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GRAPHICAL AIDS IN THE CALCULATION OF THE SHIELDING REQUIREMENTS FOR SPENT U<sup>235</sup> FUEL

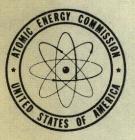
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Printed in USA. Price \$1.75. Available from the Office of Technical Services, Department of Commerce, Washington 25, D. C. GRAPHICAL AIDS IN THE CALCULATION OF THE SHIELDING REQUIREMENTS FOR SPENT U<sup>235</sup> FUEL

> BY R. L. ASHLEY

# **ATOMICS INTERNATIONAL**

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#### ABSTRACT

The shielding requirements for spent  $U^{235}$  fuel (or gross fission products) can be determined simply and rapidly by using a method in which a single predetermined energy represents the entire fission product gamma-ray spectrum. The data presented herein, in the form of a series of graphs, can be used to obtain the value of this energy. Having this energy, the requisite shielding calculations can be performed using linear absorption coefficients and buildup factors which correspond to this energy. In essence, this method for determining fission product shielding requirements has as its basis a scheme termed the "effective energy" concept in which, as noted, the entire fission product gamma-ray spectrum is replaced by a single gamma-ray line, behaving exactly as the entire spectrum for the given parameters of irradiation and decay time, shield material and thickness. The equivalence stems from the procedure utilized in determining the effective energy, i.e., the gamma rays comprising the fission product spectrum present at the time of interest are individually attenuated through a given shield and the overall dose rate attenuation determined. The effective energy corresponds to the particular gamma ray which, if present in the same intensity as the fission product spectrum, would result in an equivalent dose rate when confronted by the same shield.

The fission product spectrum considered in this report consists, for the most part, of gamma rays with energies equal to or greater than 1.60 Mev. Since the range of shield thicknesses analyzed is in the thick-shield category, it was unnecessary to consider gamma rays with lower energies as they would contribute negligibly to the dose rate at the shield surface. The decay schemes of the particular isotopes emitting gamma rays above 1.60 Mev are an important part of this analysis and, due to the limited amount of information available, have mostly been inferred from data in the literature. Details of the decay schemes utilized are discussed in the report.

The range of exposure times considered in the graphs presented is from 100 hours to 300 days, with decay times of from 20 minutes to 300 days. Monolithic shields of 6 to 12 inches of lead, 6 to 12 feet of water, 3 to 6 feet of ordinary concrete, and 2 to 4.25 feet of magnetite concrete are analyzed.

A comparison of the results obtained using the effective energy method with several experimental measurements indicates that this method yields data which are consistently lower than measured values. The ratio of calculated to measured values generally is greater than 0.5, but less than 0.9, with the greater deviation occurring for cooling times less than 45 minutes. The difference can, for the most part, be attributed to the inaccuracy of the inferred decay schemes and errors associated with the techniques used in performing the shielding calculations which were required in order to make the comparisons with the experimental data. The agreement achieved between calculation and experiment was very satisfactory, indicating that the effective energy method is not only simple and rapid, but extremely accurate as well.

## I. INTRODUCTION

#### A. METHODS PRESENTLY AVAILABLE AND THEIR LIMITATIONS

The problem of determining the shielding requirements for spent fuel is one that confronts the shielding engineer many times during the course of design of any reactor system. In the past, the solution of this type of problem could be accomplished by a number of different methods which, in general, require examination of the structure of the  $U^{235}$  fission product gamma-ray spectrum present at the particular time of interest. Analysis of this structure (or spectrum) has been performed by Moteff, <sup>1</sup> Clark, <sup>2</sup> and Beeley, et al. <sup>3</sup> However, in order to establish the dose rate at the surface of a given shield using their data, it is required that shielding calculations be performed for a number of different energy bands (or groups), the resultant dose rate being the sum of these values.

In addition, the shortest decay time considered in almost all of these and other previous efforts along this line has been one or two days. The major uncertainty in attempting to proceed towards shorter cooling times arises from the lack of information relating to the decay schemes of many of the shorter-lived gammaray emitting fission product isotopes, a number of which it was thought could conceivably become significant in any shielding calculation. Therefore, a portion of the effort in this analysis was directed towards extending the fission product gamma-ray data to as short a cooling time as possible.

#### **B. THE EFFECTIVE ENERGY METHOD**

The new method developed here for determining the shielding requirements for spent fuel will permit the easy and rapid assessment of the dose rate from a given shield or the shielding required for a given spent fuel source. This method has as its basis an "effective energy" concept, wherein the entire fission product gammaray spectrum is replaced by a single gamma-ray line, which behaves exactly as the entire spectrum for the given parameters of irradiation and decay time, and shield thickness. The range of parameters evaluated includes irradiation times of 100 hours to 300 days, cooling times of 20 minutes to 300 days, and shields consisting of 6 to 12 inches of lead, 6 to 12 feet of water, 3 to 6 feet of ordinary concrete, and 2 to 4.25 feet of magnetite concrete. To determine the radiation level from a given shield using the effective energy method, only the following steps are necessary:

- 1) Obtain the gamma-ray energy intensity of the source for the particular irradiation and decay time under consideration directly from the curves in Fig. 1 and 2.
- 2) Obtain the effective energy of the source for the given shield thickness from the curves in Fig. 10 to 25.
- 3) Use this effective energy and perform the required shielding calculation to obtain the dose rate. The values of the linear absorption coefficient, dose conversion factor, and buildup factor used in this calculation should all correspond to the effective energy.

To determine the shielding requirements for a given source, it is necessary to iterate Steps 2 and 3, i.e., using different shield thicknesses and hence, different effective energies, until sufficient points have been obtained to determine the proper thickness from a curve of dose rate vs shield thickness.

The results are quite accurate, probably better than within a factor of two for the range of parameters evaluated.

#### II. BASIS OF METHOD

#### A. CHOICE OF FISSION PRODUCT ISOTOPES

In establishing the shielding requirements for spent U<sup>235</sup> fuel, the shield thickness for thick shields is determined by the "harder" of the gamma-ray emitting fission product isotopes. Softer radiations, even though they may be present in greater quantity, will be more easily shielded due to the rapid increase in the shield absorption coefficient with decreasing gamma-ray energy (and the even more rapid decrease in the resultant attenuation factor), and thus will make only a very small contribution to the dose rate at the shield surface. Furthermore, since in practice the shields required for spent fuel can be classified in the "thick" shield category, thin shields will not be considered in this analysis. As a result, it is intended that this method be applied only to the evaluation of thick shields and therefore, the fission product isotopes considered are, for the most part, those emitting gamma rays with energies equal to or greater than 1.60 Mev. This cutoff point in the fission product gamma-ray spectrum was selected because, since the 1.60-Mev La<sup>140</sup> gamma ray is frequently the controlling factor in spent fuel shield design, at least for moderate cooling periods, it was felt that it would make a convenient cutoff point. The result is that monolithic shields of less than 6 inches of lead, 6 feet of water, 3 feet of ordinary concrete, or 2 feet of heavy concrete cannot be analyzed with the accuracy claimed for the thicker shields.

The particular isotopes considered, their yields, and decay schemes (or applicable portion thereof) are listed in Table I. Most of these decay schemes have not been definitely established, so that comments on the contents of the table, and in particular on the data in columns 2 and 3, are included.

#### **B. RANGE OF IRRADIATION PARAMETERS CONSIDERED**

In determining the energy output of these hard gamma rays, calculations were performed for six different irradiation times: 100 hours, 14, 30, 60, 100, and 300 days. The range of decay times examined was from 20 minutes to 300 days. The analysis was divided into two portions, cooling periods of less than and greater than 35 days. The reason for these two separate regions is that beyond 35 days, both  $Zr^{95}$  and Nb<sup>95</sup> contribute significantly to the dose rate at the shield surface and, as a result, they were included in the source spectrum for this latter part of the analysis.

#### **III. CALCULATIONS**

## A. ENERGY OUTPUT OF HARD FISSION PRODUCT GAMMA-RAY EMITTERS

The calculational methods used to determine the energy output of the hard fission product gamma rays are relatively straightforward. The energy output  $P_i$ , in Mev/sec-watt, due to each of the gamma rays listed in Table 1, is determined using the expression:

$$\mathbf{P}_{i} = \mathbf{g}_{i}\mathbf{E}_{i}\mathbf{F}\left\{\frac{\mathbf{Y}_{i}\lambda_{i}\lambda_{i-1}}{\lambda_{i}-\lambda_{i-1}}\left[\left(\frac{1-e^{-\lambda_{i-1}\tau}}{\lambda_{i-1}}\right)e^{-\lambda_{i-1}\tau} - \left(\frac{1-e^{-\lambda_{i}\tau}}{\lambda_{i}}\right)e^{-\lambda_{i}\tau}\right]\right\} \dots (1)$$

where  $g_i$  = the number of gamma rays of energy  $E_i$  emitted by the *i*th isotope per disintegration,

 $Y_i$  = fission yield of the isotope chain under consideration,

F = fission rate per watt = 3.1 x 10<sup>10</sup> fissions/sec-watt,

 $\lambda_i$  = decay constant of the isotope under consideration, in sec<sup>-1</sup>,

 $\lambda_{i-1}$  = decay constant of the parent isotope, in sec<sup>-1</sup>,

 $\tau$  = irradiation time, in sec,

and t = decay time, in sec.

The total amount of energy available from all the gamma-ray lines specified in Table I, for a given value of  $\tau$  and t, is then simply the sum of all the individual contributions. This sum has been plotted in Fig. 1 and 2 for the irradiation and decay times considered.

#### **B. THE EFFECTIVE ENERGY**

Having determined the individual intensities of each of the gamma-ray lines under consideration, it is then possible to evaluate the effective energy. As applied to this method, this energy is the gamma-ray line which, when confronted by a given shield thickness, will suffer the same attenuation as the initial source spectrum, and hence yield the same dose rate outside the shield for a given set of parameters. As mentioned earlier, these parameters are the irradiation and decay time, shield material, and thickness.

