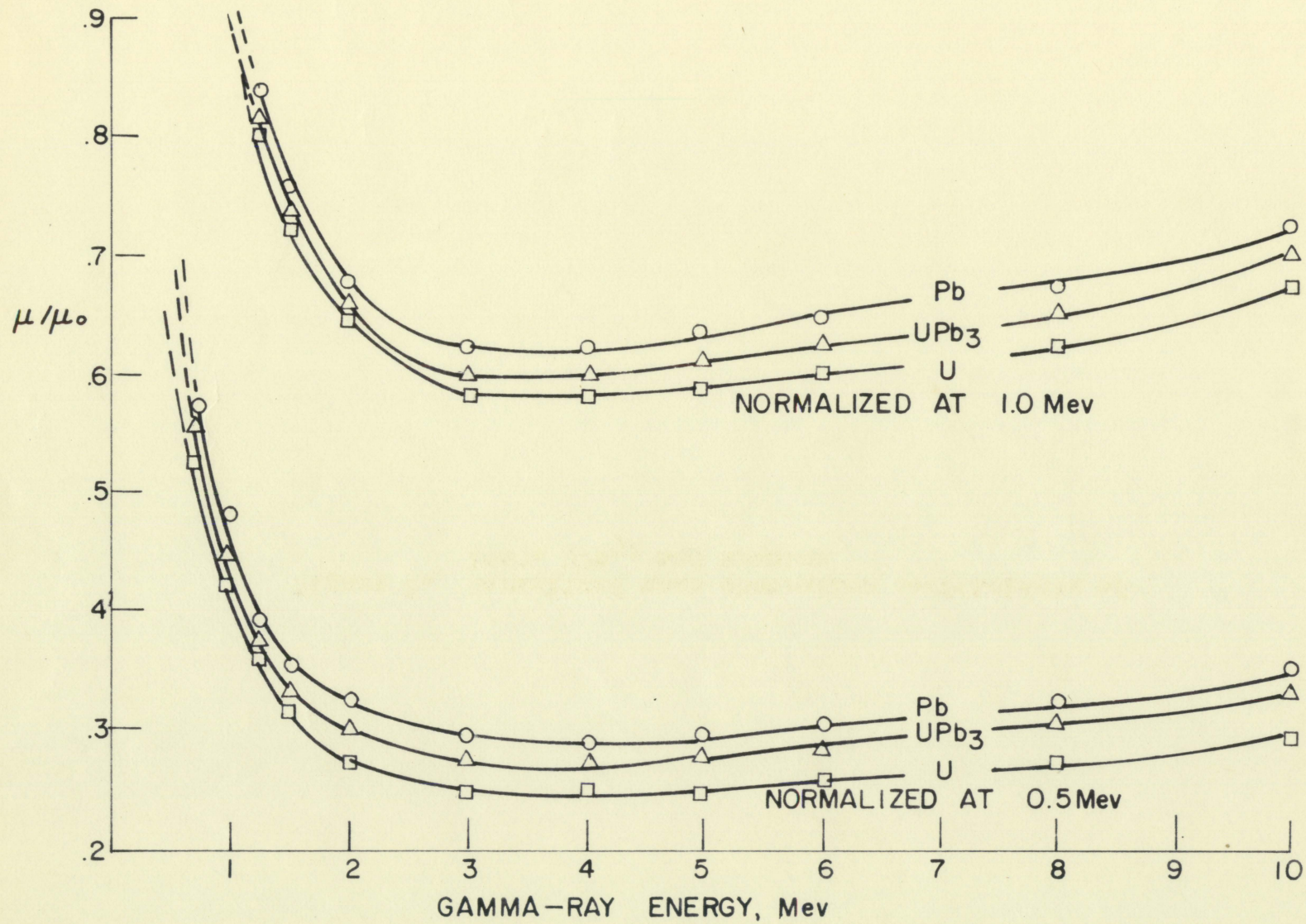
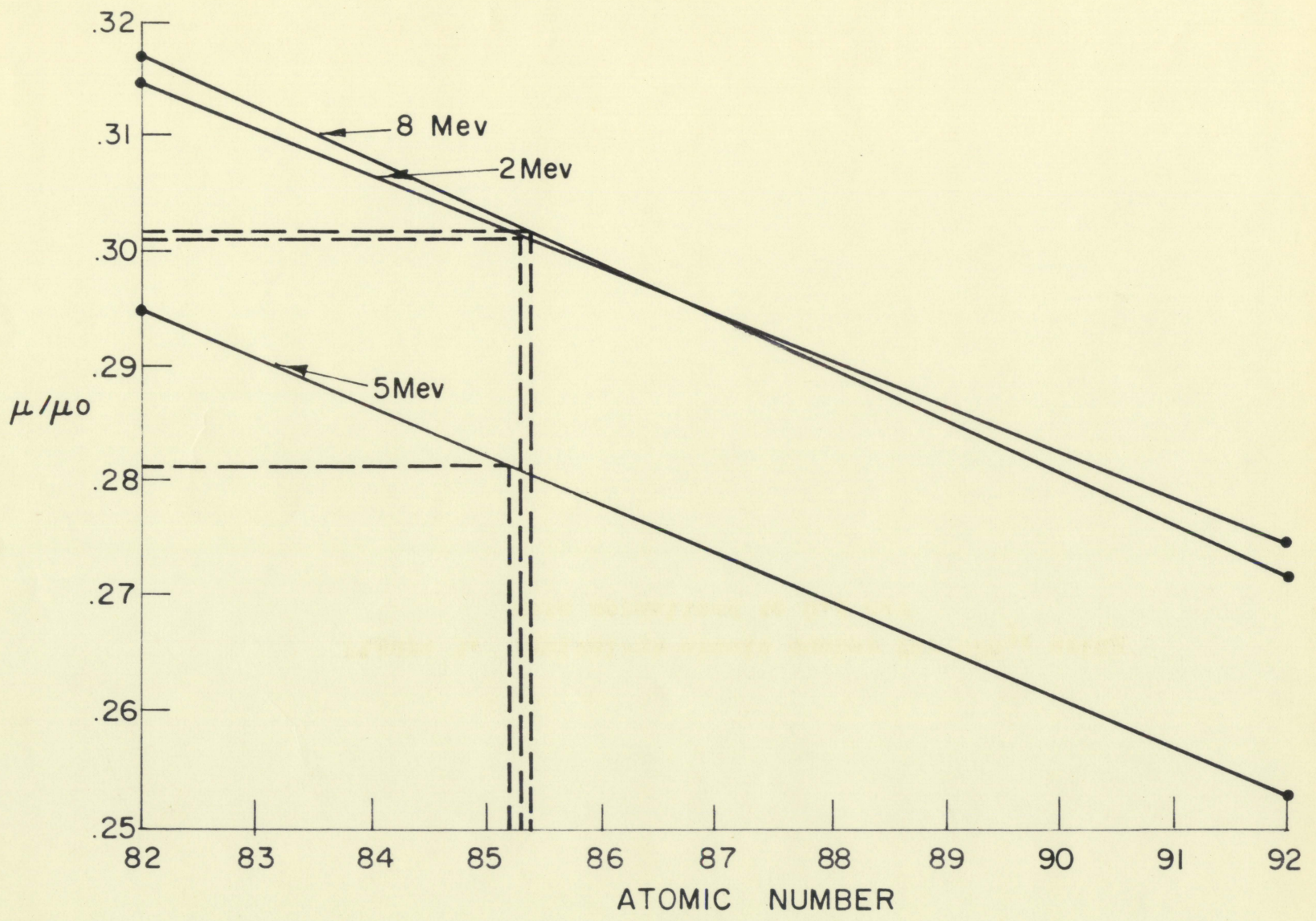


Figure 3. Equivalent atomic number for  $UPb_3$ , using  
data normalized to 0.5 Mev

FIGURE 2





these figures, it can be seen that the equivalent atomic number for  $UPb_3$  ranges from about 85.2 to 85.4, with a mean value near 85.3. Therefore this equivalent  $Z$  can be used to determine the infinite buildup factor for  $UPb_3$ .

