DESIGN STUDIES ON CESIUM-137 AS A SOURCE FOR HIGH LEVEL GAMMA IRRADIATORS

by

L. Voyvodic

Prepared for

The Office of Isotopes Development
United States Atomic Energy Commission

December 5, 1960

ARMOUR RESEARCH FOUNDATION
of
Illinois Institute of Technology
Technology Center
Chicago 16, Illinois
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DESIGN STUDIES ON CESIUM-137 AS A SOURCE FOR HIGH LEVEL GAMMA IRRADIATORS

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The Office of Isotopes Development
United States Atomic Energy Commission
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FINAL REPORT

by

ARMOUR RESEARCH FOUNDATION
of
Illinois Institute of Technology
Technology Center
Chicago 16, Illinois

Report written by
L. Voyvodic

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This is the final report on ARF Project 1150 entitled, "Design Studies on Cesium-137 as a Source for High Level Gamma Irradiators." This report covers activities between June 1, 1959 to July 31, 1960, and was conducted for the U. S. Atomic Energy Commission, Office of Isotopes Development, under Contract No. AT(11-1)-779.

Personnel who have contributed to this program include: J. Ezop, I. Filosofo, T. Klippert, C. A. Stone, G. Vossler and L. Voyvodic.

Data concerning this project can be found in ARF logbooks C9729, C9994, C9999, C10084, C10158, C10163, and C10206.

Respectfully submitted,

ARMOUR RESEARCH FOUNDATION
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L. Reifel
Director of Physics Research
We have studied, analytically and experimentally, the influence of design factors on the efficiency, depth dose uniformity and dose rate of high level, plaque cesium-137 irradiators. Approximate theoretical expressions have been derived for irradiators with plaque sources and slab absorbers where the attainment of high efficiencies, together with satisfactory dose rate and dose uniformity are particularly stringent requirements. The irradiator performance relationships are expressed in terms of a geometry factor, $G'$, which governs the spatial dose rate distribution within the absorber.

The form of $G'$ found particularly informative and useful for analytical design studies is based on multiple scattering calculations:

$$G'(b) = K_o E_1(b) + K_s \beta e^{-b}$$

where the term $E_1(b)$ is due to the uncollided photon flux from an equivalent infinite plane source, $\beta e^{-b}$ is due to the presence of scattered radiation, and the factors $K_o$ and $K_s$ represent the total corrections required for finite source lateral dimensions, non-homogeneity in atomic composition, presence of air gaps, etc. For sources with very large lateral dimensions, where $K_o \sim K_s \sim 1$, and six inch water absorbers, the calculated values of the geometry factor $G'$ appear to be nearly independent of gamma ray energy. Also, the depth dose variation quickly becomes exponential due to the dominant effect of the scattered radiation. The scattered photons are expected from theory to be strongly peaked in the region of 60 Kev in a water absorber, again nearly independent of the initial source photon energy.
energies.

For a single plaque source irradiating alternate faces of a slab absorber of thickness $t$, the effective dose rate, at half-depth in the absorber, becomes

$$\bar{D} = 2 \pi k_a h S_m \overline{G'}.$$  

The efficiency of the irradiator, based on the energy absorption at half-depth, is

$$F' = \left( \frac{\mu_r t}{\mu_a} \right) \left( \frac{H_t}{H_s} \right) \left( \frac{L_t}{L_s} \right) \overline{G}.'$$

The depth dose rate uniformity is expressed as

$$U = \left( \frac{G_f' + G_b'}{2} \right) \overline{G'}$$

where

$$\overline{G'} = G'(t/2)$$

is the geometry factor at half-depth,

$h, H_s, L_s$ are the source thickness, height and length,

$H_t, L_t$ are the effective target height and length over which the effective dose rate has the value $\bar{D}$,

$S_m$ is the source material specific activity,

and $G_f', G_b'$ are the geometry factors at the front and back surfaces of the slab absorber.

Experiments carried out with low-level cesium-137 plaque sources, with water and paraffin slab absorbers, were designed to test the theoretical predictions. Using scintillation dose rate meters and photon spectrometers, the observations shown in Figs. 1 to 11 on the absolute dose rates, the dose rate distribution and $G'$ factor, as well as the spectral distribution of gamma radiation within the absorbers, were all found to be in satisfactory agreement with the predicted performance.
The economic attractiveness of cesium-137 as source material for high level irradiators is found to be particularly sensitive to the overall efficiency of the irradiator. Based on plausible estimates for capital charges and radiation cell costs, plus a high duty cycle, it appears that cesium-137 could provide irradiation costs as low as $1 per Kilowatt-hour of absorbed energy, or 0.12 cents/megarad pound, if it were available at about 10 cents/curie and used in large irradiators at about 67 percent useful conversion efficiency.