To determine this effective energy, each of the gamma-ray lines comprising the source spectrum (as listed in Table I) is attenuated through a given thickness of shielding material. The shielded dose rate,  $D_i(s)$  in r/hr-watt, due to  $P_i$ , when a thickness of shielding material is interposed, is obtained from

$$D_{i}(s) = D_{i}(u) \left[ (B_{r})_{i} e^{-\mu_{i}x} \right] \qquad \dots (2)$$

where  $(B_r)_i$  = the dose buildup factor,

#### TABLE I

#### DATA ON HARD GAMMA-RAY EMITTING FISSION PRODUCTS

Isotope	Gamma-Ray Energy (Mev)	Gamma Rays Emitted per Disintegration (%)	Portion of Decay Chain Determining Time Dependence After Shutdown	Fission Yield <sup>2</sup> (%)	Cor
Br <sup>84</sup>	1.89	35	$Se^{84}(2m) \rightarrow Br^{84}(32m)$	0.65	The decay scheme was infer Hollander, et al., <sup>4</sup> and is t
Kr <sup>87</sup>	1.89 2.3	12.5 12.5	Br <sup>87</sup> (56s) - Kr <sup>87</sup> (78m)	2.0	The gamma-rav branching r on the decay scheme of Holl
Kr <sup>88</sup>	1.8 2.18	12 68	$Br^{88}(16s) \rightarrow Kr^{88}(2.77h)$	3.1	The intensity of the gamma- data in Hollander, et al. <sup>4</sup>
Rb <sup>88</sup>	1.86	20 2.0	$Kr^{88}(2.77h) \rightarrow Rb^{88}(17.8m)$	3.1	The absolute intensities of the from relative intensity mea
Rh <sup>106</sup>	2.4	0.25	$\operatorname{Ru}^{106}(1.0y) \longrightarrow \operatorname{Rh}^{106}(30s)$	0.52	The intensity of the 2.4-Mer Hollander, et al. <sup>4</sup>
Sn <sup>125</sup>	1.9	5.0	$\operatorname{Sn}^{125}(9.4d) \rightarrow$	0.028	The intensity of the 1.9-Me Hollander, et al. <sup>4</sup>
1 <sup>132</sup>	2.0	2.7	$Te^{132}(77.7h) \rightarrow I^{132}(2.4h)$	4.4	The intensity of the 2.0-Me Ergen. <sup>5</sup>
1 <sup>134</sup>	2.2	1.0	$Te^{1.34}(44m) \rightarrow I^{1.34}(52.5m)$	5.7	The intensity of the 2.2-Me to be the same as used by C
1 <sup>135</sup>	1.8 2.4	73 2.0	$Te^{135}(2m) \rightarrow I^{135}(6.68h)$	5.9	The decay scheme was infe The intensity of the 2.4-Me
La <sup>140</sup>	1.6 2.5 2.9	95 4.9 0.10	$Ba^{140}(12.8d) \rightarrow La^{140}(40h)$	6.17	The absolute intensities of from data in Hollander, et was taken directly from the
Pr <sup>144</sup>	2.18	0.73	$Ce^{144}(275d) \rightarrow Pr^{144}(17.5m)$	4.64	The intensity of the 2.18-M 0.86-Mev beta ray, as show cent of the decays.
Eu <sup>156</sup>	2.0	60	$Sm^{156}(10h) \rightarrow Eu^{156}(15.4d)$	0.013	The intensity of the 2.0-Me et al, $4$ and is the same as the same state of the same as t
Zr <sup>95</sup>	0.721	99	$Y^{95}(10.5m) \rightarrow Zr^{95}(65d)$	6.0	The decay scheme was take
Nb 95	0.745	100	$Zr^{95}(65d) \rightarrow Nb^{95}(35d)$	6.0	The decay scheme was take

#### omments

erred from the compilation of the same as used by Clark.<sup>2</sup>

ratio was assumed and was based ollander, et al.<sup>4</sup>

a-ray lines was inferred from the

the gamma-ray lines were taken asurements noted in Hollander, et al.<sup>4</sup>

ev line was inferred from data in

ev line was inferred from data in

ev line was taken from the work of

ev line is uncertain, but was assumed Clark.<sup>2</sup>

erred from data in Hollander, et al.<sup>4</sup> fev line agrees with that used by Clark.<sup>2</sup>

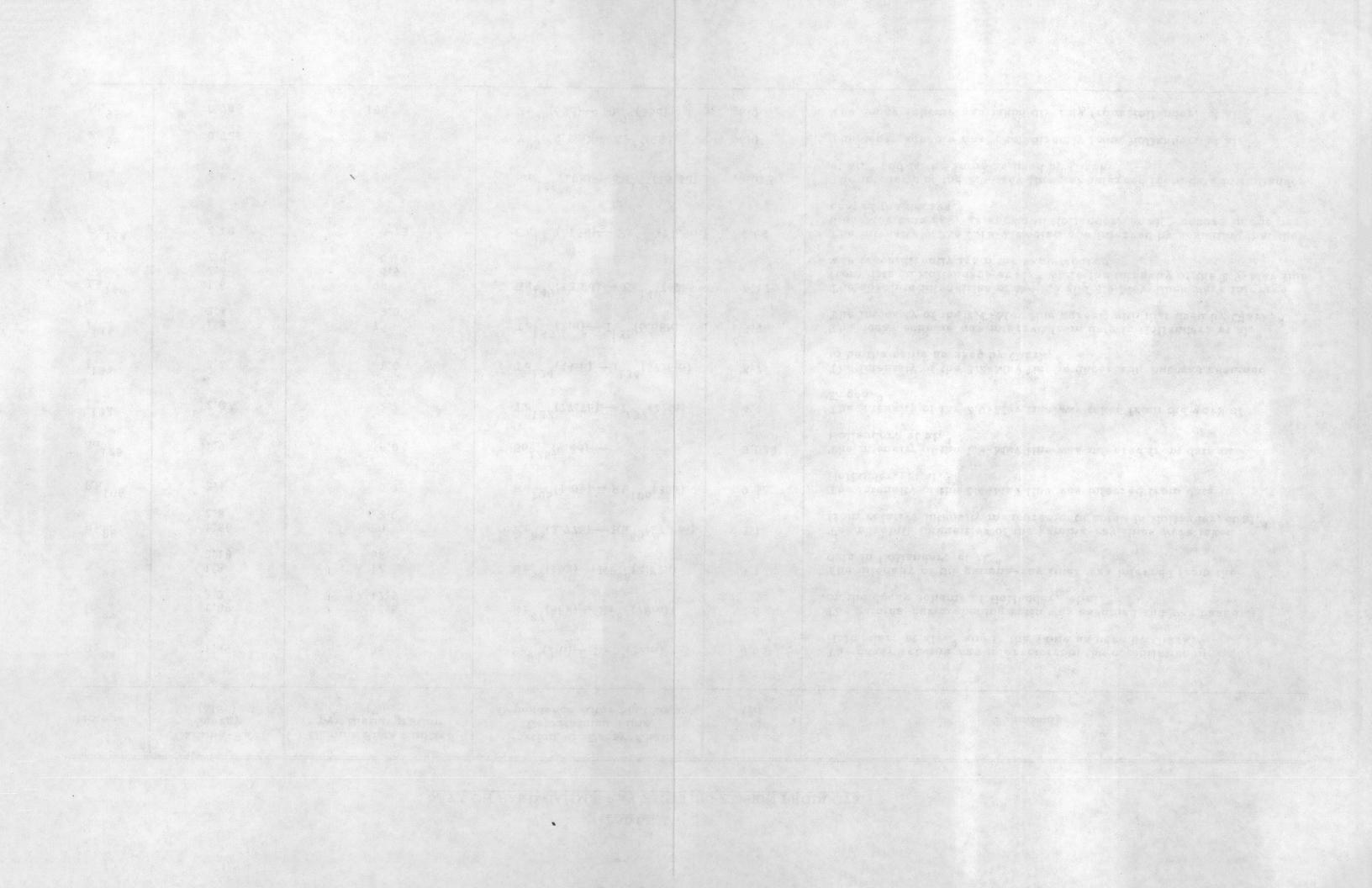
f the 1.6 and 2.5 Mev-lines were inferred t al, <sup>4</sup> while the intensity of the 2.9-Mev line he same source.

Mev line was inferred by assuming that the own in Hollander, et al,<sup>4</sup> occurs in one per

ev line was inferred from data in Hollander, used by Clark.<sup>2</sup>

ken directly from Hollander, et al.<sup>4</sup>

ken directly from Hollander, et al.<sup>4</sup>



- $\mu_i$  = the linear absorption coefficient of the shield material for the gamma ray under consideration, in cm<sup>-1</sup>,
  - x = shield thickness, in cm,

and 
$$D_i(u) = unshielded$$
 dose rate due to  $P_i$ , in r/hr-watt.

The unshielded dose rate is evaluated by using a suitable conversion factor (see Fig. 3) to convert energy flux to dose rate.<sup>6</sup> The sum of  $D_i(s)$  is then the total shielded dose rate after penetration through x centimeters of a given shield.

The ratio,  $G(\tau, t, x)$ , of the total dose rate after attenuation to the total unshielded value, corresponds to the actual attenuation offered by the shield thickness x to the spectrum under consideration for a specific value of irradiation  $\tau$  and decay t. In this analysis the ratio  $G(\tau, t, x)$ , or what is usually termed the attenuation factor, is given by

$$G(\tau, t, x) = \frac{\sum_{i}^{j} D_{i}(s)}{\sum_{i}^{j} D_{i}(u)} \qquad \dots (3)$$

From a curve of attenuation, i.e.,  $B_r e^{-\mu t}$ , <u>vs</u> photon energy for the same shield thickness, it is found that this attenuation factor corresponds exactly to that for a specific gamma-ray line. This line is then termed the effective energy of the source spectrum, for a particular irradiation time, decay time, and shield thickness.

#### C. LINEAR ABSORPTION COEFFICIENTS AND DOSE BUILDUP FACTORS

The shield materials considered in the calculations in Section III B above, include lead (6 to 12 inches), water (6 to 12 feet), ordinary concrete (3 to 6 feet) and magnetite concrete (2 to 4.25 feet). The composition and linear absorption coefficients for ordinary concrete were taken from White, <sup>7</sup> whereas the composition of magnetite concrete was taken from Henrie. <sup>8</sup> The density of the ordinary concrete was taken as 2.32 gm/cm<sup>3</sup>, whereas for magnetite concrete a value of 3.7 gm/cm<sup>3</sup> was used. The linear absorption coefficients for the magnetite concrete were evaluated using standard techniques. The resultant coefficients for both concretes are shown in Fig. 4. The dose buildup factors for these concretes were evaluated using the "equivalent Z" method of Goldstein and Wilkins;<sup>9</sup> the results are shown in Fig. 6 and 7. The gamma-ray linear absorption coefficients and dose buildup factors used for lead (density taken to be 11.3 gm/cm<sup>3</sup>) and water were taken from White<sup>7</sup> and Goldstein and Wilkins,<sup>9</sup> and are plotted in Fig. 5, 8, and 9.

It should be noted that the linear absorption coefficients listed in White<sup>7</sup> include coherent scattering, whereas the values as plotted have omitted this term. This is as it should be for penetration calculation, since coherent scattering will not result in any change in energy and only a very slight change in direction. Furthermore, for this same reason, coherent scattering was omitted from the linear absorption coefficients used in the theoretical determination of the dose buildup factors for these materials.<sup>9</sup>

### **IV. RESULTS**

The effective energy was computed as indicated in Section III B above, and is plotted in Fig. 10 to 25 for the different thicknesses of the four shielding materials considered. As mentioned in Section II, each curve is divided into two regions, one covering the period from 20 minutes to 35 days after shutdown and the other, from 35 to 300 days.

A comparison of the results obtained by using data from the two curves at their common decay point (in conjunction with the appropriate value from either Fig. 1 or Fig. 2) indicates that the difference in the dose rates obtained is less than 15 per cent for the two greatest thicknesses of each material, less than 20 per cent for the next lowest thickness, and less than 25 per cent for the smallest shield thickness considered. The latter is true except for the 300-day irradiation curve for a 6-foot shield, in which case the difference is 34 per cent. In all cases, the results obtained using values from the second region were higher, as would be expected.

It is interesting to note that the effective energy for all irradiation times and all shield materials considered is relatively constant for cooling times from about 2 to 35 days. In fact, in this range of cooling times, a rough rule of thumb for the effective energy for any of the thicknesses of shield materials considered and within the range of irradiations analyzed, is  $1.74 \pm 0.10$  Mev.

## **V. APPLICATIONS**

The results of this analysis can be used in solving two types of spent fuel shielding problems. In either case the operational history of the spent fuel, i.e., the irradiation and cooling times and the fission rate or operating power level, must be known.

#### A. TYPE 1: DOSE RATE FROM FIXED SHIELD

If it is desired to determine the radiation level from a given shield thickness, the following procedure should be used.