In general, finding an accurate expression for the effects of scattered radiation has proven to be one of the most difficult problems in reactor shielding. A number of mathematical approaches have been tried with varying degrees of success. Of the different methods, one that has proven particularly useful is the technique known as the "moments method". This treatment was developed by L. V. Spencer and U. Fano (13), and was used in an extensive program of shielding calculations performed by Nuclear Development Associates, Inc. A part of their efforts (8) has been the calculation of dose buildup factors as a function of atomic number for a wide range of photon energies and depths of penetration. The results were shown on a series of graphs of buildup factor vs atomic number. From these graphs, buildup factors for  $UPb_3$  were found using an equivalent atomic number of 85.3. These values are tabulated in Table 2.

In Table 2 are listed the buildup factors for  $UPb_3$  that can be expected from gamma radiation of eight different energies ranging from 0.5 to 10.0 Mev. It can be seen that the value of the buildup factor is a function of the shield thickness, which is given in terms of the relaxation length  $\mu_0 r$ . Table 2 lists specific data for seven different relax-

Table 2. Dose buildup factors for UPb<sub>3</sub>

Gamma-ray energy, Mev	Relaxation length, $\mu_0 r$						
	1	2	4	7	10	15	20
0.5	1.31	1.49	1.69	1.89	2.12	2.44	2.64
1.0	1.43	1.71	2.18	2.84	3.48	4.41	5.25
2.0	1.46	1.80	2.47	3.55	4.54	6.17	8.00
3.0	1.40	1.73	2.42	3.61	5.07	7.95	11.73
4.0	1.33	1.62	2.26	3.52	5.18	9.25	16.44
6.0	1.23	1.46	1.94	3.23	5.32	13.46	29.36
8.0	1.17	1.39	1.80	2.81	4.94	14.14	36.14
10.0	1.15	1.31	1.67	2.48	4.21	12.70	37.67

ation lengths in a range from 1 to 20. A better understanding of the data can be obtained by referring to Figure 4 and 5, which are plots of the buildup factor vs shield thickness. The general position of these points indicates that they could perhaps be represented by a sum of exponential terms. With such an expression, the specific data in Table 2 could be expanded into the form of a general equation which could be applied to find buildup factors for the intermediate gamma energies and shield thicknesses. A general equation of the form

$$B(\mu r) = A e^{-\alpha \mu r} + (1 - A) e^{-\beta \mu r} \quad (11)$$

has been suggested by Taylor (14) in which the three parameters  $A$ ,  $\alpha$ , and  $\beta$  are energy dependent. This equation has been used by other experimenters with very good results.

Figure 4. Dose buildup factors for  $UPb_3$  for gamma-ray energies of 0.5, 1.0, and 2.0 Mev.

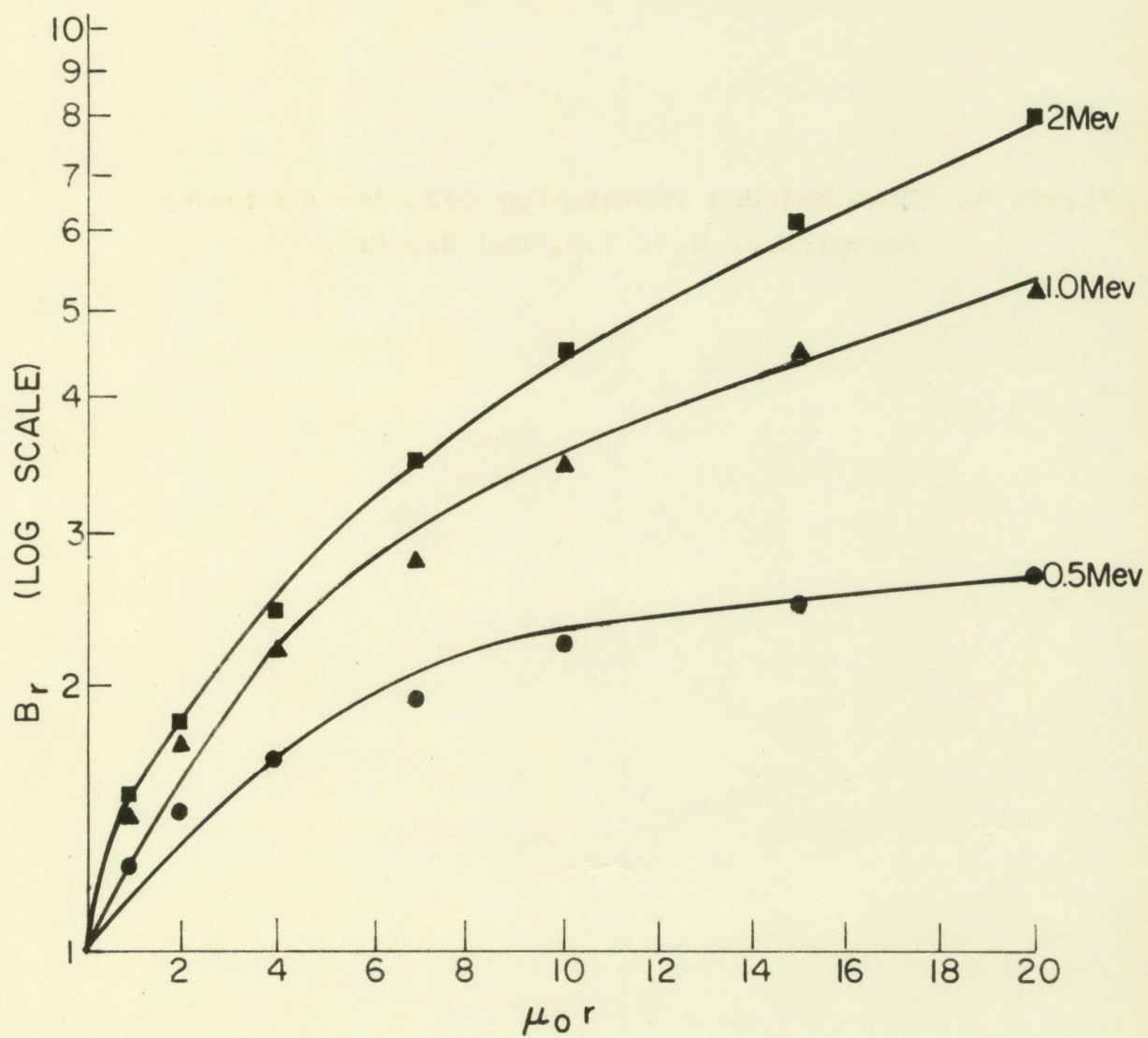
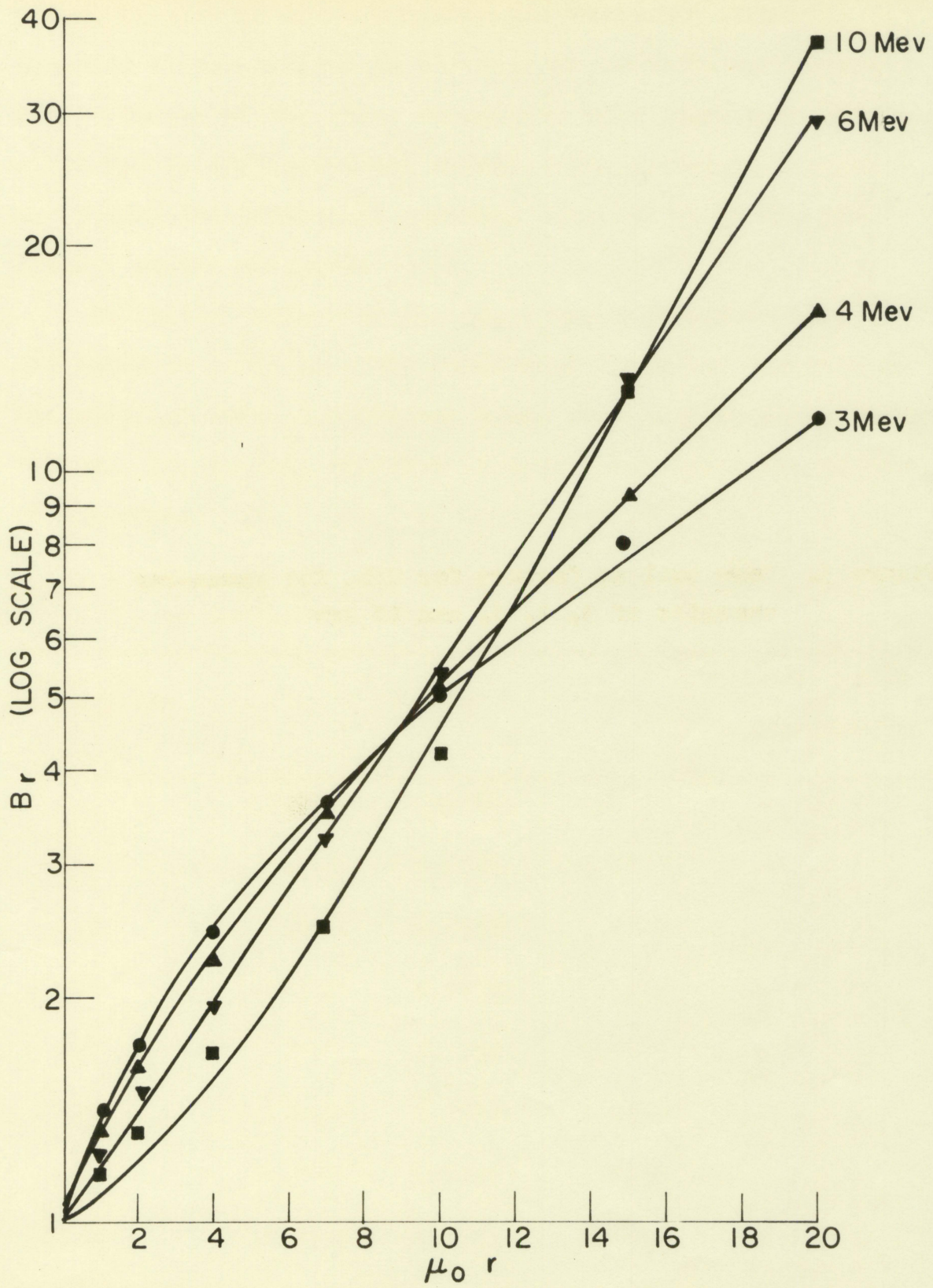