It is recommended that similar studies be carried out for irradiators with other designs and geometries of interest in high level radiation processing such as cylindrical source systems. The potential economic attractiveness coupled with increased dose uniformity and reduced shielding requirements indicate the desirability of further study.
# Table of Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABSTRACT</td>
<td>v</td>
</tr>
<tr>
<td>LIST OF FIGURES</td>
<td>ix</td>
</tr>
<tr>
<td>LIST OF TABLES</td>
<td>xi</td>
</tr>
<tr>
<td>I. INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>II. RADIATION PROPERTIES OF CESIUM-137 SOURCES</td>
<td>1</td>
</tr>
<tr>
<td>A. Decay Scheme and Power Output</td>
<td>1</td>
</tr>
<tr>
<td>B. Absorption and Scattering Cross-Sections</td>
<td>2</td>
</tr>
<tr>
<td>C. Dose Conversion Factors</td>
<td>4</td>
</tr>
<tr>
<td>D. Source Material</td>
<td>4</td>
</tr>
<tr>
<td>III. ANALYTICAL STUDIES</td>
<td>4</td>
</tr>
<tr>
<td>A. Source Economics</td>
<td>4</td>
</tr>
<tr>
<td>B. Irradiator Performance Relationships</td>
<td>7</td>
</tr>
<tr>
<td>C. Plaque Source Geometry Factor, G'</td>
<td>9</td>
</tr>
<tr>
<td>D. Effect of Source Gamma Energy</td>
<td>11</td>
</tr>
<tr>
<td>E. Finite Plaque Sources</td>
<td>11</td>
</tr>
<tr>
<td>F. Effect of Source Thickness and Filters</td>
<td>13</td>
</tr>
<tr>
<td>IV. EXPERIMENTAL STUDIES</td>
<td>16</td>
</tr>
<tr>
<td>A. Experimental Arrangement</td>
<td>16</td>
</tr>
<tr>
<td>B. Dose Rate Measurements on Plaque Sources</td>
<td>18</td>
</tr>
<tr>
<td>C. Photon Spectral Measurements</td>
<td>27</td>
</tr>
<tr>
<td>V. INTERPRETATION AND CONCLUSIONS</td>
<td>33</td>
</tr>
<tr>
<td>REFERENCES</td>
<td>39</td>
</tr>
<tr>
<td>Figure No.</td>
<td>Description</td>
</tr>
<tr>
<td>-----------</td>
<td>------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>Fig. 1</td>
<td>Depth dose variation in water for three cesium-137 plaque sources with dimensions 20 inches x 20 inches, 20 inches x 60 inches and 40 inches x 40 inches, and calculated variation for an infinite plane isotropic source.</td>
</tr>
<tr>
<td>Fig. 2</td>
<td>Depth dose variation in water with 40 inches x 40 inches plaque cesium-137 source, showing effect of water absorber thickness.</td>
</tr>
<tr>
<td>Fig. 3</td>
<td>Depth dose variation in water due to 40 inches x 40 inches plaque cesium-137 source, showing effects of composition of the filter placed between source and water absorber. All filters, iron, cadmium and lead, were 1/16 inch thickness sheets.</td>
</tr>
<tr>
<td>Fig. 4</td>
<td>Geometry factor G' derived for experimental curves A and D of Fig. 3. Calculated curves for the uncollided flux contribution (E_1(b')) and total flux component (G'(b')) are shown for an infinite plane isotropic cesium-137 source in water.</td>
</tr>
<tr>
<td>Fig. 5</td>
<td>Depth dose variation in paraffin with 40 inches x 40 inches plaque cesium-137 source, 1/16 inch steel filter, showing effect of air gap between source and absorbers.</td>
</tr>
<tr>
<td>Fig. 6</td>
<td>Depth dose variation in paraffin with 40 inches x 40 inches plaque cesium-137 source, with 2 inch air gap, showing filtration effect of steel absorbers.</td>
</tr>
<tr>
<td>Fig. 7</td>
<td>Depth dose variation in paraffin with 40 inches x 40 inches plaque cesium-137 source, with 2 inch air gap, showing filtration effects of composite filters between source and absorber.</td>
</tr>
<tr>
<td>Fig. 8</td>
<td>Off-axis dose rate variations at three depths in paraffin slab absorber with 40 inches x 40 inches cesium-137 plaque source. In the set shown with open circles (o), the absorbers A and B were present on both sides of the detector. In the set shown crossed (x), only absorber A was present.</td>
</tr>
</tbody>
</table>
### LIST OF FIGURES

(Continued)

<table>
<thead>
<tr>
<th>Figure No.</th>
<th>Description</th>
<th>Page No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fig. 9</td>
<td>Observed photon spectral distribution at several depths in a paraffin absorber irradiated by a 40 inches x 40 inches cesium-137 plaque source. The dashed curve illustrates the observed spectrum in the absence of the absorber.</td>
<td>30</td>
</tr>
<tr>
<td>Fig. 10</td>
<td>Observed photon spectral distributions at several depths in water, irradiated by a 40 inches x 40 inches cesium-137 plaque source.</td>
<td>31</td>
</tr>
<tr>
<td>Fig. 11</td>
<td>Spectral profiles at front surface of paraffin absorber, showing the effects of various filters in the source assembly</td>
<td>32</td>
</tr>
</tbody>
</table>
# LIST OF TABLES