- Step 1. Use Fig. 1 or 2, depending on the specific cooling time involved, to determine the gamma-ray source strength to be used in the subsequent calculations. Note that it is implicit in the use of these two curves that the power level or fission rate be constant over the operation time. If this was not the case, or if the irradiation was of either a cyclic or irregular nature, each different constant power interval in the operational history should be evaluated separately.
- Step 2. From the appropriate curve of shield material and thickness, Fig. 10 to 25, find the effective energy corresponding to the specific irradiation and decay times being considered.
- Step 3. Use this effective energy to calculate the dose rate at the surface of the shield being evaluated, or at any more distant point which may be of interest. In performing the calculation, the values of  $\mu_i$ ,  $B_r$ , and  $k_i$  should correspond to that for the effective energy obtained in <sup>i</sup>Step 2 (see Fig. 3 to 9).

When dealing with solid sources, where self-absorption is a consideration, appropriate corrections to the effective energy as obtained from Step 2, should be made.

Note: Such corrections can be made by using the effective energy obtained from Step 2 to estimate the self-absorption thickness (for example, see Foderaro and Obenshain<sup>10</sup>). Then, using either density ratios or, more accurately, linear absorption coefficient ratios, find an equivalent thickness of the shield material which would correspond to this source self-absorption thickness. The effective energy which should be used in the dose rate calculation is, then, that corresponding to a shield consisting of the sum of the two thicknesses of the <u>same</u> material. The exact value of the effective energy can be obtained by interpolation of the data on the curves of Fig. 10 to 25. It should be borne in mind, however, that to preserve the accuracy claimed, this method is applicable to thick sources only as long as the actual shield dimension is at least equal to the smallest shield thickness for which effective energy data have been plotted.

#### **B. TYPE 2: DETERMINATION OF SHIELD THICKNESS**

To determine the shield requirements for a given spent fuel source, perform Step 1, estimate a shield thickness, and perform Steps 2 and 3, making any appropriate self-absorption corrections which may be required. The result will be the dose rate at the surface of, or at any more distant position from, the estimated shield. Then, depending on whether the dose rate is high or low, choose either a thicker or thinner shield, and iterate Steps 2 and 3. Continue this procedure until sufficient points have been established to permit the determination of the required shield from a curve of dose rate vs shield thickness.

Several examples illustrating the application of these methods are included in the Appendix.

#### VI. LIMITATIONS OF THE METHOD

One limitation of the effective energy method is due to the cutoff point in the fission product gamma-ray spectrum used in establishing the source spectrum for this analysis. Since the contribution from all lower energy fission product gamma-ray emitters has been omitted, the dose rate through "thin" shields cannot be determined. "Thin", in this case, refers to thicknesses less than the smallest shield for which effective energies have been evaluated. Actually, the effective energy data can probably be extrapolated and used for preliminary

evaluation of shields up to one thickness increment smaller than that for which calculations have been performed (i.e., 4 inches of lead, 4 feet of water, 2 feet of ordinary concrete, or 1.25 feet of magnetite concrete), and the accuracy of the results will be well within an order of magnitude. However, when shielding spent fuel or gross fission products, such thin shields are not frequently encountered.

Another limitation is that the accuracy of the results is dependent upon the specific decay schemes chosen. Actually, as noted in Section II, most of the decay schemes listed in Table I were either assumed or inferred, usually using data in Hollander, et al,  $^4$  as a basis. In the case of  $I^{135}$ , for example, recent experimental information<sup>11</sup> indicates a decay scheme different from, and much more complex than, that used in this analysis. The adequacy of the decay schemes used, and hence the extent of this limitation, can only be determined by more detailed experimental spectral measurements, or by comparison of data from extensive experimental spent fuel shielding parameter studies with results obtained using the effective energy method.

Since the fission yields used in Table I are those for  $U^{235}$ , the effective energy method is not strictly applicable to the determination of the shielding requirements for  $U^{233}$  and  $Pu^{239}$  fission products. However, since the change in the mass-yield curve is fairly small, the use of this method should yield results which are adequate for the purpose of preliminary analyses. It should be noted however, that if the yield of a given isotope changes by a factor of two, the resultant dose rate from this isotope will change accordingly, whereas the dose rate from the source will be affected to a lesser degree.

The effective energy method is only applicable to monolithic shields, due to the still-unknown character of the gamma-ray dose buildup factor in thick shields containing regions of two or more distinctly different shielding media, thus presenting another limitation. Some work on the behavior of the buildup factor in thin two-media shields has recently been reported, <sup>13, 14</sup> but it has not been ascertained whether the formulation holds for deeper penetrations. When the shield consists of two different layers, the atomic numbers of which differ appreciably, and the attenuation in each layer is comparable, or nearly so, the deep penetration problem will have to be handled in a more specialized manner, one beyond the scope of this report. However, in a deep penetration problem where most of the attenuation is in one of the shield materials, the buildup factor may be adequately represented by assuming all the penetration in that material. Furthermore, when the atomic numbers are reasonably close, as in the case of aluminum and ordinary concrete or iron and magnetite concrete, the buildup factor can be represented by assuming all of the penetration in either of the materials. The assumption can even be extended to an iron-water shield. Many other recipes have been contrived to increase the accuracy of evaluating this factor, but again, a discussion of this aspect of shielding is beyond the scope of this report.

#### VII. COMPARISON WITH EXPERIMENT

The accuracy of this method for determining the shielding requirements for spent fuel depends on comparison with accurate experimental data. Only one such piece of experimental information<sup>15</sup> was available to the author at this writing. This consisted of a series of measurements performed in the water surrounding the Oak Ridge Bulk Shielding Reactor following a 100-hour run at a power level of 100 kilowatts. The fuel elements used in the reactor were fresh, i.e., none had operated at any significant power level previous to the experiment. A comparison of the experimental data with the results obtained using this method is included in Table II below.

#### TABLE II

		Dose	Dose Rate	
Decay Time (min)	Water Shield Thickness (ft)	Calculated (mrem/hr)	Measured (mrem/hr)	
20	8	35.2	80.3	
30	8	33.2	64	
45	8	31.6	53	
60	8	30.4	43.5	
1 20	8	25.9	30.5	
240	8	15.1	20.5	
1 20	6	471	540	

#### AFTER SHUTDOWN DOSE RATES FROM THE BSR

Comparing the calculated and measured dose rates in Table II reveals that the isotopes used in Table I are not sufficient to adequately represent the fission product gamma-ray spectrum at short cooling times, with the result that the dose rate at times less than 45 minutes after shutdown is underestimated by a greater amount than at longer cooling times. This is not surprising however, as many attempts have been made to calculate the gross fission product gamma-ray energy intensity following a burst by summing up the contributions from the individual known fission product gamma-ray emitters. All such calculations have resulted in a decided departure from the well-known Way-Wigner expression, 1.26t<sup>-1.2</sup> Mev/fission-sec. At short cooling times, the best of these calculations have always yielded results lower than that predicted by the Way-Wigner expression, the departure occurring at about 60 minutes. At 20 minutes after the burst, the calculated data are low by at least a factor of two and at 30 minutes, by a factor of 1.5, indicating that some important short-lived fission product activities have been omitted. If then, only a fraction of the unaccounted-for energy is "hard," as defined in this analysis, the results obtained using the effective energy method for short cooling times (less than 45 minutes) might well be brought into much better agreement with the measured data.

For decay times greater than 45 minutes, the ratio of the calculated to measured values is fairly consistent, further supporting the contention that there are probably some short-lived hard gamma-ray emitting fission products which have not been identified, or whose decay schemes have not been resolved. The relative constancy of this ratio also indicates that the source spectra chosen in this analysis are adequate for evaluation of spent fuel shielding requirements when the decay is at least 45 minutes.

One other piece of experimental data<sup>16</sup> was available, the result of a series of measurements performed in the Materials Testing Reactor (MTR) Canal on a spent MTR fuel element. However, the source-to-detector separation distances quoted were reported to have been only approximate, since both the source and detector were manually supported. In this case the fuel element had been irradiated for 8.9 days at a constant power level, and the measurements were performed between 5 and 6 hours after shutdown. Dose rate measurements were made at 6-, 8-, 10-, and 12-foot source-detector separation distances. The calculated results were, as in the case of the BSR calculations, consistently lower than the measured values. The ratio of the calculated to measured values was between 0.5 and 0.8 which is in agreement with the ratios obtained in the BSR comparison, indicating that the spectrum chosen is quite adequate, at least in the range of irradiation and decay times for which comparisons with experimental data have been made.

#### VIII. DISCUSSION OF THE POSSIBLE ERRORS

Although no attempt has been made to analyze in detail the possible sources of error which could effect the results presented here, or those calculated using this data, it was thought advisable to at least summarize their origin and, when possible, estimate the associated error.

The linear absorption coefficients and buildup factors used in Sections III and VII, as well as those that would be used in any shield evaluation, are all subject to some error. The absorption coefficients are, in many cases, known only to an accuracy of about one per cent, and the buildup factors to about five per cent.<sup>9</sup> However, a one per cent error in the absorption coefficient would result in an error of about 10 to 15 per cent in the dose rate, for the shield thicknesses evaluated in the BSR comparison calculations. For a 12-foot water shield, the error could be as high as 30 per cent, depending on the decay time, i.e., the effective energy, and hence  $\mu$ . Furthermore, the buildup factors which were utilized were taken from tabulations of point isotropic infinite media data, <sup>9</sup> whereas in most instances the shields encountered in design problems are finite. The use of infinite media buildup factors in finite shield problems will introduce some additional error, yielding results which are high. The magnitude of the difference has only recently been investigated.<sup>12</sup> This error will, however, not be very significant, probably no more than about 15 per cent for the 6-foot water shield data in Fig. 10. For greater thicknesses and for higher atomic number shield materials, the error will be less.

Another source of error can be attributed to the very method by which the values of  $E_{\rm eff}$  were determined, i.e., the use of point source geometry, which is different from the geometry in most calculations. This, it is expected, will lead to a very small and perhaps negligible error, but is mentioned here for the sake of completeness.

In determining  $P_i$  (Section III), the entire fission yield was assigned to the parent activities shown in column 4 of Table I. The presence of any partial yields would tend to increase the dose rates calculated for short cooling periods and decrease those for long decay times. Also, no consideration of fission product burnout was made in any of the calculations performed to determine  $P_i$ . Using the method presented here, determination of the shielding requirements for spent fuel which has been irradiated to a high  $U^{235}$  burnup, would be expected to yield results which overestimate the actual dose rate. The effect of neglecting burnout will result, in most cases, in a very small and probably negligible error.

With regard to the calculations performed in Section VII, several other sources of error should be mentioned. One source of error is the calculational methods used in evaluating the dose rate from the BSR. In these calculations, the method and curves of Foderaro and Obenshain<sup>10</sup> were used and can lead to an error of about 10 per cent. The method of determining the magnitude of the buildup factor (see Eq. 8A in the Appendix) may also lead to some additional error.

In spite of all these potential sources of error, it is interesting to note that when the results obtained using the effective energy method were compared with experimental data, the average ratio of calculated to measured values was about 0.80, for cooling times greater than 45 minutes. Even taking into consideration the magnitude of potential errors discussed above, dose rates calculated for times beyond those for which experimental data is available should be accurate within a factor of two.

#### IX. CONCLUSION

The effective energy method for determining the shielding requirements for spent  $U^{235}$  fuel will greatly simplify and reduce the amount of labor involved in the solution of such problems. From the results obtained by comparing calculated dose rates with experimental data, it is evident that this method is extremely accurate, at least over the range of parameters for which experimental data is available. The method also has the advantage that it is based on a very simple concept, i.e., effective energy.