Figure 5. Dose buildup factors for  $UPb_3$  for gamma-ray energies of 3, 4, 6, and 10 Mev



Hence, in view of its simplicity and indicated accuracy, Equation 11 was applied to the data in Table 2, and appropriate values of the three parameters were found that gave a close fit to the individual points. The curves in Figures 4 and 5 show the results of applying Equation 11 to the particular photon energies.

Individual values of the three parameters have been tabulated in Table 3. This table also lists the position on the curve at which the maximum error results from using these values. In general, Equation 11 gave a very good fit to most of the data.

Table 3. Coefficients for Taylor's buildup factor equation as applied to  $UPb_3$

Gamma-ray energy, Mev	A	-a	$\beta$	Worst error of approximation %
0.5	1.920	0.0160	0.2350	- 7.0 % at $\mu_0 r = 1$
1.0	2.700	0.0342	0.1794	- 3.9 % at $\mu_0 r = 1$
2.0	2.850	0.0516	0.1720	4.2 % at $\mu_0 r = 4.5$
3.0	2.500	0.0774	0.1117	- 3.1 % at $\mu_0 r = 15$
4.0	1.700	0.1136	0.1400	- 2.3 % at $\mu_0 r = 1$
6.0	1.000	0.1700	0.0000	- 4.8 % at $\mu_0 r = 15$
8.0	0.700	0.1976	0.0558	7.0 % at $\mu_0 r = 7$
10.0	0.475	0.2195	0.0674	- 8.8 % at $\mu_0 r = 2$

The values found for the terms  $A$ ,  $-\alpha$ , and  $\beta$  are shown on Figures 6, 7, and 8, respectively. The points of Figures 6 and 7 have been joined with a broken line. However, the reader is cautioned about using values on these curves which are not close to the plotted points since the behavior of the curves between the points has not been calculated. If the buildup factor for intermediate gamma-ray energies is desired, the following procedure is recommended:

1. From the data in Table 2, plot the buildup factor as a function of the gamma-ray energy. This will result in a family of 7 curves, with one curve for each relaxation length given.
2. From these curves, find the value of the buildup factor at each relaxation length for the particular energy of interest.
3. The buildup factor at penetrations up to 20 relaxation lengths can then be found by plotting the values of the buildup factor found in Step 2 as a function of relaxation length.

The presentation of the buildup factor has, up to this point, considered only that for an infinite medium. That is, the buildup at some depth  $t$  in a shield of infinite thickness would be a function of two factors: a probability that gamma radiation would penetrate to depth  $t$ , and a probability that some radiation would penetrate beyond this depth and be scattered back to  $t$ . The buildup factors for the finite thick-