<table>
<thead>
<tr>
<th>Table No.</th>
<th>Description</th>
<th>Page No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>Cross-Sections for Cesium-137 Gamma Rays.</td>
<td>3</td>
</tr>
<tr>
<td>II</td>
<td>Source Characteristics for Gamma Irradiators and Water Equivalent Absorbers</td>
<td>5</td>
</tr>
<tr>
<td>III</td>
<td>Approximate Behavior of Infinite Plaque Gamma Irradiators for 6 Inch Thick Water Slab Absorbers</td>
<td>12</td>
</tr>
<tr>
<td>IV</td>
<td>Approximate Depth Dose and Edge Effect Correction Factors for Long Plaque Sources of Height $H_s$</td>
<td>14</td>
</tr>
<tr>
<td>V</td>
<td>Approximate Dose Rate Variations For a Large Area Cesium-137 Plaque Source and 6 Inch Water Absorber.</td>
<td>15</td>
</tr>
<tr>
<td>VI</td>
<td>Results from Cesium-137 Plaque Sources and Six Inch Water Absorbers</td>
<td>34</td>
</tr>
</tbody>
</table>
DESIGN STUDIES ON CESIUM-137 AS A SOURCE FOR HIGH LEVEL GAMMA IRRADIATORS

I. INTRODUCTION

The purpose of this study has been to examine the properties and limitations of fission product cesium-137 as source material for high level gamma irradiators. The approach here emphasizes the radiation physics aspects of effect irradiator design, keeping in mind at the same time the engineering and economic aspects important to practical applications.

Considering only the penetration of the initial source photons, (uncollided flux) the analysis and design of gamma irradiators present few difficulties. However, the presence of scattered radiation introduces several important features, which arise from the fact that the energy absorption due to scattered radiation is affected by the source photon energy as well as by the configuration, dimensions and atomic composition of the irradiator.

The first part of this report covers our analytical studies on irradiator design and performance; the comparison of cesium-137 with other sources such as cobalt-60 in applications comparable to the High Intensity Food Irradiator; the expected effects of finite source dimensions, air gaps and filters on irradiator efficiency, dose rate and dose uniformity. The second part describes results obtained with low-level experimental cesium-137 sources, using both dose rate and photon spectra measuring techniques. Finally the results and conclusions of this study are summarized.

II. RADIATION PROPERTIES OF CESIUM-137 SOURCES

A. Decay Scheme and Power Output

As reviewed by Strominger et al., cesium-137 decays primarily...
by beta emission with a maximum energy of 0.514 Mev, followed by a single gamma ray of 0.662 Mev from barium-137. In 8 percent of the decays, there is a beta of 1.17 Mev maximum energy with no gamma rays. In the gamma decay of barium-137 from the 0.662 Mev level, which has a half life of 2.6 minutes, 10 percent of the transitions occur by internal conversion, giving electrons of 624 Kev as well as characteristic barium x-rays.

The average energy of gammas per disintegration should then be

\[ 0.662 \times 0.92 \times 0.90 = 0.55 \text{ Mev} \]

This corresponds to a gamma power of

\[ P_\gamma = 3.7 \times 10^{16} \times 0.55 \times 1.6 \times 10^{-6} \times 10^{-10} = 3.27 \text{ Kilowatt/Megacurie} \] (1)

The average energy of the betas and conversion electrons per disintegration is 240 Kev. The corresponding beta power is \( P_\beta = 1.43 \) Kilowatt/Megacurie, or 44 percent of the gamma power radiated. This high beta energy output of cesium-137, which is of concern in self-heating, is to be compared with the smaller fraction of only 4 percent of the gamma power in the case of cobalt-60 sources.

The half life of cesium-137 is nearly 28 years, corresponding to an annual renewal requirement of about 2.5 percent if the source strength is to be kept constant in an irradiator. Also, from the half life, the maximum possible specific activity for cesium-137 sources is \( S_m \approx 100 \text{ curies/gm} \).

B. Absorption and Scattering Cross-Sections

The values for the attenuation coefficient \( \mu \) and energy absorption coefficient \( \mu_a \) used in this investigation were obtained from Rockwell\(^2\) and Grodstein\(^3\) and are listed in Table I. The scattering cross-section
### TABLE I
CROSS-SECTIONS FOR CESIUM-137 GAMMA RAYS
(0.662 Mev)