These same comparison calculations also indicate that, whereas the ratio of calculated to measured values was  $0.8 \pm 10$  per cent for cooling times beyond 45 minutes, this ratio decreased sharply with shorter cooling times. This variance appears to indicate that there are several short-lived hard gamma-ray emitting fission products which have not been identified or whose decay schemes have not been resolved. For longer irradiation times and greater cooling times than those covered by the BSR and MTR comparisons, the effective energy method will yield results which are accurate within a factor of two. However, the accuracy of the results obtained by using this method for cooling periods less than 45 minutes will increase with increasing irradiation time (the BSR had only operated for 100 hours), because the buildup of the longer-lived hard gamma-ray emitting fission product isotopes will tend to mask the discrepancy which shows up so strongly in a shorter irradiation.

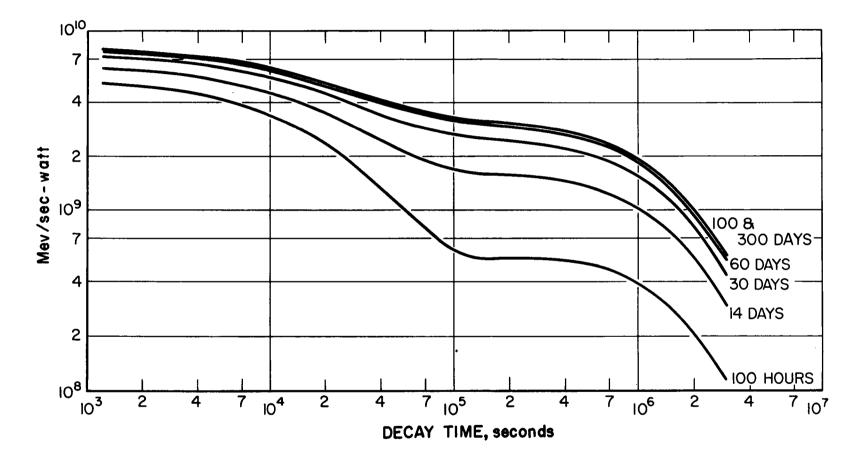


Fig. 1. Decay of Hard Gamma-Ray After-Shutdown Power for Various Irradiation Times (20 minutes to 35 days decay)

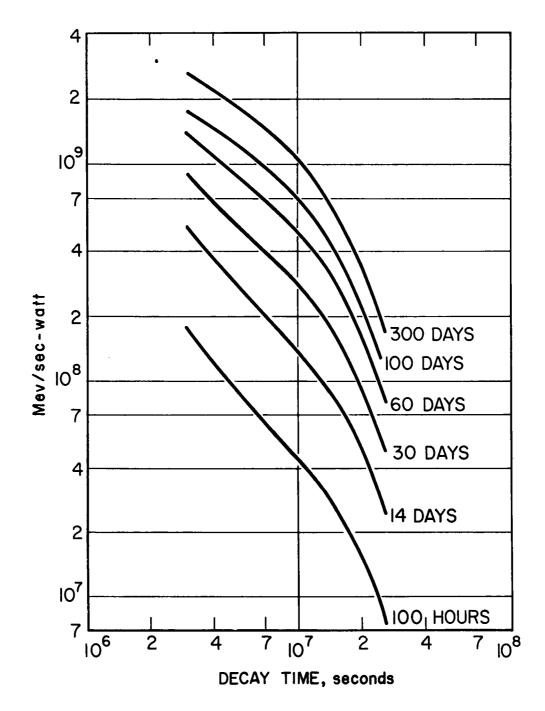


Fig. 2. Decay of Hard Gamma-Ray After-Shutdown Power for Various Irradiation Times (35 days to 300 days decay)

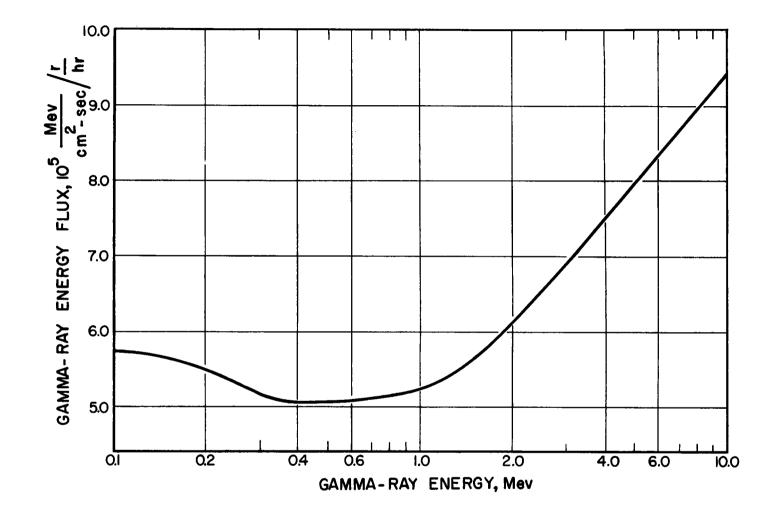


Fig. 3. Gamma-Ray Energy Flux Per Roentgen

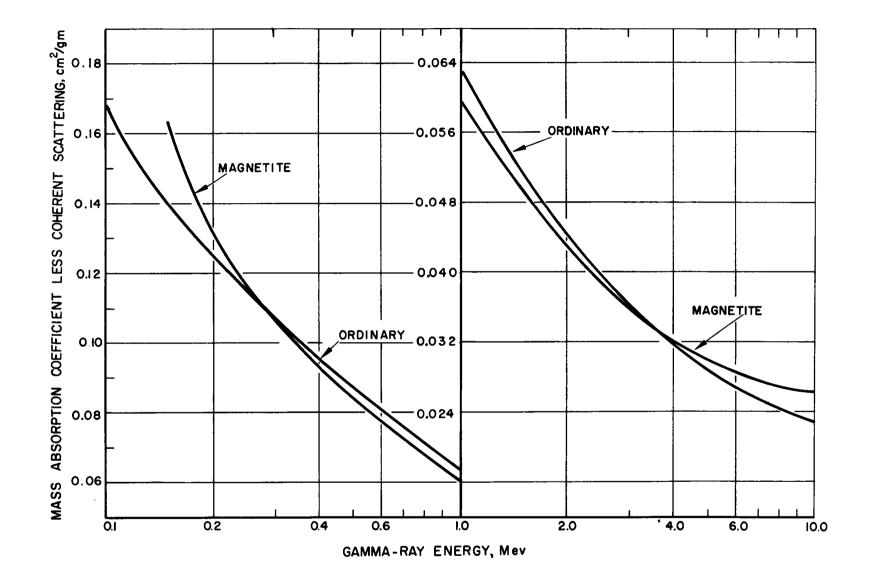


Fig. 4. Mass Absorption Coefficient Less Coherent Scattering for Ordinary and Magnetite Concretes

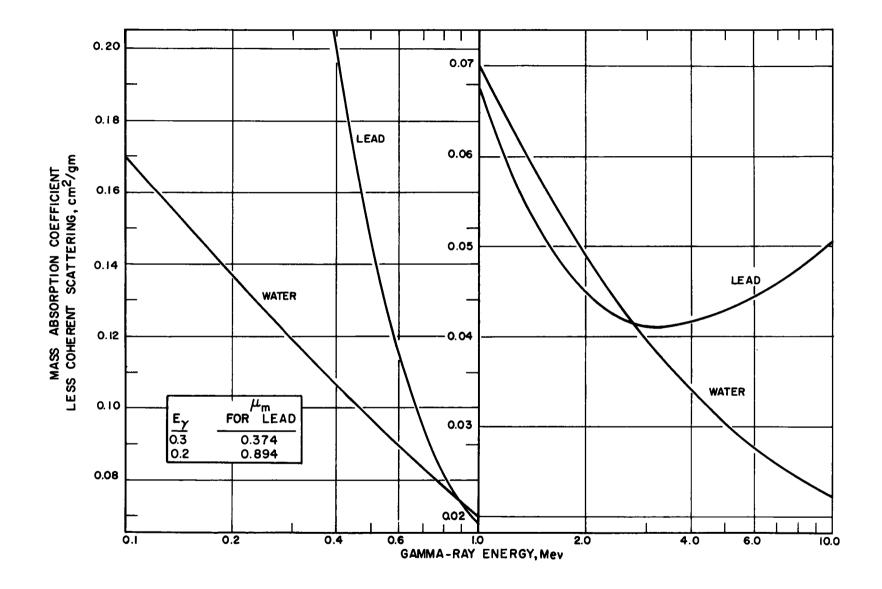


Fig. 5. Mass Absorption Coefficient Less Coherent Scattering for Lead and Water

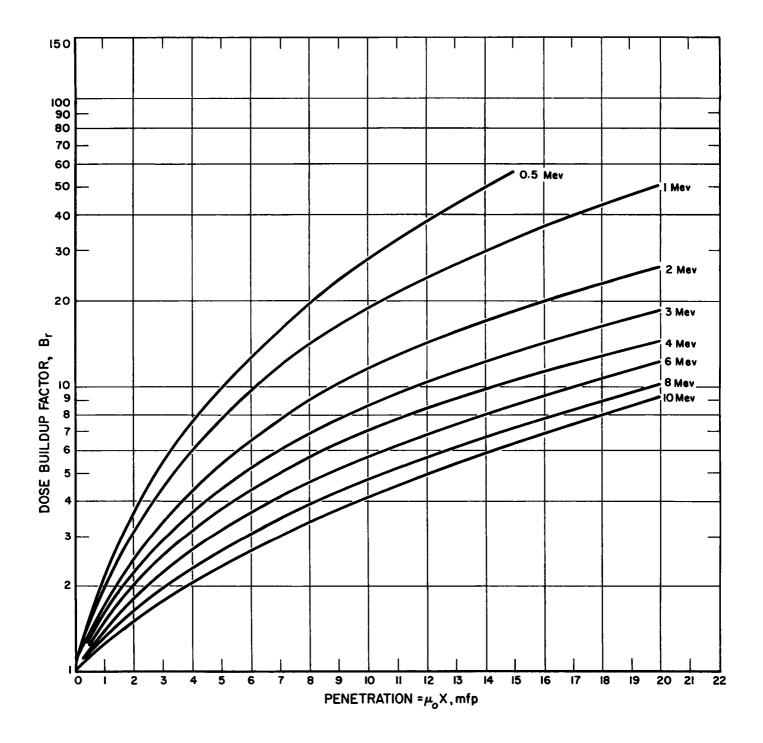


Fig. 6. Dose Buildup Factor for Magnetite Concrete (Point Isotropic Source)

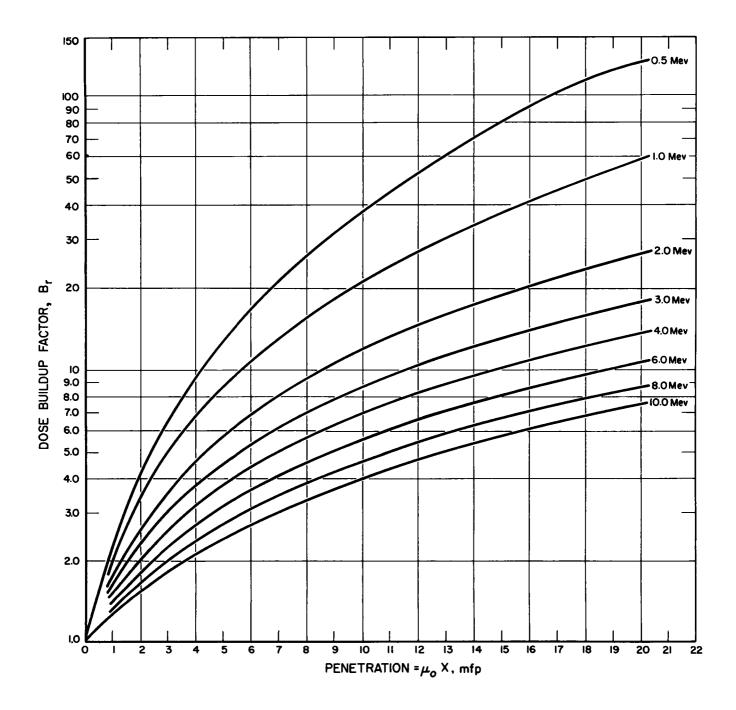


Fig. 7. Dose Buildup Factor for Ordinary Concrete (Point Isotropic Source)