*Skip*



Figure 6. Values for the constant, A

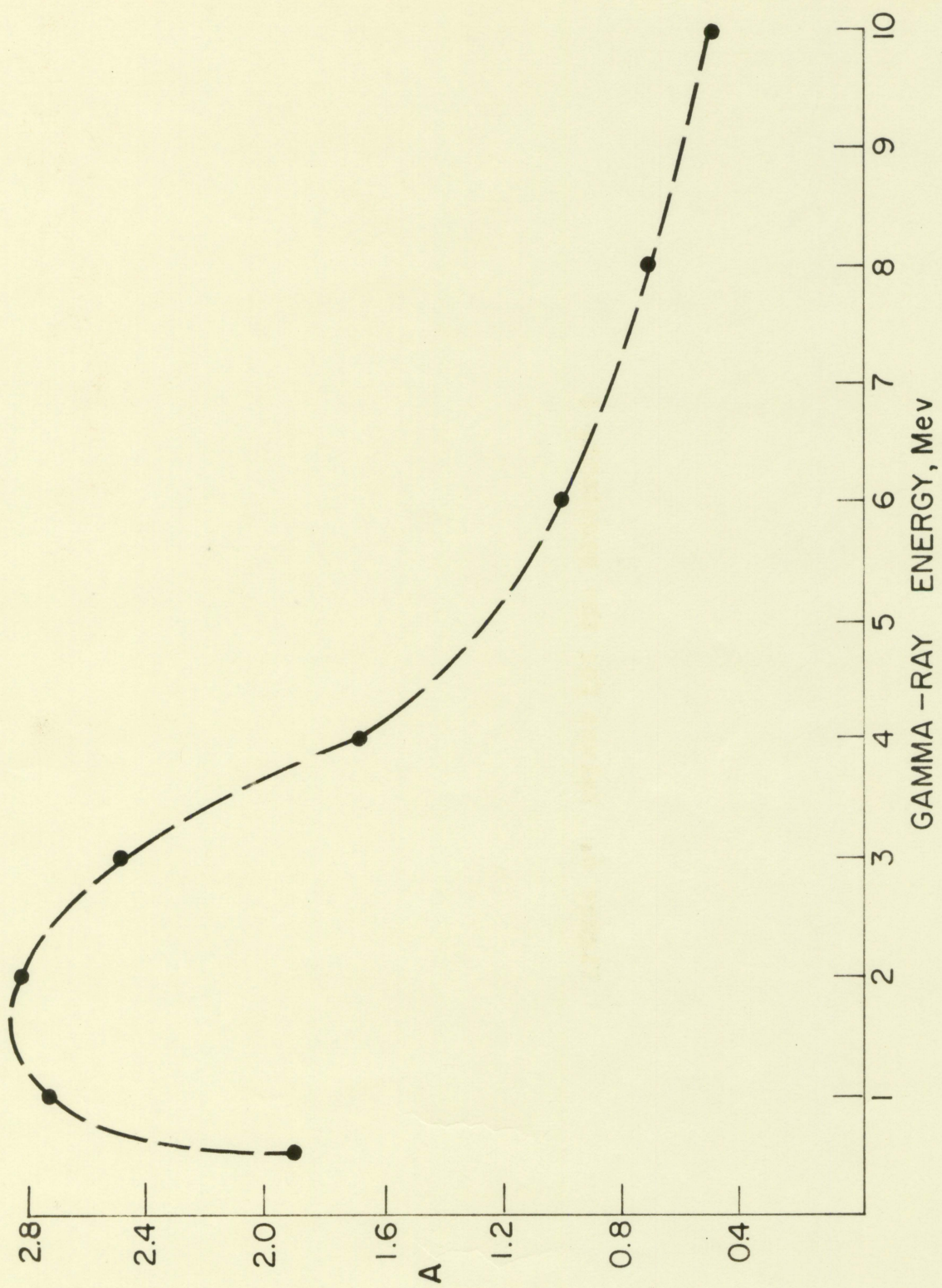


Figure 7. Values for the constant, - a

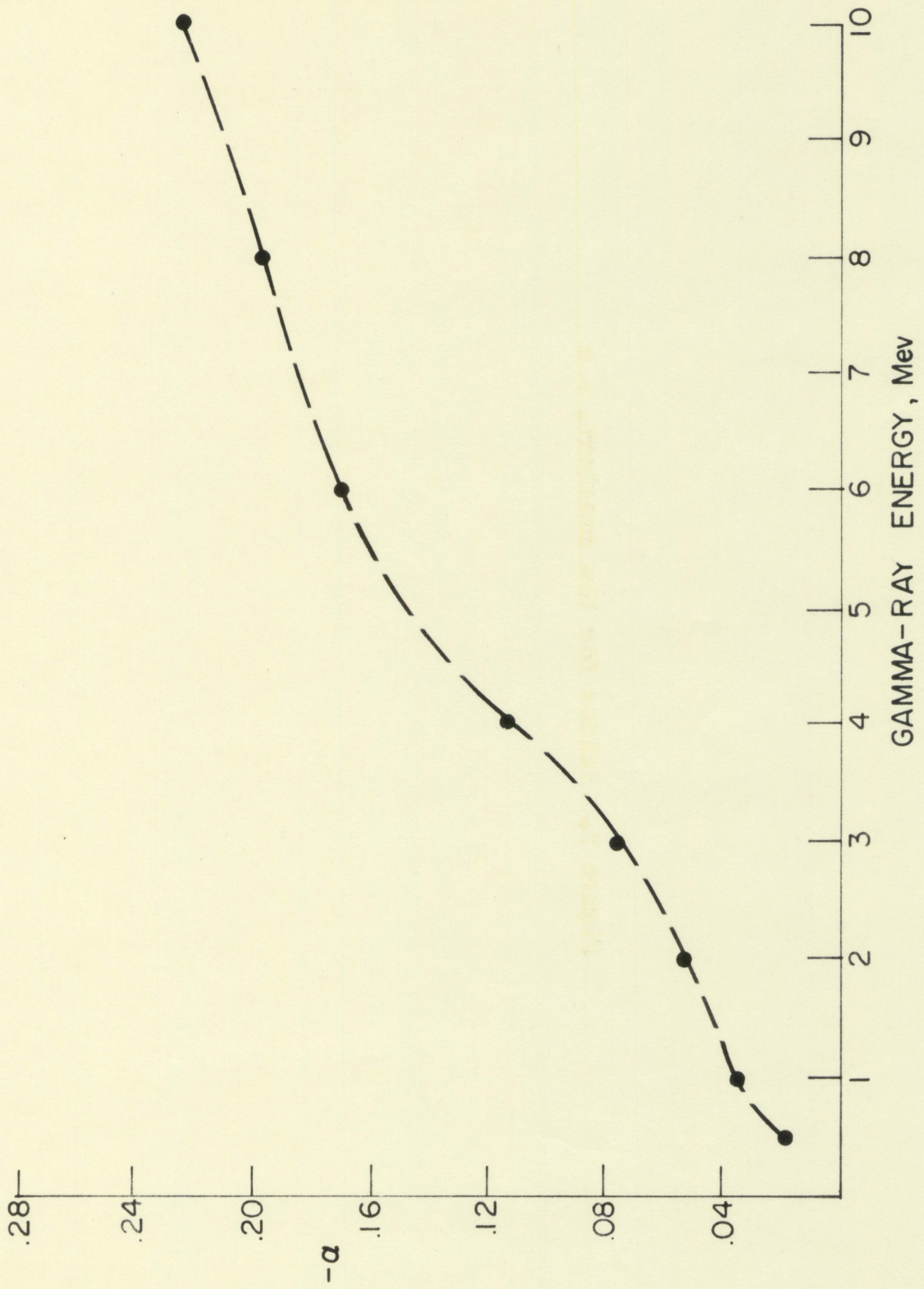
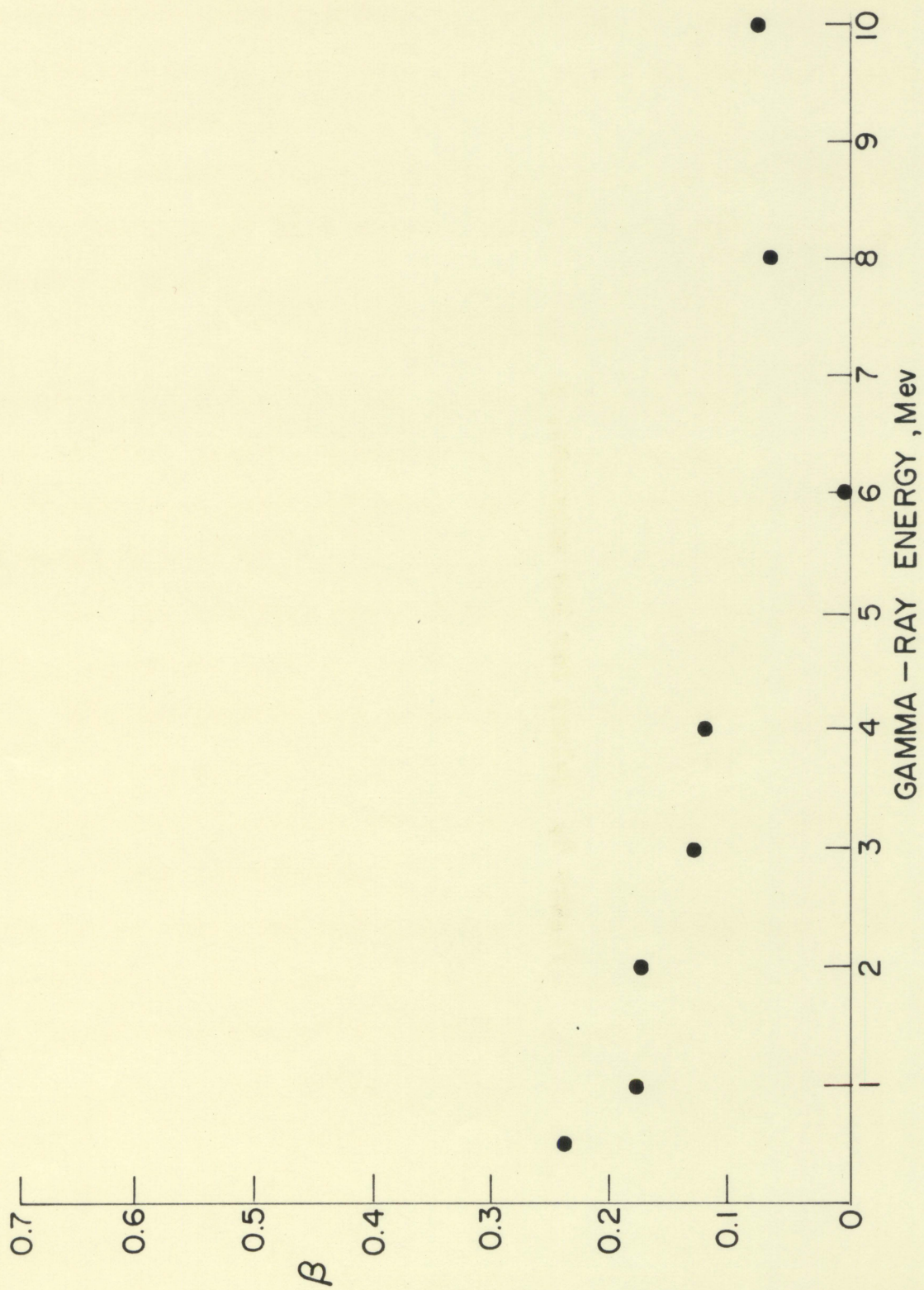


Figure 8. Values for the constant,  $\beta$



nesses that are considered in this thesis should then require some correction factor since  $t$  is now at the surface of the medium and hence, there would be no contributions from back-scattered radiation.