<table>
<thead>
<tr>
<th>Material</th>
<th>Z</th>
<th>$\mu$ \text{ cm}^2/gm</th>
<th>$\mu_a$ \text{ cm}^2/gm</th>
<th>$\sigma_s = \mu - \mu_a$ \text{ cm}^2/gm</th>
<th>$\sigma_s / \mu_a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrogen</td>
<td>1</td>
<td>0.152</td>
<td>0.059</td>
<td>0.093</td>
<td>1.57</td>
</tr>
<tr>
<td>C, N, O, Air</td>
<td>6-8</td>
<td>0.0765</td>
<td>0.0295</td>
<td>0.047</td>
<td>1.60</td>
</tr>
<tr>
<td>Tissue</td>
<td>-</td>
<td>0.0825</td>
<td>0.0320</td>
<td>0.0505</td>
<td>1.58</td>
</tr>
<tr>
<td>Water</td>
<td>-</td>
<td>0.0855</td>
<td>0.0328</td>
<td>0.0527</td>
<td>1.60</td>
</tr>
<tr>
<td>Aluminum</td>
<td>13</td>
<td>0.0755</td>
<td>0.0287</td>
<td>0.0468</td>
<td>1.62</td>
</tr>
<tr>
<td>Iron</td>
<td>26</td>
<td>0.0715</td>
<td>0.0270</td>
<td>0.0445</td>
<td>1.65</td>
</tr>
<tr>
<td>Copper</td>
<td>29</td>
<td>0.069</td>
<td>0.0255</td>
<td>0.0435</td>
<td>1.72</td>
</tr>
<tr>
<td>Tin</td>
<td>50</td>
<td>0.0715</td>
<td>0.0332</td>
<td>0.0383</td>
<td>1.15</td>
</tr>
<tr>
<td>Tantalum</td>
<td>73</td>
<td>0.098</td>
<td>0.059</td>
<td>0.039</td>
<td>0.66</td>
</tr>
<tr>
<td>Lead</td>
<td>82</td>
<td>0.113</td>
<td>0.074</td>
<td>0.039</td>
<td>0.53</td>
</tr>
<tr>
<td>Uranium</td>
<td>92</td>
<td>0.135</td>
<td>0.098</td>
<td>0.037</td>
<td>0.38</td>
</tr>
<tr>
<td>Sodium Iodide</td>
<td></td>
<td>0.073</td>
<td>0.032</td>
<td>0.041</td>
<td>1.28</td>
</tr>
</tbody>
</table>
(\sigma_s = \mu - \mu_a\), and the ratio \(\sigma_s/\mu_a\), shown in Table I, were derived for estimating scattering build-up effects.

C. **Dose Conversion Factors**

Using the values of \(\mu_a\) given in Table I, the expected specific dose rates for cesium-137 point sources are

\[
k_r = \left(\frac{3.7 \times 10^{10}}{4\pi}\right) \times (0.55 \times 1.6 \times 10^{-6}) \times 3600 \times \left(\frac{0.0295}{87}\right)
\]

\[= 3.15 \times 10^3 \text{ r/hr/curie at 1 cm in air}\] (2)

\[
k_a = \left(\frac{3.7 \times 10^{10}}{4\pi}\right) \times (0.55 \times 1.6 \times 10^{-6}) \times 3600 \times \left(\frac{0.0328}{100}\right)
\]

\[= 3.05 \times 10^3 \text{ rads/hr/curie at 1 cm in water}\] (3)

D. **Source Material**

The fission product cesium-137 source material, as described in the Radioisotope Catalogue and in a brochure by the Isotopes Division of ORNL, is presently available in the form of cesium chloride with a specific activity \(S_m = 22 \text{ curies per gram of solid}\). The density of source material \(\rho_s\) is expected to approach \(3 \text{ gm/cm}^3\). From its similarity to the composition of sodium iodide, source attenuation coefficients for cesium chloride have been assumed to approximate those shown for sodium iodide at the bottom of Table I.

Table II lists the more important cesium-137 characteristics which have been found of interest for irradiator analysis, as well as the corresponding features for several other gamma sources.

III. **ANALYTICAL STUDIES**

A. **Source Economics**

In reviewing the factors governing the economic use of cesium-137
TABLE II

SOURCE CHARACTERISTICS FOR GAMMA IRRADIATORS AND WATER EQUIVALENT ABSORBERS

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$</th>
<th>$E$</th>
<th>$P_y$</th>
<th>$\mu$</th>
<th>$\mu_a$</th>
<th>$\sigma_s/\mu_a$</th>
<th>$k_a \times 10^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>yrs.</td>
<td>Mev</td>
<td>Kw/Mc</td>
<td>gm/cm$^2$</td>
<td>gm/cm$^2$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cesium-137</td>
<td>28</td>
<td>0.66</td>
<td>3.27</td>
<td>0.086</td>
<td>0.033</td>
<td>1.6</td>
<td>3.05</td>
</tr>
<tr>
<td>Cobalt-60</td>
<td>5.2</td>
<td>1.17</td>
<td>1.33</td>
<td>0.063</td>
<td>0.030</td>
<td>1.1</td>
<td>12.6</td>
</tr>
<tr>
<td>Sodium-24</td>
<td>14 hrs.</td>
<td>1.38</td>
<td>2.76</td>
<td>0.050</td>
<td>0.026</td>
<td>0.9</td>
<td>17.8</td>
</tr>
<tr>
<td>Iridium-192</td>
<td>74 d.</td>
<td>0.4</td>
<td>5.9</td>
<td>0.11</td>
<td>0.033</td>
<td>2.4</td>
<td>5.2</td>
</tr>
</tbody>
</table>
in high level gamma irradiators, the earlier study by Beeley has been found to be a convenient point of reference.