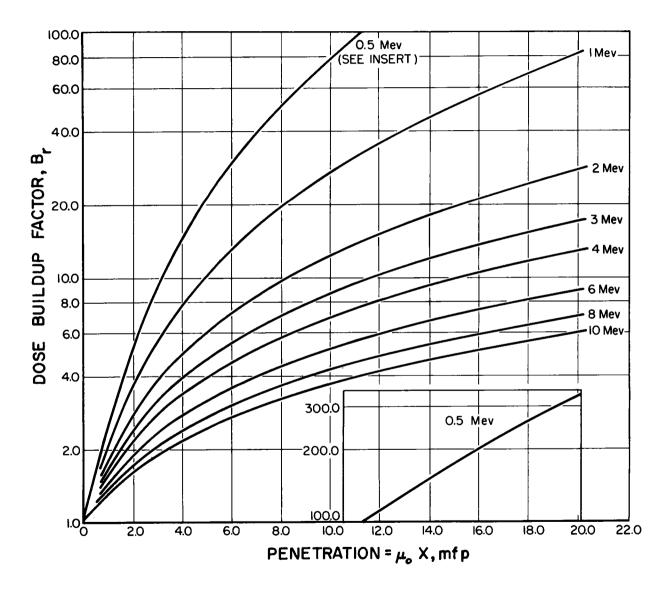


Fig. 8. Dose Buildup Factor for Water (Point lsotropic Source)

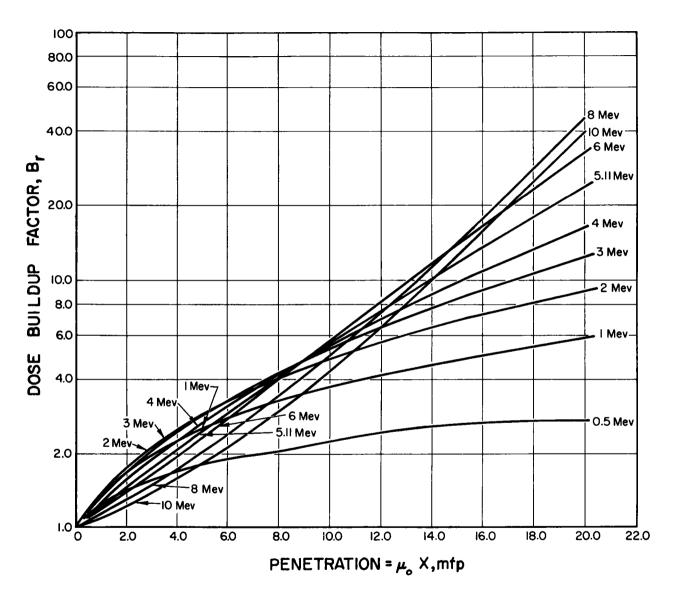


Fig. 9. Dose Buildup Factor for Lead (Point Isotropic Source)

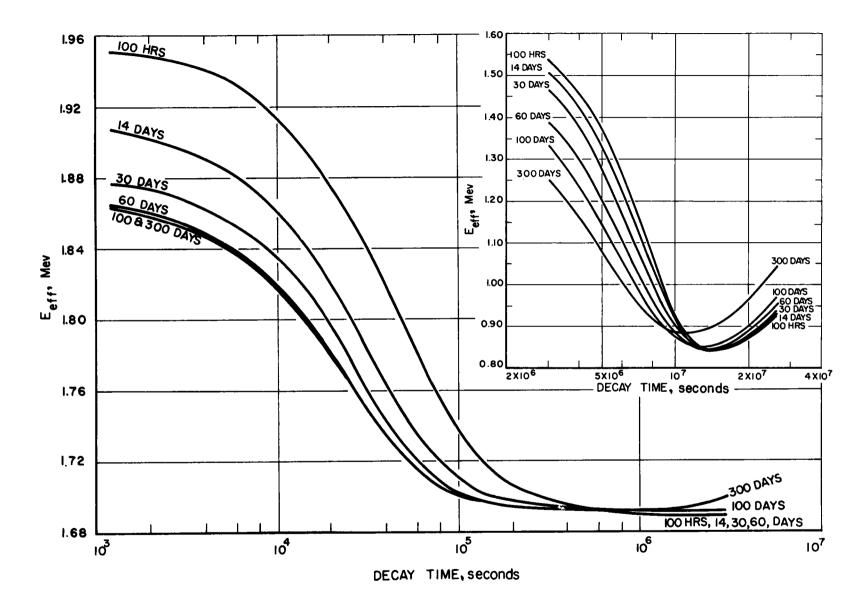


Fig. 10. Effective Energy as a Function of Decay and Irradiation Times for a 6-foot Water Shield

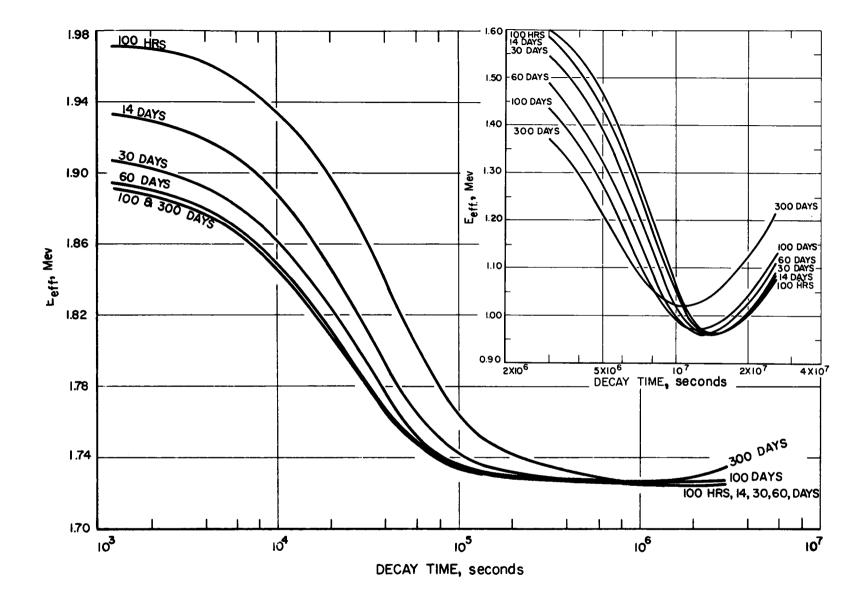


Fig. 11. Effective Energy as a Function of Decay and Irradiation Times for an 8-foot Water Shield

ω ω

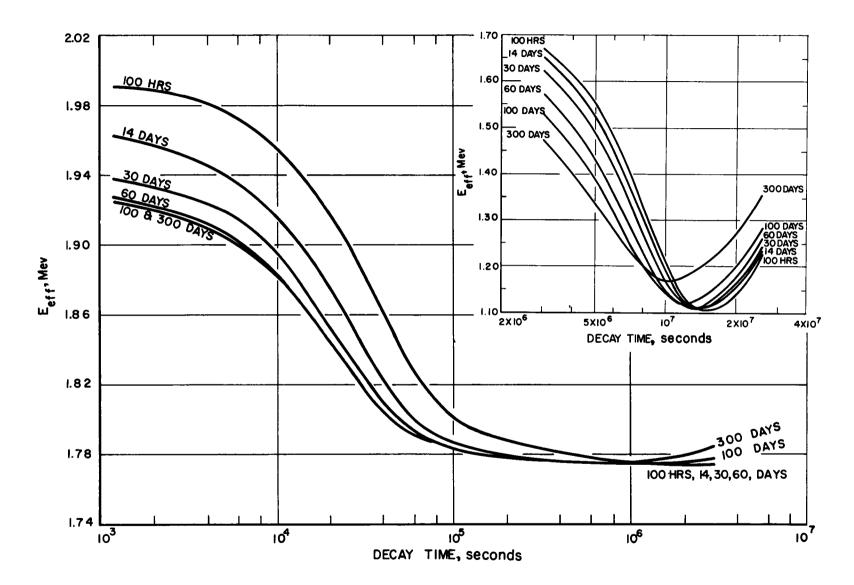


Fig. 12. Effective Energy as a Function of Decay and Irradiation Times for a 10-foot Water Shield

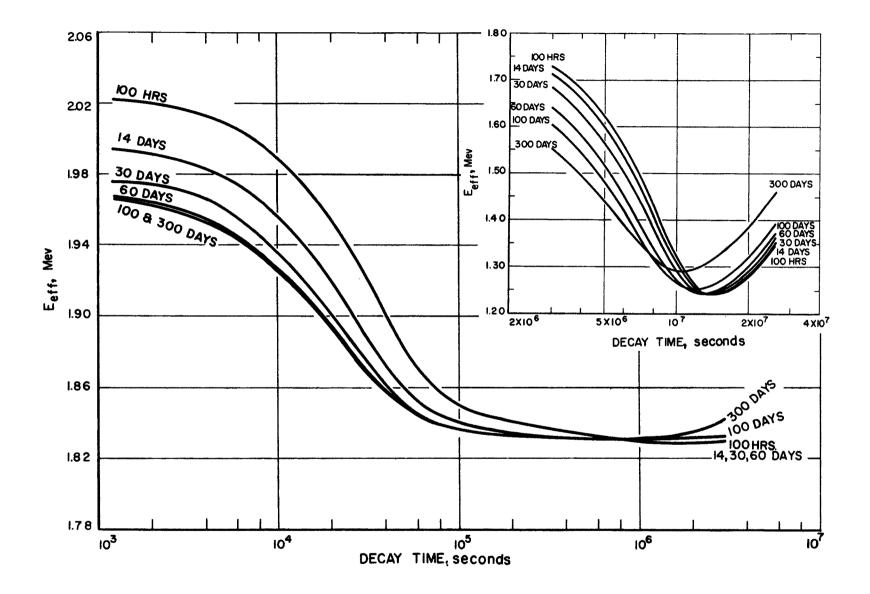


Fig. 13. Effective Energy as a Function of Decay and Irradiation Times for a 12-foot Water Shield