Berger and Doggett (2) have investigated this problem using Monte Carlo techniques. They consider the correction factor  $k(t)$  as

$$k(t) = \frac{B(t,t) - 1}{B(t,\infty) - 1} \quad (12)$$

where  $B(t,t)$  and  $B(t,\infty)$  are the buildup factors for a finite and infinite medium, respectively. The findings of these investigations have indicated that  $k(t)$  differs substantially from unity only if

1. the material has a fairly low (less than iron) atomic number;
2. the photons are in the low-energy (less than 1 Mev) range;
3. short penetrations (less than 4 mean free paths) are considered.

For the material and the energies that are being considered here,  $k(t)$  is very close to unity; hence this factor will not be included in the calculations which follow.

## IV. PROCEDURE

Equation 8 will be used in the following analyses of the  $UPb_3$  shielding material. Recall that

$$D = 5.24 \times 10^6 \frac{EC}{R} B(\mu r) e^{-\mu r} \text{ mr/hr}$$

is applicable if

1. the energy absorption coefficient of air is taken as  $3.5 \times 10^{-5} \text{ cm}^{-1}$ ;
2. one gamma ray is emitted for each nuclear disintegration;
3. the gamma-ray source is considered to be concentrated at a point.

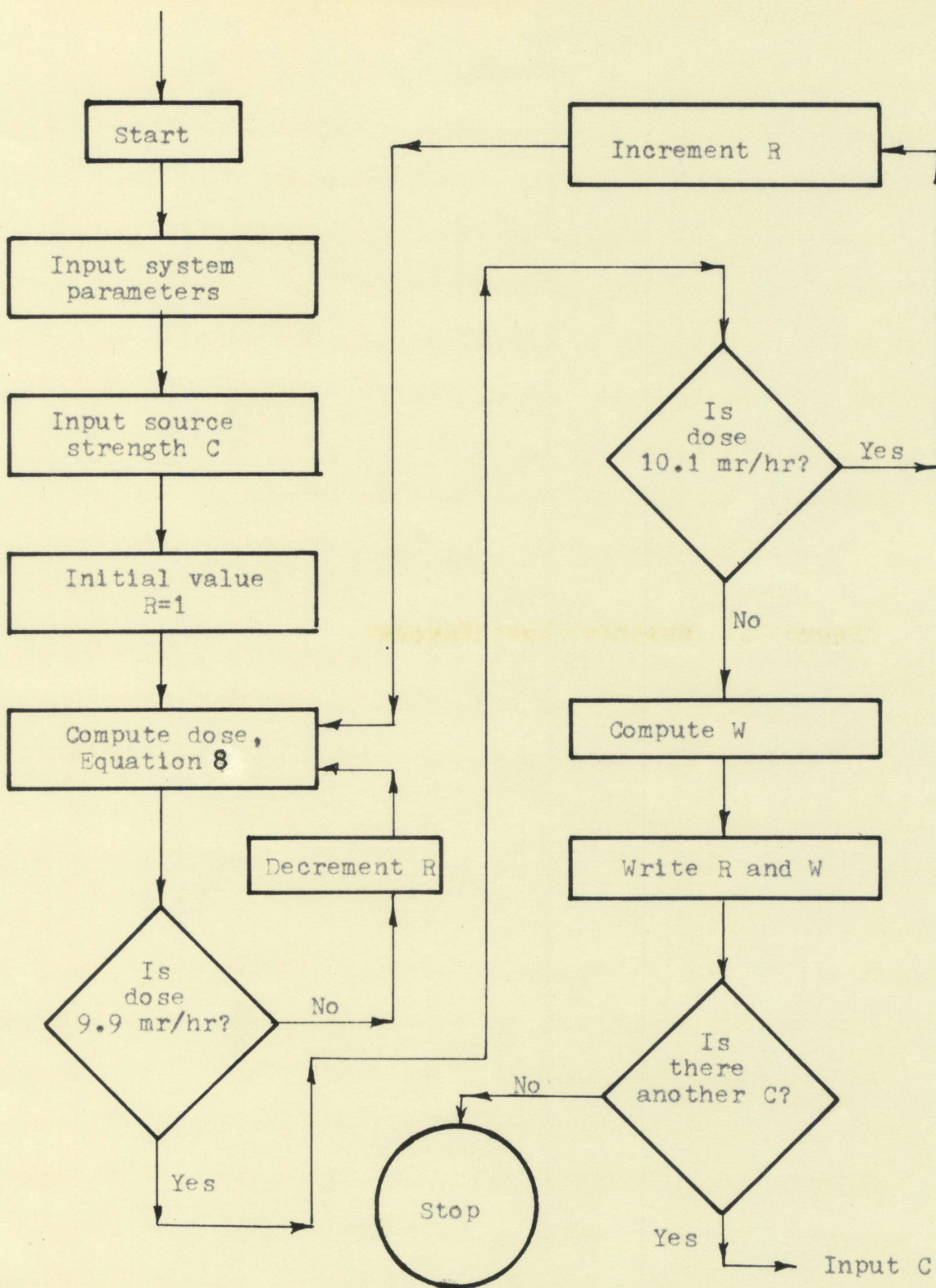
Solutions to the above equation were found using the IBM 360/50 computer. A dose rate  $D$  having a range between 9.9 and 10.1 mr/hr was specified, and the required radius  $R$  was then calculated. Since this dose rate is at the surface of the sphere, then  $R = r$  in Equation 8. This equation was evaluated for source strengths of 1, 10, 100, 1000, and 5000 curies, and for gamma-ray energies of 0.5, 1, 2, 3, 4, 6, 8, and 10 Mev. The values for the input system parameters  $A$ ,  $\alpha$ , and  $\beta$  in the buildup factor expression (Equation 11) were taken from Table 3, and values for the absorption coefficients  $\mu$  were taken from Table 1. Thus in Equation 8,  $R$  was the only variable; it was assigned an initial value of 1 cm, and then varied until the dose rate fell into the de-



sired range. After the required value of  $R$  had been determined, the shield weight  $W$  was calculated and the corresponding values of  $R$  and  $W$  were printed. A flow diagram of the computer program used is shown in Figure 9.

This same procedure was repeated for the elements lead and uranium so as to aid in comparing the data from the  $UPb_3$  compound with that of its component elements. The corresponding values of  $A$ ,  $\alpha$ , and  $\beta$  for lead and uranium were taken from data compiled by Buscaglione and Manzini (4). Reference (19) furnished the necessary data for the linear absorption coefficients.

Figure 9. Computer flow diagram



## V. DISCUSSION OF RESULTS

The information obtained from the computer program has been tabulated in Tables 4 and 5. The data in Table 4 refer only to the  $UPb_3$  compound, and is a compact analysis of the shielding requirements that will be met in using  $UPb_3$  as a shielding material. This information is shown in Figures 10 and 11 as plots of the shield radius vs the source strength. An interesting feature of these curves is their nearly straight line variation over the entire range of investigation. This consistency permits an accurate determination of shielding requirements for the intermediate source strengths.

Referring to Figure 11, it can be seen that gamma radiation with initial energy of 6.0 Mev requires the maximum amount of shielding; the material necessary to shield against 8.0 and 10.0 Mev gammas is somewhat less. Similar results were observed with lead and uranium. It can also be noted that for  $UPb_3$  the values at 4.0 and 8.0 Mev are almost identical.

The data in Table 5 are a comparison of the shield weights of lead,  $UPb_3$ , and uranium required for gamma rays of various energies. The results for a few selected energies are shown in Figures 12, 13, and 14 which are plots of the shield weight vs the source strength. The general shape of these curves is very similar and they indicate a smoothly varying function. Because of the very slow change in their slopes,

Table 4. Shield radius (in cm) of  $UPb_3$  required for a surface dose rate of between 9.9 and 10.1 mr/hr.