An irradiation cost of 0.15 cent per megarad pound (or $1.20 per kilowatt-hour of absorbed energy) was estimated by Beeley for capital charges and maintenance, exclusive of source costs, for a radiation cell containing 7.8 megacuries of cesium-137; irradiation was assumed to be on a continuous basis (90 percent plant factor or 8,000 hours per year). Taking 0.36 cent per megarad pound as the competitive total irradiation cost for a sodium 24 reactor gamma facility (which is also near the mean of the estimated values for projected electron accelerator costs), the allowance permitted for competitive cesium-137 source costs in an irradiator with 20 percent efficiency came out to 3.7 cents per curie. The latter figure was based on 12 percent annual capital charges, plus 2.3 percent per year to make up for radioactive decay. Considering that the costs of source fabrication, shipping and installation will likely be at least several cents per curie, it may readily be seen that cesium-137 source material has very little economic attraction on this basis since current cesium costs are $1.00 per curie.

The situation is drastically altered if the cesium-137 gamma irradiator can be designed for high efficiency. Keeping all other factors the same, but assuming an improved irradiator to give 67 percent conversion efficiency, which is the value used by Beeley for projected high energy electron accelerators, the cesium-137 radiation cell charges drop from 0.15 to 0.045 cents per megarad pound. The allowance permitted for source costs then increased from 3.7 to 22 cents per curie of cesium-137 in the example above.

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Again, using the same parameters as above for source capital charges, plus annual renewal for radioactive decay, it may be shown that the source costs for cesium-137 can be expressed as

\[ Y_{Cs} = 18 FX \text{ cents/curie} \quad (4) \]

where \( X \) dollars/Kilowatt-hour absorbed are the source costs allowed for irradiation, and \( F \) is the irradiator fractional efficiency. On the same basis cobalt-60, with a 13 percent annual charge for source renewal, results in a source cost of

\[ Y_{Co} = 48 FX \text{ cents/curie} \]

In this form we see that if radiation processes are economic in the region of \( X \sim \$3/\text{Kilowatt-hour absorbed} \), or \( \sim 0.38 \text{ cent per megarad pound} \), and if irradiators of \( \sim 67 \text{ percent efficiency} \) are practical, then cesium-137 source costs up to about 36 cents/curie can be tolerated. (The corresponding source costs for cobalt-60 on this basis would be 96 cents/curie, as installed in the radiation cell.) Conversely, it is also evident from the above expression that if cesium-137 were available at \( \sim 10 \text{ cents/curie} \), by processing fission products in large quantities with low-cost encapsulation techniques, and irradiators of over 50 percent efficiency were employed, the costs of irradiation could be reduced to less than \$1 per Kilowatt-hour absorbed, i.e., less than 0.12 cent/megarad pound.

B. Irradiator Performance Relationships

The design analysis developed in the HIFI program\(^5,6\) for cobalt-60 sources has been examined in terms of cesium-137 as the source material. In the process it has also been found that the irradiator performance relationships can be expressed in simpler, and at the same time, in more general terms.
It was shown previously\(^5,6\) that for a uniform rectangular source approaching infinite dimensions, and with tightly packed 6 inch water equivalent package absorbers on either side, the total source activity required for irradiation of 3 megarad tons/hour or useful energy absorption at the rate of 7.6 Kilowatts, can be expressed in terms of a specific dose rate factor, \(\bar{G}\), as follows:

\[
S = 8.9 \times 10^4 / \bar{G} \text{ megacuries.} \tag{6}
\]

The effective dose rate \(\bar{D}\), at half-depth in the absorber, is similarly expressed as:

\[
\bar{D} = S_a \bar{G} = S_m h \bar{G} \text{ rads/hour} \tag{7}
\]

where \(S_a\) is the effective source strength per unit area, \(S_m\) is the mass specific activity and \(h\) the source thickness.

In the present study we have examined the irradiator performance in terms of a more convenient reduced geometry factor \(G'\), where

\[
G'(b) = G(b)/2 \pi k_a. \tag{8}
\]

Consider now a uniform plaque source of strength \(S\), with finite dimensions (height \(H_s\), length \(L_s\) and thickness \(h\)), producing a minimum dose \(I_{\text{min}}\) in a residence time \(T_r\) (or \(\bar{D} = I_{\text{min}}/T_r\)) for alternate face exposures of a slab target with thickness \(t\), height \(H_t\) and effective length \(L_t\). Next we express the source power \((SP_e)\) times efficiency \((F)\) as being equal to the rate of energy absorption at points of minimum dose in the absorber. One then obtains for the effective dose rate

\[
\bar{D} = 2\pi k_a S_a \bar{G}' \tag{9}
\]
where \( S_a = h S_m = S/H_s L_s \).  

The depth dose uniformity is given by

\[
U = \frac{I_{\text{max}}}{I_{\text{min}}} = \frac{[G'(0) + G'(t)]}{2 G'}.
\]  

The fractional irradiator efficiency can now be expressed as

\[
F = \left(\mu_a t\right) G' \left(\frac{H_t}{H_s}\right) \left(\frac{L_t}{L_s}\right).
\]  

The source requirements for an irradiator throughput capacity of \( C \) megarad tons/hour, or \( 2.53 C \) Kilowatts absorbed, is then simply

\[
S = 2.53 C/F P_\gamma \text{ Megacuries}
\]

where \( P_\gamma \) is the gamma power, as listed in Table II.