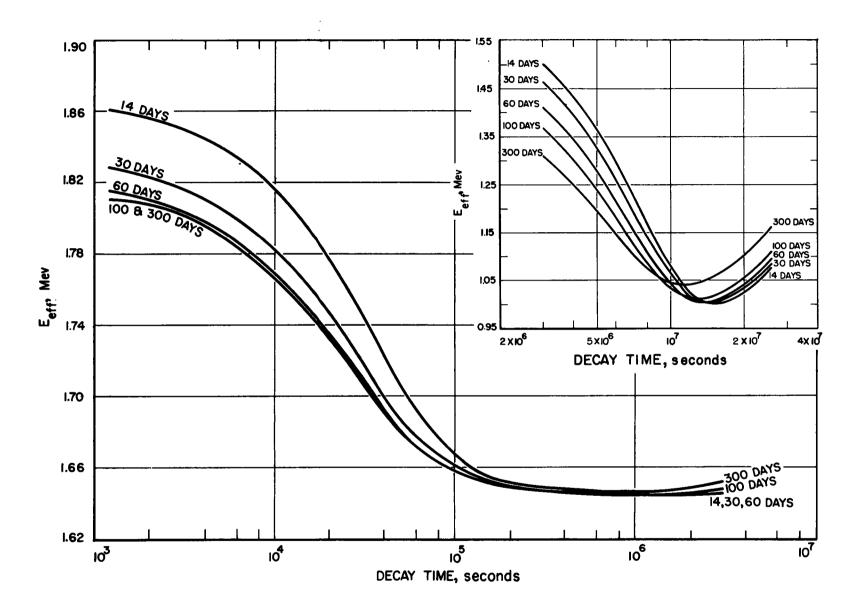


Fig. 14. Effective Energy as a Function of Decay and Irradiation Times for a 6-inch Lead Shield

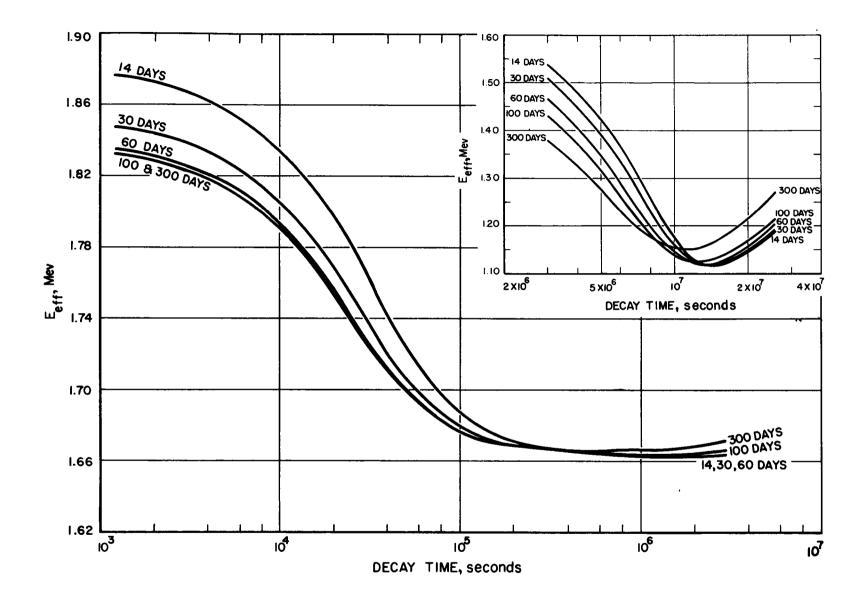


Fig. 15. Effective Energy as a Function of Decay and Irradiation Times for an 8-inch Lead Shield

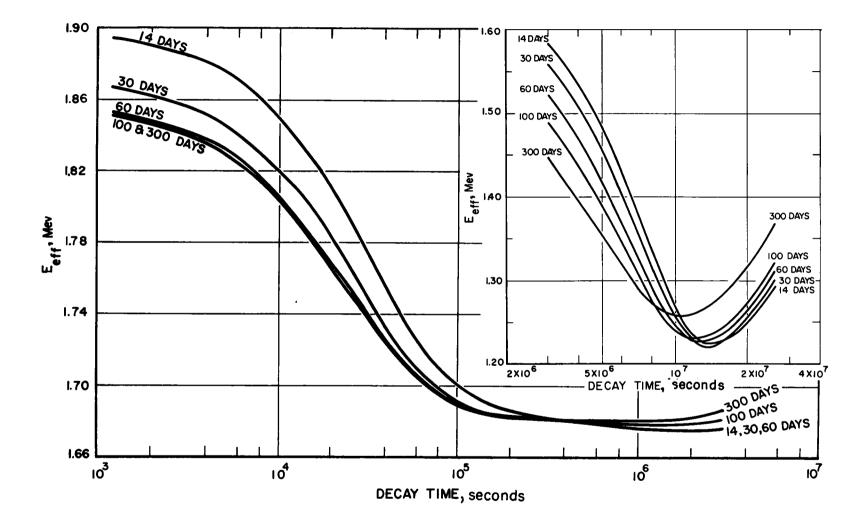


Fig. 16. Effective Energy as a Function of Decay and Irradiation Times for a 10-inch Lead Shield

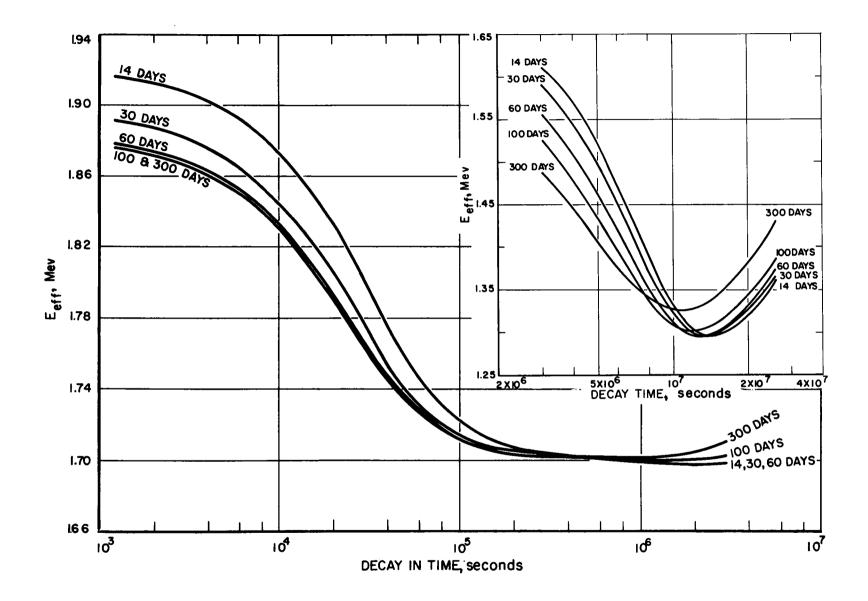


Fig. 17. Effective Energy as a Function of Decay and Irradiation Times for a 12-inch Lead Shield

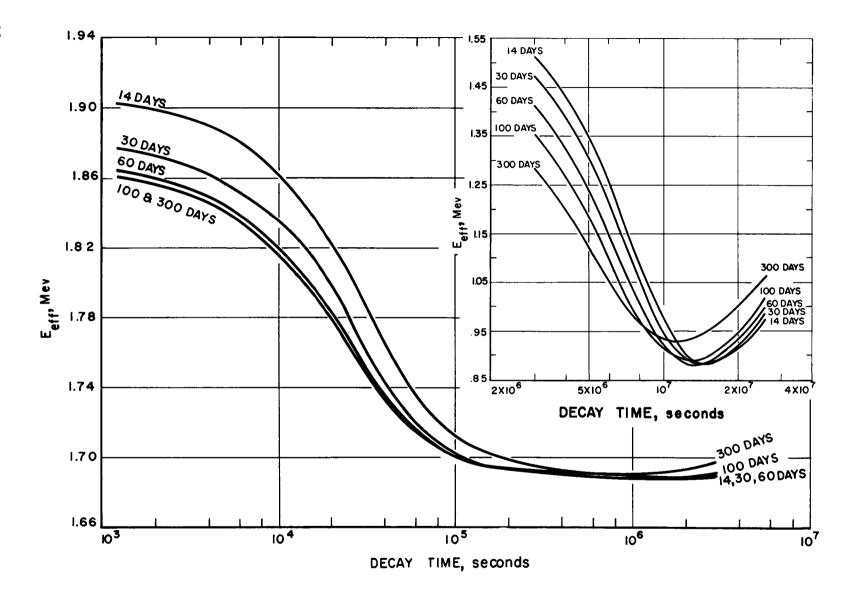


Fig. 18. Effective Energy as a Function of Decay and Irradiation Times for a 3-foot Ordinary Concrete Shield

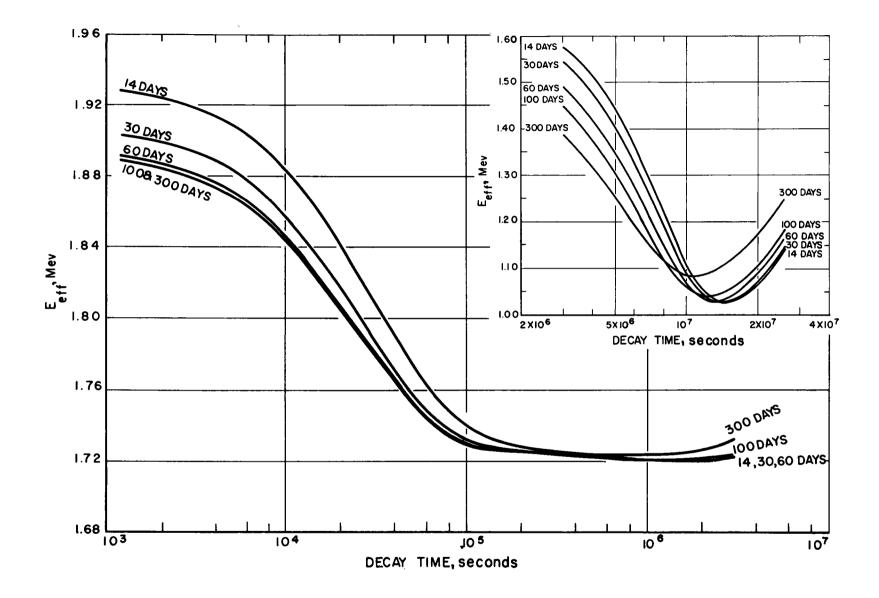


Fig. 19. Effective Energy as a Function of Decay and Irradiation Times for a 4-foot Ordinary Concrete Shield

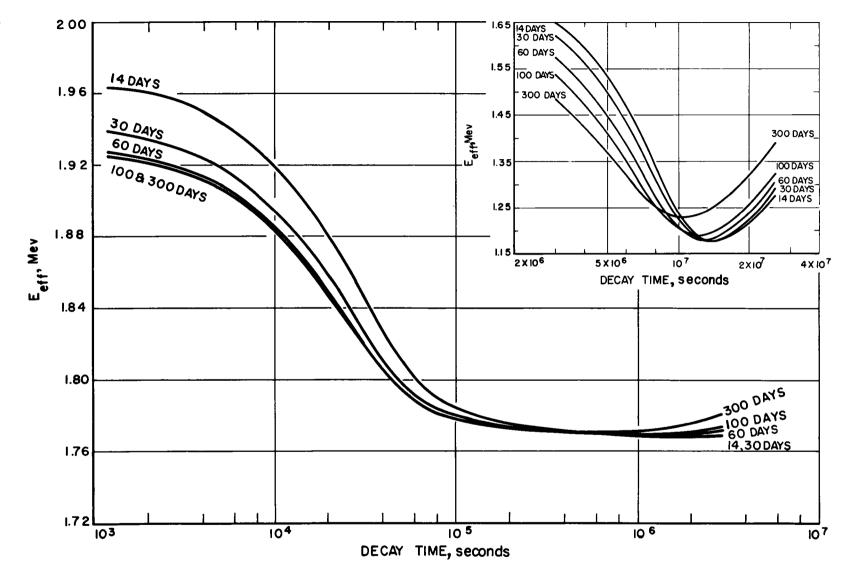


Fig. 20. Effective Energy as a Function of Decay and Irradiation Times for a 5-foot Ordinary Concrete Shield