Gamma-ray energy, Mev	Point source strength, curies				
	1	10	100	1000	5000
0.5	5.04	6.04	7.06	8.10	8.83
1.0	10.60	12.81	15.06	17.37	18.99
2.0	16.24	19.65	23.14	26.69	29.21
3.0	18.19	22.01	25.92	29.89	32.70
4.0	18.77	22.72	26.76	30.88	33.80
6.0	19.10	23.15	27.30	31.54	34.55
8.0	18.76	22.71	26.78	30.95	33.90
10.0	17.94	21.72	25.63	29.63	32.47

Table 5. Material comparison of shield weight, in kilograms, required for a surface dose rate of between 9.9 and 10.1 mr/hr

Material	Point source strength, curies				
	1	10	100	1000	5000
<u><math>E_0 = 0.5</math> Mev</u>					
Lead	9.8	17.2	28.0	42.7	55.7
$UPb_3$	6.9	12.0	19.1	28.9	37.4
Uranium	2.7	4.6	7.1	10.6	13.6
<u><math>E_0 = 1.0</math> Mev</u>					
Lead	86	154	254	392	514
$UPb_3$	65	114	186	285	373
Uranium	30	52	83	125	162
<u><math>E_0 = 2.0</math> Mev</u>					
Lead	300	540	890	1374	1806

Table 5. (Continued)

Material	Point source strength, curies				
	1	10	100	1000	5000
UPb <sub>3</sub>	233	412	674	1034	1355
Uranium	118	204	327	493	6640
<u>E<sub>0</sub> = 3.0 Mev</u>					
Lead	420	753	1239	1911	2507
UPb <sub>3</sub>	327	580	947	1452	1902
Uranium	168	289	461	695	899
<u>E<sub>0</sub> = 4.0 Mev</u>					
Lead	460	825	1356	2091	2745
UPb <sub>3</sub>	360	637	1042	1601	2099
Uranium	188	324	516	776	1005
<u>E<sub>0</sub> = 6.0 Mev</u>					
Lead	478	862	1430	2225	2939
UPb <sub>3</sub>	379	674	1106	1706	2242
Uranium	197	339	544	823	1071
<u>E<sub>0</sub> = 8.0 Mev</u>					
Lead	445	807	1350	2120	2818
UPb <sub>3</sub>	359	637	1045	1611	2118
Uranium	184	318	510	774	1009
<u>E<sub>0</sub> = 10.0 Mev</u>					
Lead	389	701	1168	1828	2426
UPb <sub>3</sub>	314	557	916	1415	1861
Uranium	161	278	446	678	884

Figure 10. UPb<sub>3</sub> shield radius requirements for gamma-ray energies of 0.5, 1.0, and 2.0 Mev

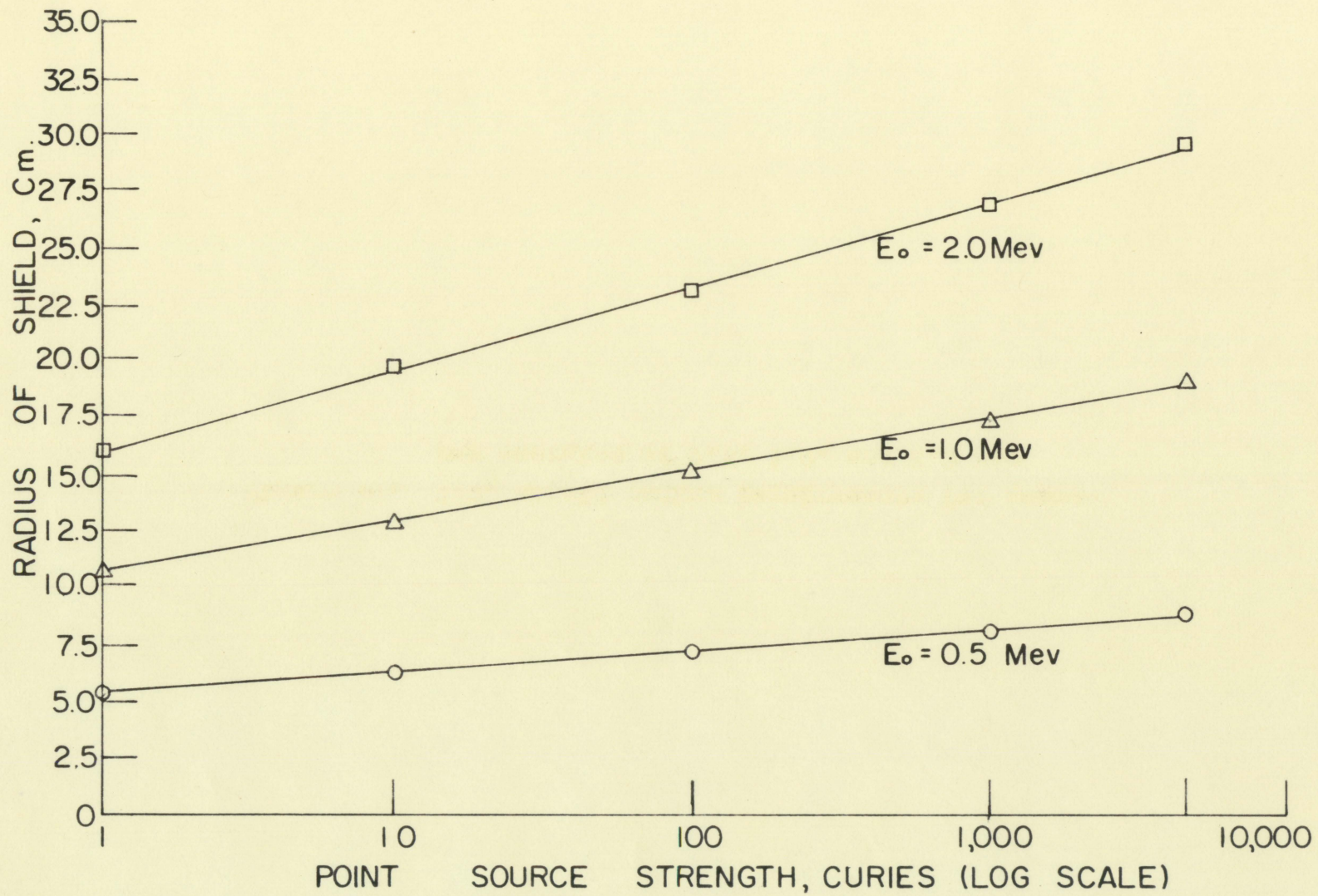




Figure 11.  $UPb_3$  shield radius requirements for gamma-ray energies of 3.0, 4.0, 6.0, 8.0, and 10.0 Mev