If the irradiator consists of multiple plaques and/or multiple absorber passes, this procedure can be readily extended for each plaque-absorber combination for deducing the effective dose rate, dose uniformity and efficiency values for the total irradiator, as will be illustrated later in Table III and Table VI.

C. Plaque Source Geometry Factor, \( G' \)

Several approaches have been used to obtain expressions for the geometry factor \( G' \), which may be useful under a variety of experimental conditions.

As in previous work\(^5,6\) following Goldstein\(^7\), we have used the approximation

\[
G'(b) = E_1(b) \left[ 1 + 2 \left( \frac{\sigma_s}{\mu_a} \right)^b \right]
\]

for an infinite source.

Using Taylor's form\(^2\) of expressing the NBS-NDA Moment
Method solutions for the build up factor in an infinite medium, we have

\[ G'(b) = A_1 E_1 [(1 + a_1)b] + A_2 E_2 [(1 + a_2)b] \]  \hspace{1cm} (15)

Taking the linear approximation for point source build up as

\[ B(b) = 1 + \beta b \]  \hspace{1cm} (16)

and carrying out the integration over an infinite isotropic plane, we have also derived the interesting relation

\[ G'(b) = E_1(b) + \beta e^{-b} . \]  \hspace{1cm} (17)

For the coefficient $\beta$ we have examined two approximations:

\[ \beta = \sigma_s/\mu_a \]  \hspace{1cm} (18)

\[ \beta = (A_1 a_1 + A_2 a_2) \]  \hspace{1cm} (19)

Relation (17) appears particularly useful, as it allows a direct separation of the uncollided photon flux component, $E_1(b)$, and the scattered radiation component, $\beta e^{-b}$. For finite irradiators each of these two terms can be modified by suitable correction factors. Under conditions where the scattered radiation predominates, relation (17) shows that the depth dose variation due to a plaque gamma source falls off exponentially, just as though one had a simple plane parallel beam of radiation instead of an assemblage of isotropic emitters. From a qualitative inspection of the scattered radiation energy spectrum, as derived from the Moment Method and discussed by Goldstein 7 and Rockwell,2 it would also appear that the important flux under these conditions is degraded in water to the vicinity of 60 KeV photon energy, regardless of the initial choice of gamma emitter.
D. **Effect of Source Gamma Energy**

On the basis of the preceding analysis, and the use of the approximate relation (14) for the plane source build up factor, calculations were made for infinite plaque irradiators and six inch thick water absorbers over a range of initial source photon energies from 0.4 to 3 Mev. This includes iridium-192, cobalt-60, sodium-24, as well as the cesium-137 source material of primary interest in this study. A simplifying feature which emerged is that one inch of water equivalent absorber is indicated as the required source assembly thickness, \( b_s \), for removing the steep field gradient near the source at all of the photon energies considered. Further, by selecting the source cladding to be 1/16 inch of stainless steel, this condition leads to recommended source thicknesses up to 3 gm/cm\(^2\) for all the gamma sources considered. In the case of cesium-137, this suggests the use of up to 1 cm thickness of compressed cesium chloride source material. The results of these calculations are shown in Table III.

It may be seen from Table III that the dose uniformity for applications similar to the HIFI program is expected to be quite good (\( U \lesssim 1.25 \)) and essentially independent of the gamma source material chosen. The values of the geometry factor \( \bar{G} \) at half-depth in the absorber are also indicated to be nearly equal and to a first approximation can be taken as unity for sources of very large lateral dimensions. For such sources the irradiator efficiencies are also seen to be in the 67 percent region previously mentioned as necessary for economic attractiveness.

E. **Finite Plaque Sources**

Among the factors which have been examined analytically are the effects of finite source height \( H_s \) and length \( L_s \), both on the geometry
**TABLE III**

APPROXIMATE BEHAVIOR OF INFINITE PLAQUE GAMMA IRRADIATORS FOR 6 INCH THICK WATER SLAB ABSORBERS

(Source assembly thickness \(b_s\) chosen equal to 1 inch water equivalent for uncollided source photons)

<table>
<thead>
<tr>
<th>Gamma Energy</th>
<th>(E_o) - Mev</th>
<th>0.40</th>
<th>0.66</th>
<th>1.25</th>
<th>2.76</th>
</tr>
</thead>
<tbody>
<tr>
<td>One pass on</td>
<td>(G')</td>
<td>1.19</td>
<td>1.03</td>
<td>1.02</td>
<td>1.16</td>
</tr>
<tr>
<td>either side</td>
<td>(U)</td>
<td>1.16</td>
<td>1.25</td>
<td>1.24</td>
<td>1.22</td>
</tr>
<tr>
<td></td>
<td>(F)</td>
<td>0.60</td>
<td>0.52</td>
<td>0.46</td>
<td>0.37</td>
</tr>
<tr>
<td>Two passes on</td>
<td>(G')</td>
<td>1.42</td>
<td>1.34</td>
<td>1.41</td>
<td>1.76</td>
</tr>
<tr>
<td>either side</td>
<td>(U)</td>
<td>1.20</td>
<td>1.23</td>
<td>1.20</td>
<td>1.20</td>
</tr>
<tr>
<td></td>
<td>(F)</td>
<td>0.71</td>
<td>0.67</td>
<td>0.64</td>
<td>0.56</td>
</tr>
</tbody>
</table>

(Geometry Factor \(G'\))

(Depth Dose Uniformity \(U\))

(Fractional Efficiency \(F\))
factor $G'$ and also on the parameters $(H_t/H_s)$ and $(L_t/L_s)$ which occur in the irradiator efficiency expression (12). The situation may also be examined in terms of the correction factor $K$ required for $G'$ on the source central axis, and overlap factors $(H_t/H_s)$ and $(L_t/L_s)$ as determined by the off-axis variation of $G'$ as one approaches the fringing field of the source.