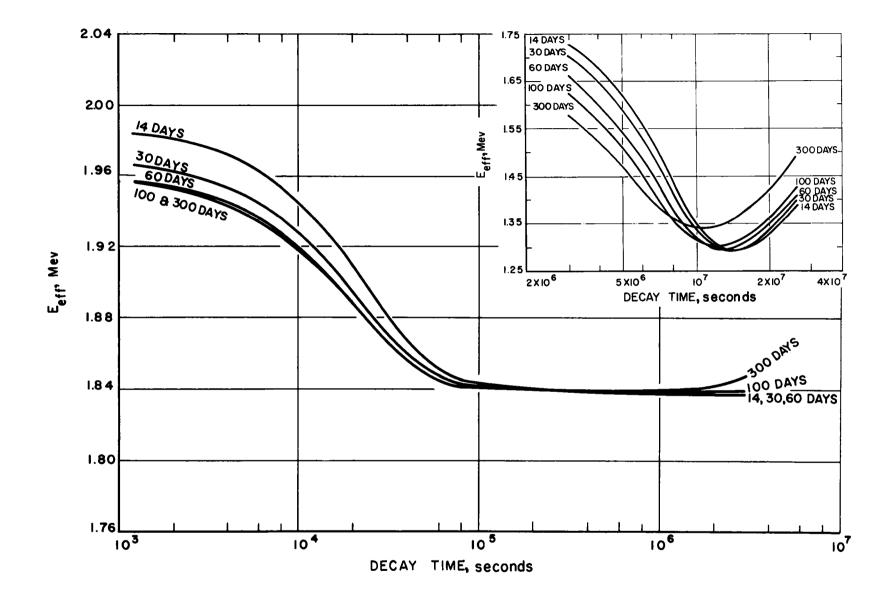


Fig. 21. Effective Energy as a Function of Decay and Irradiation Times for a 6-foot Ordinary Concrete Shield

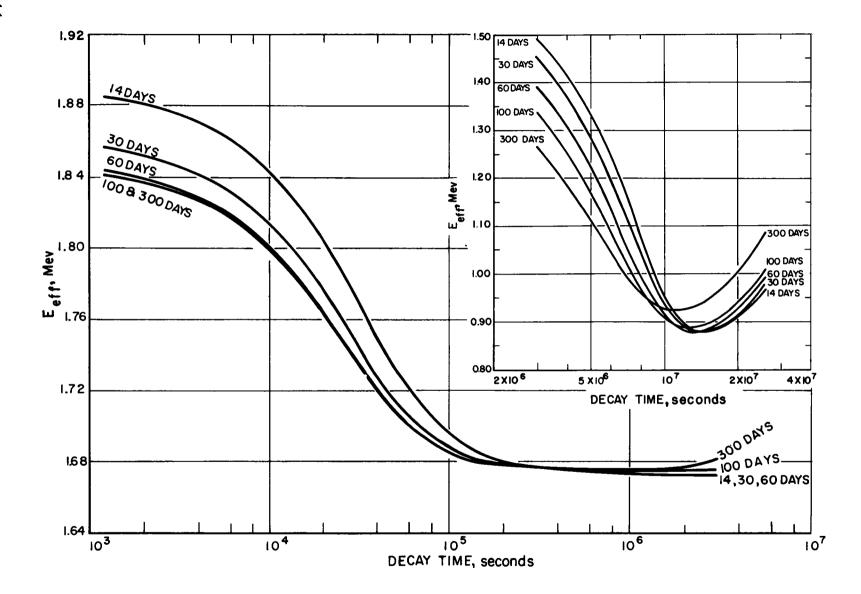


Fig. 22. Effective Energy as a Function of Decay and Irradiation Times for a 2-foot Heavy Concrete Shield

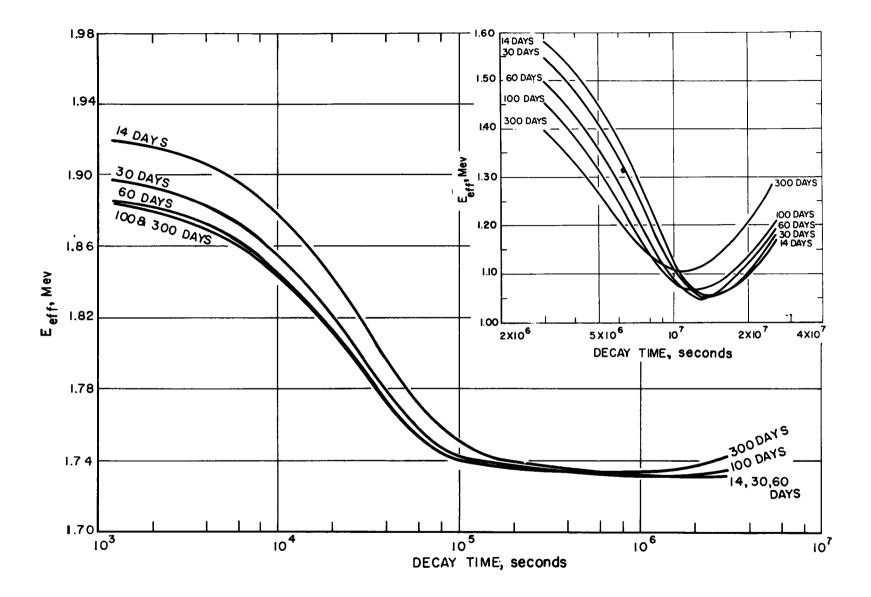


Fig. 23. Effective Energy as a Function of Decay and Irradiation Times for a 2.75-foot Heavy Concrete Shield

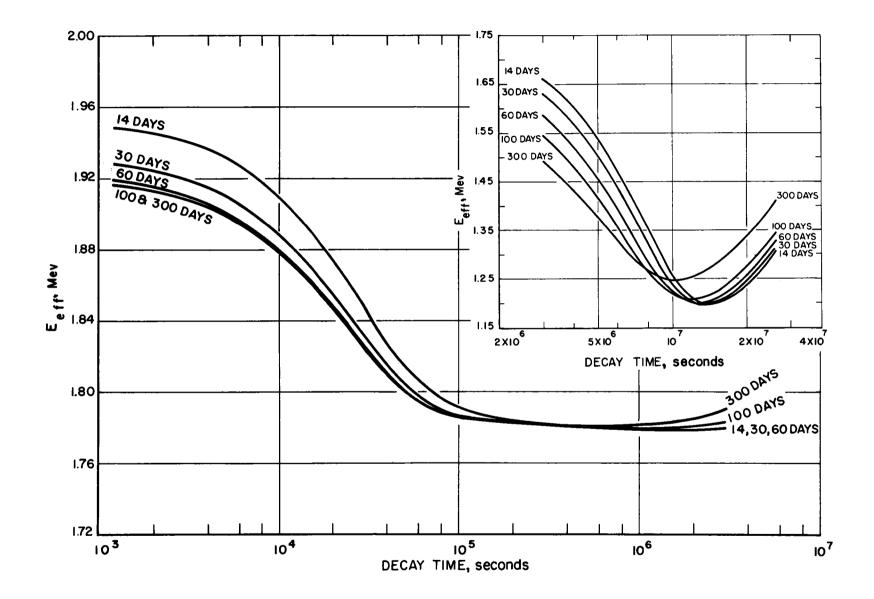


Fig. 24. Effective Energy as a Function of Decay and Irradiation Times for a 3.50-foot Heavy Concrete Shield

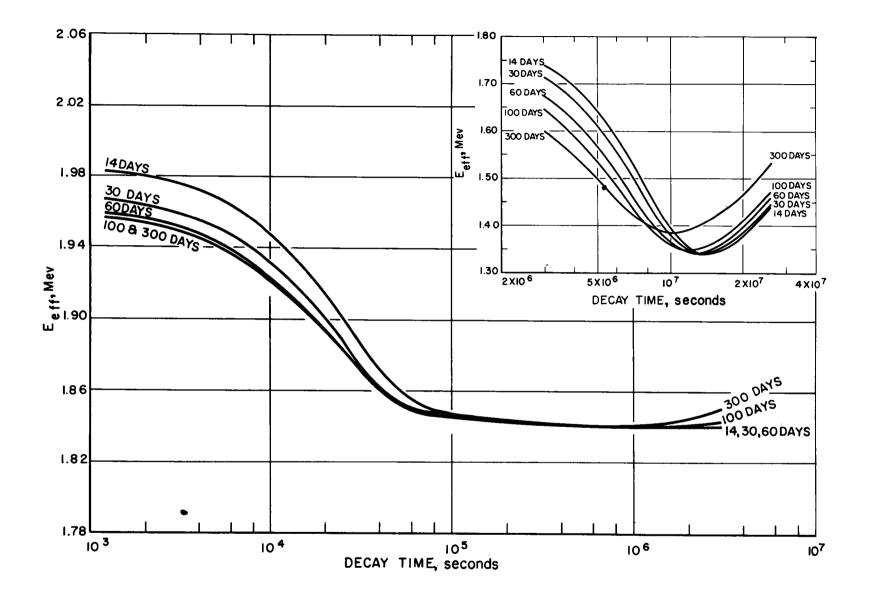


Fig. 25. Effective Energy as a Function of Decay and Irradiation Times for a 4.25- foot Heavy Concrete Shield

#### APPENDIX

# EXAMPLES USING THE EFFECTIVE ENERGY METHOD

### TYPE A. DOSE RATE FROM FIXED SHIELD

#### Case 1. Constant Power Level:

It is desired to evaluate the dose rate on the horizontal centerline at a distance of one meter from a spent  $U^{235}$  fuel element contained in a coffin with a wall thickness x of 6 inches. The fuel element is a solid right cylinder of natural uranium, 3-feet long, with an OD of 3 inches. The element has been irradiated at a constant power level P<sub>o</sub> of 150 kilowatts for a period  $\tau$  of 100 days, and cooled for a period t of 1 day. Summarizing these data:

$$\tau$$
 = 100 days,  
t = 1 day = 8.64 x 10<sup>4</sup> seconds,  
x = 6 inches,

and

 $\mathbf{P}_{c} = 150$  kilowatts.

In solving this problem, three different assumptions will be used with respect to the geometry involved. The results will become progressively more accurate.

## Solution 1. Point Source Approximation:

Taking the simplest approach, treat the source as a point, and following the procedure in Step 1 of Section V, referring to Fig. 1 for a 100-day irradiation  $\tau$  and 1-day cooling t, the hard fission product gamma-ray power  $P_{ur}$  is

$$P_w = 3.40 \times 10^9 \text{ Mev/sec-watt.}$$

Since the fuel element was operated at a constant power level of 150 kilowatts, the intensity of the hard gamma-rays  $P_{z}$  from the source will be

$$P_{g} = P_{w}P_{o} \qquad \dots (1A)$$
  
= (3.40 x 10<sup>9</sup>) (150 x 10<sup>3</sup>)  
= 5.10 x 10<sup>14</sup> Mev/sec.

Following the procedure in Step 2 of Section V, the effective energy as obtained from Fig. 14 is

$$E_{eff} = 1.662 \text{ Mev.}$$

The dose rate R, in roentgens per hour at one meter from this point source, can be determined by evaluation of the equation

$$R = \frac{P_s B_r e^{-\mu x}}{4 \pi a^2 k} \qquad \dots (2A)$$

where  $B_r = dose buildup factor for the shield material,$ 

 $\mu$  = linear absorption coefficient of the shield, cm<sup>-1</sup>,

a = horizontal distance between the source and detector, = 100 cm,

and k = conversion factor from energy flux to dose rate,  $\frac{Mev}{cm^2-sec}/\frac{r}{hr}$ .