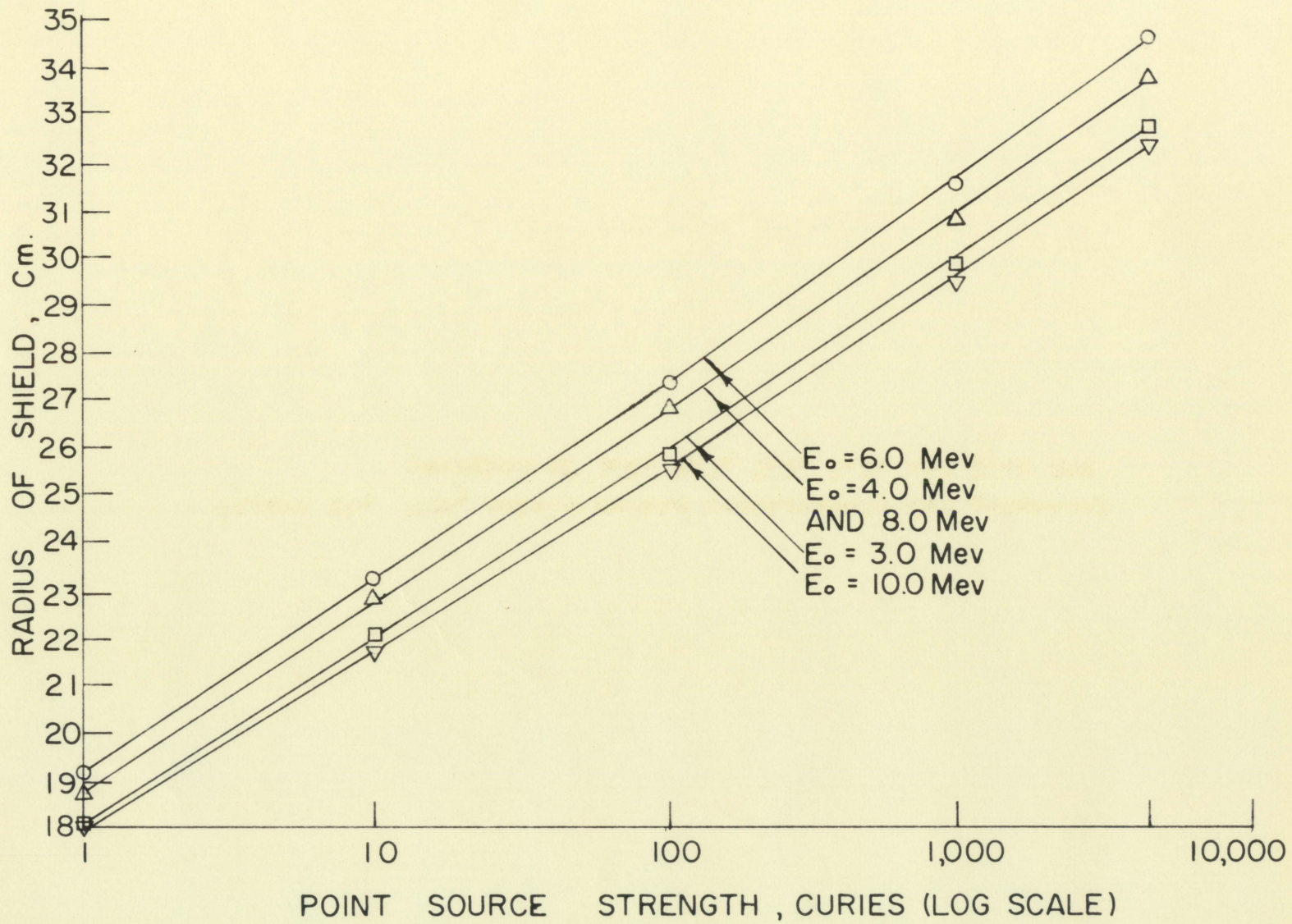


Figure 12. Material comparison of shield weight requirements for gamma rays of energy 1.0 Mev

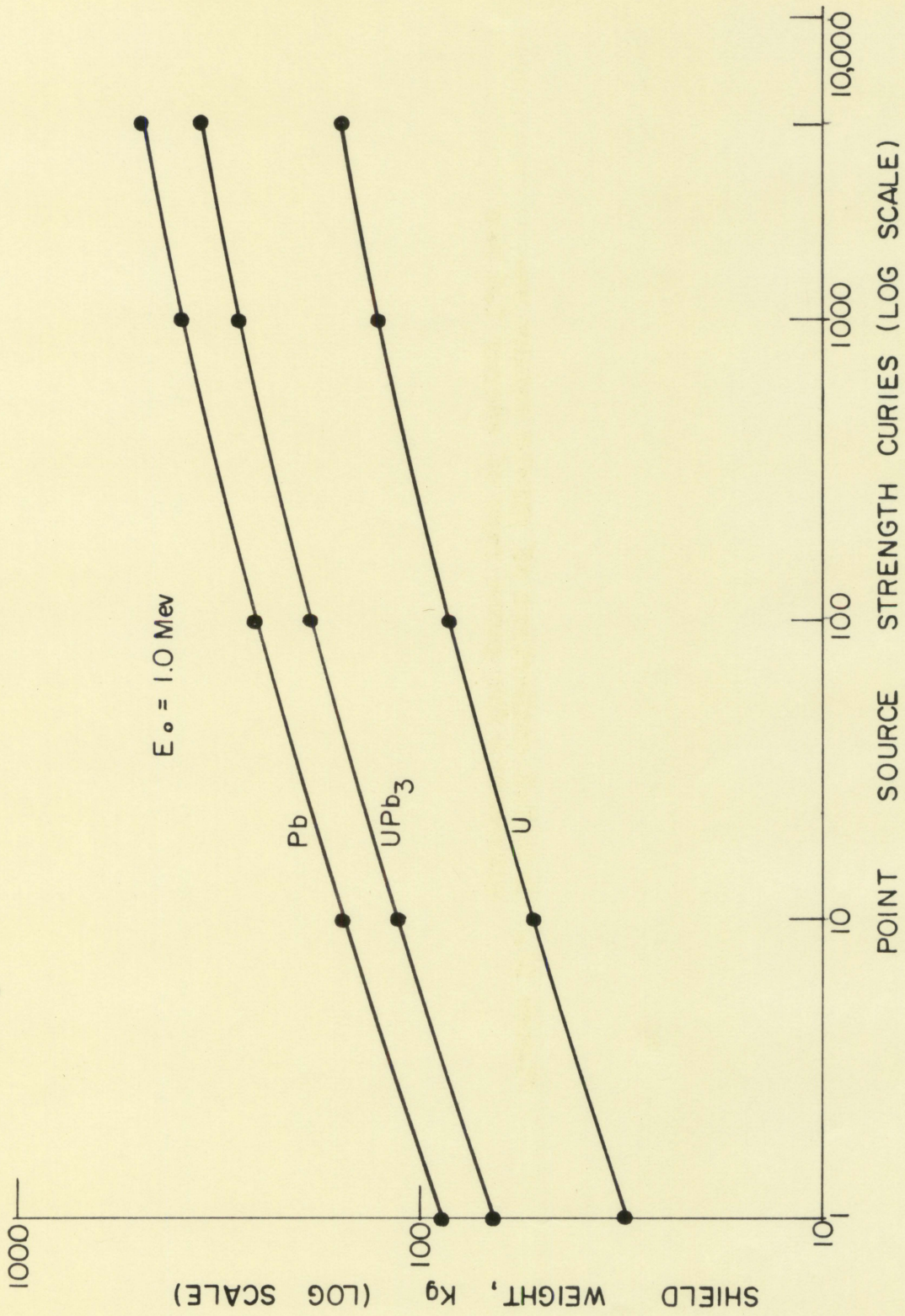


Figure 13. Material comparison of shield weight requirements for gamma rays of energy 6.0 Mev