As shown in Table IV, approximate correction factors for the fringing field can be derived on the assumption that, at a given depth, the energy absorption is uniform except within one relaxation length $(1/\mu)$ in the absorbing medium. Thus for uniform dose rate in the vertical direction, a source overlap of $1/\mu$ is indicated above and below the irradiated material, i.e., $H_s \approx H_t + 2/\mu$.

The central axis correction factor $K$ in Table IV is based on the assumption of a simple asymptotic approach to unity when the source dimensions $H_s$ and $L_s$ are a large number of relaxation lengths of absorber. Under the above assumptions, the need for relatively large source dimensions, particularly for high energy gamma sources, is strongly indicated in Table IV. In the case of cesium-137, lateral dimensions of the order of four feet are indicated for good efficiency, dose rate and dose uniformity.

F. Effect of Source Thickness and Filters

Since the build up factors in relations (14) to (19) are dependent on atomic number as well as photon energy, estimates have been made of the effects caused by varying the source assembly composition.

Table V lists the results derived for $G'$, governing the dose rate and final dose at half-depth in a 6 inch water absorber, and for the depth dose uniformity $U = \frac{[G'(0) + G'(t)]}{2G'(t/2)}$. The estimates, based on relation (11), were made for two limiting types of composition...
### TABLE IV

APPROXIMATE DEPTH DOSE AND EDGE EFFECT CORRECTION FACTORS FOR LONG PLAQUE SOURCES OF HEIGHT $H_s$  

<table>
<thead>
<tr>
<th>$E_0$ - Mev</th>
<th>0.40</th>
<th>0.66</th>
<th>1.25</th>
<th>2.76</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1/\mu$ - inches $H_2O$</td>
<td>3.6</td>
<td>4.6</td>
<td>6.25</td>
<td>9.6</td>
</tr>
</tbody>
</table>

| $H_s = 72$ inches |  
|------------------|---|---|---|---|
| $H_t/H_s$ | 0.90 | 0.87 | 0.83 | 0.73 |

|  
|------------------|---|---|---|---|
| $H_s = 48$ inches |  
|------------------|---|---|---|---|
| $H_t/H_s$ | 0.85 | 0.81 | 0.74 | 0.60 |

|  
|------------------|---|---|---|---|
| $H_s = 24$ inches |  
|------------------|---|---|---|---|
| $H_t/H_s$ | 0.70 | 0.62 | 0.48 | 0.20 |

Based on:

$$\bar{K} = 1 - 1/\mu H_s \text{ when } \bar{\mu} L_s \gg 1;$$

$$H_s = H_t + 2/\mu$$
TABLE V

APPROXIMATE DOSE RATE VARIATIONS FOR A LARGE AREA CESIUM-137 PLAQUE SOURCE AND 6 INCH WATER ABSORBER

<table>
<thead>
<tr>
<th>Case</th>
<th>$b_g$</th>
<th>$\mu_g h$</th>
<th>$h$ (gm/cm²)</th>
<th>$G'(t/2)$</th>
<th>$U$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.15</td>
<td>0.10</td>
<td>1.37</td>
<td>1.10</td>
<td>1.26</td>
</tr>
<tr>
<td></td>
<td>0.20</td>
<td>0.20</td>
<td>2.75</td>
<td>1.05</td>
<td>1.24</td>
</tr>
<tr>
<td></td>
<td>0.25</td>
<td>0.30</td>
<td>4.1</td>
<td>1.00</td>
<td>1.21</td>
</tr>
<tr>
<td></td>
<td>0.35</td>
<td>0.50</td>
<td>6.9</td>
<td>1.00</td>
<td>1.00</td>
</tr>
</tbody>
</table>

Case B

<table>
<thead>
<tr>
<th>$G'(t/2)$</th>
<th>$U$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>1.13</td>
</tr>
<tr>
<td>0.90</td>
<td>1.10</td>
</tr>
<tr>
<td>0.80</td>
<td>1.05</td>
</tr>
<tr>
<td>0.75</td>
<td>1.00</td>
</tr>
</tbody>
</table>
for cesium-137 source assemblies; case A for a dilute source in a low Z source mixture and cladding material, and case B for high Z (lead equivalent) source and cladding combination. It was also assumed that the source cladding thickness $b_c = 0.10$ relaxation lengths, such that the source attenuation thickness $\mu_s h = 2(b_s - 0.10)$.