To solve this expression, the specific values of  $\mu$ ,  $B_r$ , and k must correspond to the effective energy of 1.662 Mev.

From Fig. 5,  $\mu$  for lead for 1.662 Mev gamma rays is

$$\mu = \mu_{m} \rho \qquad \dots (3A)$$
  
= (0.0480) (11.3)  
= 0.542 cm<sup>-1</sup>

where  $\mu_{\rm m}$  = mass absorption coefficient, cm<sup>2</sup>/gm, obtained from Fig. 5, and  $\rho$  = shield density, gm/cm<sup>3</sup>.

Therefore,  $\mu x = (0.542)(15.24) = 8.26$ 

The dose buildup factor B<sub>r</sub>, for a penetration of 1.662 Mev gamma rays through 8.26 mean free paths, is 3.95 (Fig. 9). From Fig. 3,  $k = 5.84 \times 10^5 \frac{Mev}{cm^2-sec/hr}$ .

Substituting in Eq. (2A), the dose rate is

$$R = \frac{(5.10 \times 10^{14}) (3.95) e^{-8.26}}{4 \pi (100)^2 (5.84 \times 10^5)}$$
$$= 7.06 r/hr.$$

Solution 2. Line Source Approximation (No Self-absorption):

Representing the source by a line of length h, the source strength per unit length  ${\bf S}_{\rm h}$  is

$$S_{h} = \frac{\frac{P_{s}}{h}}{\frac{5.10 \times 10^{14}}{91.44}}$$
  
= 5.58 x 10<sup>12</sup> Mev/sec-cm.

For this source geometry, the dose rate R along the horizontal centerline from a shielded line source may be evaluated using the equation

$$R = \frac{S_h B_r}{2 \pi a k} F(\theta, \mu x), \qquad \dots (5A)$$

where

$$\mathbf{F}(\theta,\mu\mathbf{x}) = \int_{0}^{\theta} e^{-\mu\mathbf{x} \sec \phi} d\phi,$$

and

$$\theta = \tan^{-1} \frac{h}{2a} = \tan^{-1} \frac{91.44}{2(100)}$$

and since  $\mu \mathbf{x} = 8.26$ , as in Solution 1 above,  $\mathbf{F}(\theta, \mu \mathbf{x}) = 8.57 \times 10^{-5}$ . Substituting in Eq. (5A), the dose rate is

$$R = \frac{(5.58 \times 10^{12}) B_r}{2 \pi (100) (5.84 \times 10^5)} (8.57 \times 10^{-5})$$
$$= 1.30 B_r r/hr.$$

To evaluate the dose buildup factor, proceed as follows:

The unshielded dose rate on the horizontal centerline at one meter from this line source is given by

$$R_{u} = \frac{S_{h}}{2 \pi a k} tan^{-1} \frac{h}{2a}$$
 ... (6A)

Since the unscattered dose rate arriving at the detector after penetration through the shield is  $R/B_r$ , the effective attenuation that the shield offers to this source is simply

$$\frac{\frac{R}{B_r}}{\frac{R_u}{R_u}} = \frac{F(\theta, \mu_x)}{\tan^{-1} \frac{h}{2a}}, \dots (7A)$$

The absolute magnitude of the natural logarithm of Eq. (7A) corresponds to the effective number of mean free paths,  $(\mu x)_{eff}$ , through which the source radiation had penetrated, and therefore can be used to obtain the buildup factor. In this case

$$(\mu \mathbf{x})_{eff} = \left| \ln \frac{\mathbf{F}(\theta, \mu \mathbf{x})}{\tan^{-1} \frac{h}{2a}} \right|$$
 ... (8A)  
=  $\left| \ln \frac{8.57 \times 10^{-5}}{0.4288} \right|$   
= 8.51.

From Fig. 9, the dose buildup factor corresponding to the penetration of 1.662 Mev gamma rays through 8.51 mean free paths is 4.03. Therefore, the dose rate at one meter is

$$R = (1.30) (4.03) = 5.24 r/hr.$$

# Solution 3. Cylindrical Source (With Self-absorption):

Using the method of Foderaro and Obenshain<sup>10</sup> which includes the effect of self-absorption, the source cylinder is replaced by a line source of equivalent total source strength. The line source is positioned a distance z within the true source cylinder, the distance z being just sufficient to yield an attenuation equivalent to the actual self-absorption. The dose rate for this source geometry<sup>10</sup> is

$$R = \frac{S_h B_r}{2\pi (a+z)k} F\left[\theta, (\mu x + \mu_s z)\right] \qquad \dots (9A)$$

where  $\mu_s$  = linear absorption coefficient of the source material, for gamma rays of the effective energy, cm<sup>-1</sup>,

and a = distance from the surface of the true source to the detector, 100 cm. To determine z, proceed as follows:

As was shown previously for a 6-inch lead shield,  $E_{eff} = 1.662$  Mev. From a curve of the linear absorption coefficient for uranium,  $^7\mu_{g} = 0.971$  cm<sup>-1</sup>. Since a/R<sub>0</sub>>10, where R<sub>0</sub> = actual fuel element radius = 3.81 cm,

$$\mu_{\mathbf{s}} \mathbf{R}_{\mathbf{o}} = (0.971)(3.81)$$
$$= 3.70.$$

From the graph on page 24 of the reference,  ${}^{10}$   $\mu_{g}z = 1.73$ . Hence,

$$z = \frac{1.73}{0.971}$$
  
= 1.78 cm

Using linear absorption coefficient relationships, the source self-absorption thickness is equivalent to a lead thickness  $z_{Pb}$  of

$$z_{Pb} = \frac{\mu_{s} z}{\mu}$$
 ...(10A)  
=  $\frac{1.73}{0.542}$   
= 3.19 cm.

Therefore, the total lead shielding  $x_{eff}$  offered to this line source, is equivalent to

$$x_{eff} = x + z_{Pb}$$
 ...(11A)  
= 6 +  $\frac{3.19}{2.54}$ 

From a curve of  $E_{eff} \underline{vs}$  lead thickness, which was obtained by cross plotting data from Fig. 14 through 17 at the specific values of  $\tau$  and t under consideration,  $E_{eff}$  for 7.26 inches of lead is 1.671 Mev. Thus,  $\mu_m$  for this shield will be 0.0480 cm<sup>2</sup>/gm and hence,  $\mu = 0.542$  cm<sup>-1</sup>.

= 7.26 inches.

Since the line source is now positioned within the actual source, the angle  $\theta$  will change, and becomes

$$\theta = \tan^{-1} \frac{h}{2(a+z)} \dots (12A)$$
  
=  $\tan^{-1} \frac{91.44}{2(100+1.78)}$   
= 24°12' = 0.4225 radians.

Since  $E_{eff}$  has changed, there will be a change in k. Substituting in Eq. (9A), with  $k = 5.85 \times 10^5$ ,  $\mu_x + \mu_s z = 8.26 + 1.73 = 9.99$ , and  $F[\theta, (\mu_x + \mu_s z)] = 1.44 \times 10^{-5}$ , the dose rate R becomes

$$R = \frac{5.58 \times 10^{12} B_r (1.44 \times 10^{-5})}{2\pi (100 + 1.78) (5.85 \times 10^5)}$$
$$= 0.215 B_r r/hr.$$

The buildup factor is evaluated in the same manner as for Solution 2, and it is assumed that all the penetration is in the lead. (In the example considered, this assumption will lead to a very small error. However, when the source is very large and  $\mu_{\rm g}$ z approaches  $\mu$ x, the method of assuming all the buildup in one

material can lead to large errors, especially when the atomic numbers of the two media differ appreciably.) For the case at hand, using Eq. (7A),

$$\frac{\frac{R}{B}_{r}}{R_{u}} = \frac{1.44 \times 10^{-5}}{0.4225}$$
$$= 3.40 \times 10^{-5},$$

and

$$(\mu x)$$
 =  $|\ln 3.40 \times 10^{-5}|$   
eff

= 10.3.

From Fig. 9, for 1.671 Mev gamma rays,  $B_r = 4.67$ . Therefore, the dose rate is

$$R = (4.67) (0.215)$$
$$= 1.00 r/hr.$$

### Case 2. Operation at Different Power Levels:

In evaluating problems in which the fuel element was not operated at a constant power level, the procedure to be used is the same as in the above calculations, with only one modification. Specifically, every discrete period of constant power operation should be evaluated. Then, the solution will be the sum of the dose rates due to each of these intervals. The procedure is more lengthy, though no more complicated, than that for Case 1. However, with proper averaging of the power levels in adjacent time intervals, the amount of labor can be reduced considerably, the entire procedure requiring much less time than other techniques for the solution of this type of problem.

#### TYPE B: SHIELD THICKNESS TO BE DETERMINED

Suppose it is desired to evaluate the shielding required by the spent fuel described in Type A, Case 1, above, in order that the spent fuel may be transported by public carrier to another site. To meet Interstate Commerce Commission (ICC) tolerance requirements, a lead shield thickness must be determined which will reduce the maximum radiation level to 10 mr/hr at a centerline distance of one meter from the fuel. (Note that for the size of the shields being investigated in these examples, satisfying the 10 mr/hr requirement will also satisfy the 200 mr/hr ICC shield surface tolerance requirement.)

## Case 1. Constant Power Level:

Solution 1. Point Source Approximation:

In the previous point source calculation relative to this fuel element, the dose rate was found to be 7.06 r/hr, or 7060 mr/hr. This dose rate is greater than permissible, hence additional shielding will be required. Let us try a 10-inch shield. (Any intermediate thickness could be used but  $E_{eff}$  would have to be obtained by cross plotting of the data from Fig. 14 through 17.) From Fig. 16, for a 10-inch lead shield,  $E_{eff} = 1.692$  Mev.

From Fig. 5,  $\mu_{\rm m} = 0.0476 \text{ cm}^2/\text{gm}$ , hence  $\mu = 0.539 \text{ cm}^{-1}$ . Therefore,

$$\mu = (0.539) (25.4)$$
  
= 13.7,

and from Fig. 9 for 1.692 Mev gamma rays penetrating through 13.7 mean free paths,  $B_r = 5.86$ . From Fig. 3, k is 5.86 x  $10^5 \frac{Mev}{cm^2 - sec} / \frac{r}{hr}$ .

Substituting in Eq. (2A), the dose rate is

$$R = \frac{(5.10 \times 10^{14}) (5.86) e^{-13.7}}{4 \pi (100)^2 (5.86 \times 10^5)}$$
$$= 4.55 \times 10^{-2} r/hr.$$

This dose rate is still too high, so let us evaluate a 12-inch lead shield. Repeating the procedure above, the dose rate from the 12-inch shield is found to be  $3.92 \times 10^{-3}$  r/hr, which is too low. Obviously then, the required thickness is somewhere between 10 and 12 inches of lead.

In order to determine this value, a curve of dose rate vs shield thickness can now be made using the data obtained from the three point-source calculations, and the required shield is then obtained by interpolation. Following this procedure, the required shield thickness is found to be 11.2 inches of lead.

## Solutions 2 and 3:

The same iterative type of procedure as carried out in Solution 1 above will yield the desired shield thickness, the procedures being just somewhat more lengthy.

## Case 2. Operation At Different Power Levels:

Again, the same procedure is used, except that the contribution from every discrete period of constant power operation should be evaluated. The resultant dose rate from any given shield thickness will be the sum of the individual contributions.

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