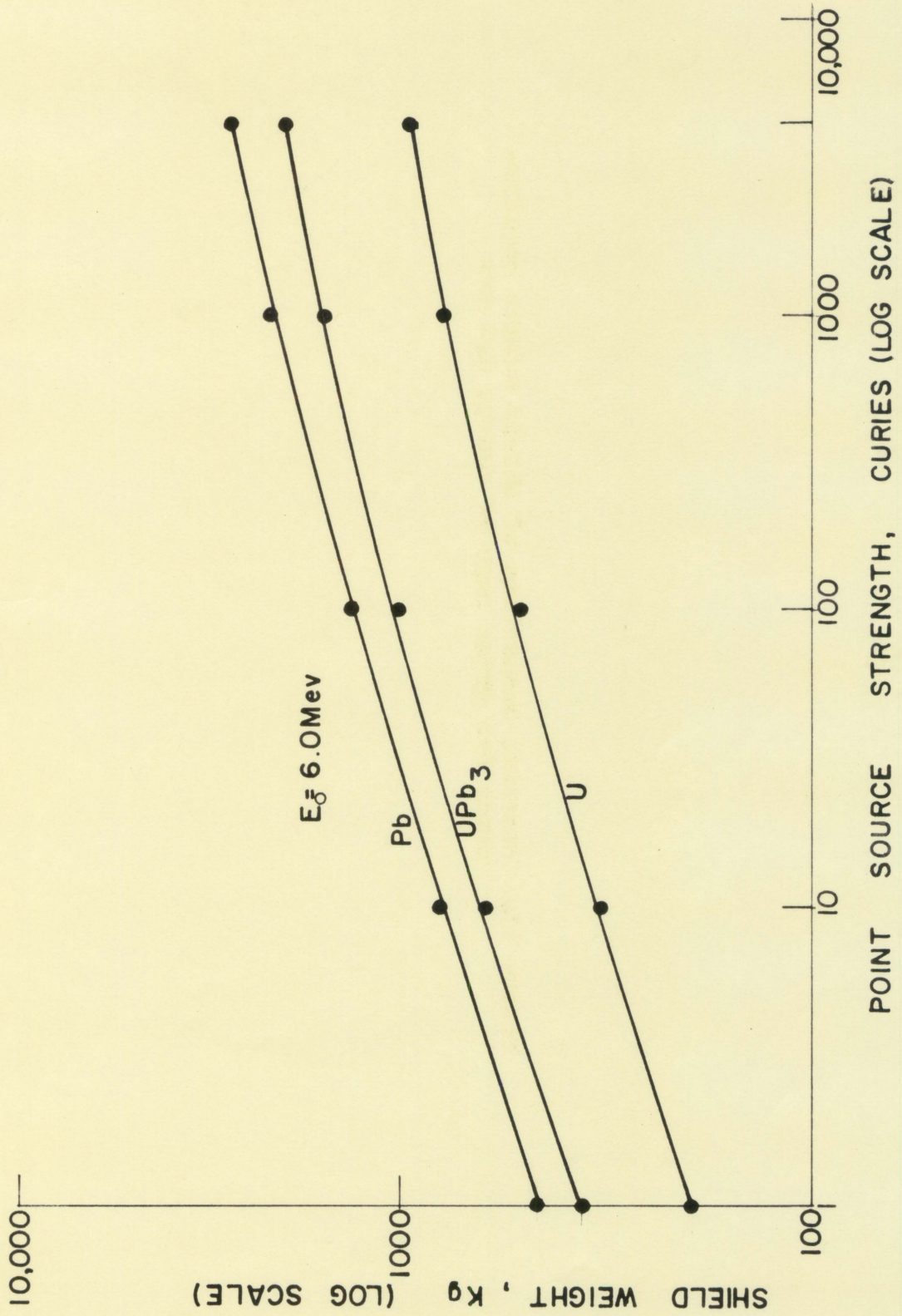
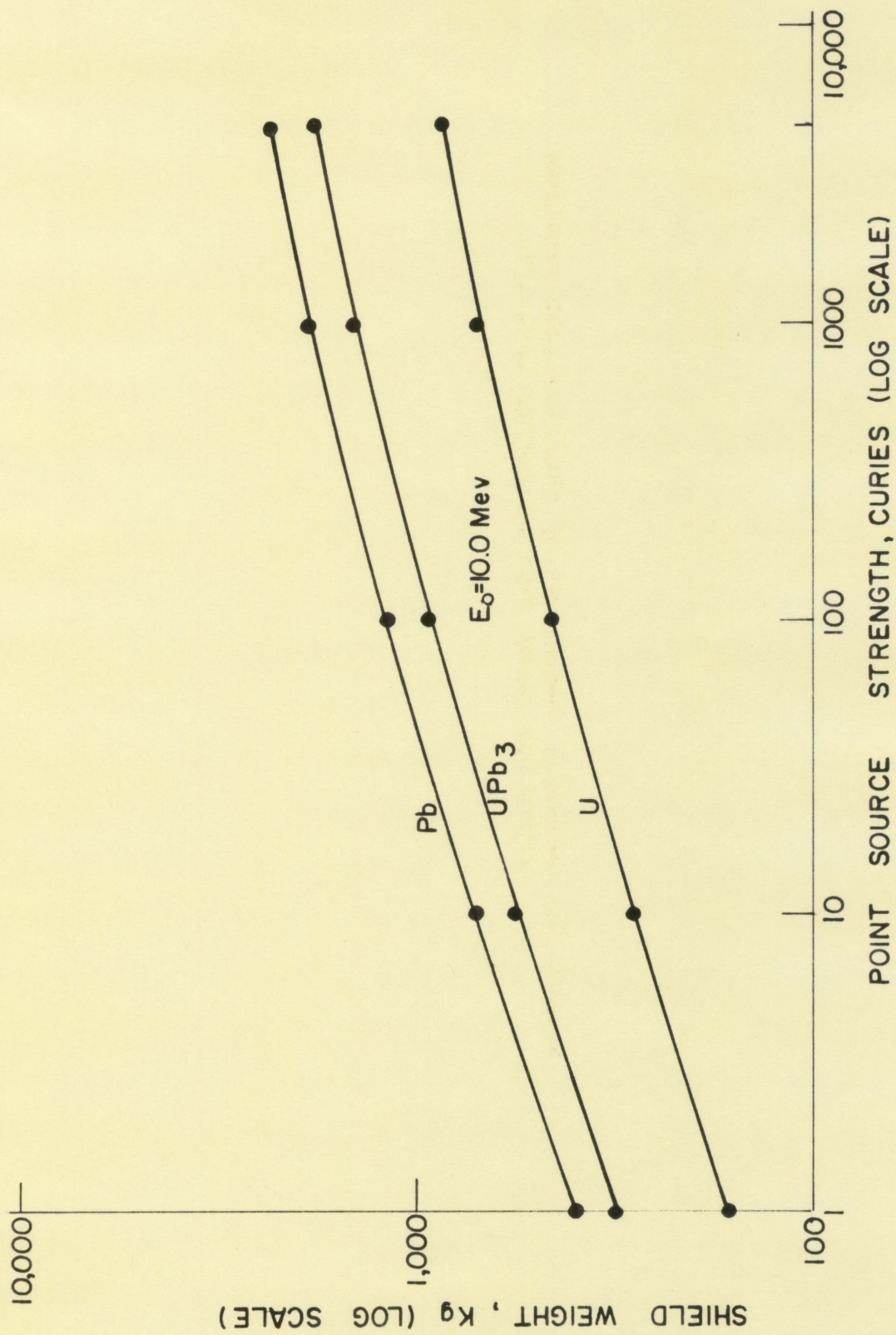


Figure 14. Material comparison of shield weight requirements for gamma rays of energy 10.0 Mev





these curves also provide an accurate means of determining shielding requirements for source strengths other than those that have been calculated.

The source energies and activities that have been considered in these calculations should be of sufficient range to cover many of the problems encountered in shielding work. Because any arbitrary radiation source can be considered as a distribution of point sources, this information can be readily adapted to a variety of source geometries such as line or plane sources. Methods for making these transformations are given in a number of standard texts including Glasstone and Sesonske (6) and Jaeger (10).

The data as presented here are applicable only to homogeneous shields of  $UPb_3$ . For shield compositions other than this, i.e.,  $UPb_3 + UPb$  or  $Pb + UPb_3$ , modifications would be necessary, dependent upon the relative amounts of each component. However, since alloys with uranium content less than the composition of  $UPb_3$  have a melting point which is essentially that of pure lead, little would be gained in their use as a shield. In addition, as previously noted, the pyrophoric nature of  $UPb$  and the difficulties encountered in preparing alloys of higher uranium content seem to rule out compositions which differ greatly from  $UPb_3$ .

In considering the uranium content of a  $UPb_3$  shield, economic reasons would quite naturally indicate that depleted uranium be used. Although the price and availability of this

material may change, it appears that finding uses for depleted uranium has been a problem. A fairly recent report (22) stated: "Rapidly rising stockpiles of depleted uranium, a byproduct in the manufacture of fissionable  $U^{235}$ , have prompted a search for uses for the depleted  $U^{238}$ ."

## VI. CONCLUSIONS

The results of this analysis have demonstrated that the use of  $UPb_3$  in place of lead will effect a substantial reduction in both the size and weight of a shield. The savings are quite significant when considering the high-energy gammas and large source strengths. If size and weight were the only considerations in shielding, uranium would be quite superior to either lead or  $UPb_3$ ; however, a great many other factors including cost, ease of fabrication, and compatibility with surroundings must also be considered. The features of  $UPb_3$  which make it worthy of consideration as a shielding material can be summarized as follows:

1. The high melting point of  $UPb_3$  gives it a definite advantage over lead.
2. The foregoing analysis has demonstrated that  $UPb_3$  should be a very effective shield against gamma radiation.

In addition, indications are that this compound can be produced on a price-competitive basis.

Although a number of different aspects of  $UPb_3$  have required consideration in the discussion, the analysis of this compound has been concerned only with its shielding properties. No economic or feasibility analysis has been attempted; their inclusion here should only be interpreted as trends indicated by the literature. Clearly, many questions about  $UPb_3$  remain

unanswered. Properties such as tensile strength, ductility, specific heat, thermal conductivity and expansion coefficients are yet to be determined. These properties must be proven to be compatible with shielding requirements before  $UPb_3$  can be termed acceptable as a shielding material. Such questions serve as worthwhile subjects for future investigations.

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