The dose uniformity figures in Table V indicate that relatively small values of $b_s^1$, with cesium-137 source thickness of several gm/cm$^2$, should remove most of the depth dose non-uniformity if large area sources are employed. Under these conditions $U < 1.25$ and $G \approx 1.0$, as deduced previously. The values in Table V also suggest improved uniformity if the source assembly contains high Z materials, i.e., more uniform depth dose variations are expected with lead equivalent filters in case B than for water equivalent filters in case A, but at the expense of lower dose rate and efficiency.

IV. EXPERIMENTAL STUDIES

A. Experimental Arrangement

1. Sources

Guided by the analytical studies, the main experimental source developed for this program has dimensions 40 inches x 40 inches x 1/2 inch in thickness. It was composed of four panels, each a brass tray 20 inches x 20 inches in area, 1/2 inch in depth and 1/16 inch wall thickness, containing $0.52 \pm 0.03$ mc cesium-137 in aqueous solution. The solution source strength was determined by a comparison of gamma ray spectrometer photopeak intensities of a prepared sample and a calibrated cesium-137 solution of comparable strength.
The expected dose rate at any point in a water absorber due to this source is, from relations (3), (9) and (10),

\[ D = \frac{S_a G}{\alpha} = 2\pi h S_m G' = 4.0 \text{ G' mrad/hr} \]  

(20)

where \( G' \) is the geometry factor discussed previously.

A second cesium-137 source, approximately 5 mc strength, was employed for calibration of the dose rate probes. After dilution, this solution also provided calibration samples for scintillation counting.

Tests were also made of metallurgical techniques for preparing thin flat source elements suitable for high level cesium-137 irradiators. Barium chloride samples have been sealed in copper, low-carbon steel and stainless steel tubes and flattened by cold-rolling.

The following example illustrates the results achieved. A 15 inch length of type 304 stainless steel tube, 2-1/4 inches O.D. and 1/16 inch wall, was first partially flattened and then loose-filled with barium chloride. After "Heliarc" welding, the tube was cold-rolled to 1/2 inch thick by 3-3/8 inches wide, with a resultant reduction of 23 percent in wall thickness and an increase in barium chloride density from 1.6 to 2.99 g/cc. Further rolling resulted in a weld rupture due to the high pressure exerted by the salt. While this technique appeared promising, it was discontinued in view of possibly simpler methods of producing compressed cesium chloride pellets for later encapsulation in pipes or trays. The latter methods are presently under investigation at the Oak Ridge National Laboratories.

2. Detectors

Two scintillation probes were constructed with small phototubes
and crystals in order to obtain good spatial resolution of the radiation flux distribution.

The first probe employed a 3/4 inch six stage phototube, DuMont Type K 1303, with 4 inch long lucite light pipe, and a 10 mm diameter by 5 mm thick anthracene crystal, all covered with waterproof, light-tight tape. The current was measured with a Keithley Micro-microammeter. An output stage was added which provided means for cancelling the phototube dark current, as well as providing a further gain in sensitivity up to a factor of 30. Still further gain in sensitivity was obtained by cooling the probe with running tap water. At 1200 volts, the photomultiplier dark current was reduced to approximately $2 \times 10^{-10}$ amperes by cooling.

The second detector employed a ten stage 1-1/4 inch phototube, type 6467 with lucite light pipe and 0.12 inch aluminum waterproof cover. For dose rate measurements, an anthracene crystal was used, 1-1/4 inch diameter and 1/4 inch thick, together with the current measuring system described above.* The same probe, with a small sodium iodide crystal, was used for examining the photon spectral distribution, feeding into an RCL 256 channel pulse height analyzer.

B. Dose Rate Measurements on Plaque Sources

Figures 1 to 4 show the dose rate observations made on the central axis of our cesium-137 plaque sources and water absorbers. In this set of tests the source panels were placed horizontally on a concrete floor and a steel tank, 20 inches by 20 inches in area, 12 inches deep and 1/16

* The dark current here was $\sim 10^{-9}$ amps, but due to the higher phototube sensitivity it was found that dose rates in the range of 1 mrad/hr could be read with either probe without difficulty.
Fig. 1 Depth dose variation in water for three cesium-137 plaque sources with dimensions 20 inches x 20 inches, 20 inches x 60 inches and 40 inches x 40 inches, and calculated variation for an infinite plane isotropic source.
Fig. 2 Depth dose variation in water with 40 inches x 40 inches plaque cesium-137 source, showing effect of water absorber thickness.
Fig. 3  Depth dose variation in water due to 40 inches x 40 inches plaque cesium-137 source, showing effects of composition of the filter placed between source and water absorber. All filters, iron, cadmium and lead, were 1/16 inch thickness sheets.
Fig. 4 Geometry factor $G'$ derived for experimental curves A and D of Fig. 3. Calculated curves for the uncollided flux contribution ($E_1(b')$) and total flux component ($G'(b')$) are shown for an infinite plane isotropic cesium-137 source in water.
inch wall thickness, was placed directly on the sources, with no air gaps. Paraffin blocks surrounded the water tank to simulate a large water absorber area.

Figure 1 shows how the absolute dose rate values and uniformity of depth dose both vary as the source dimensions are increased. Figure 2 shows that finite absorber thickness has only a secondary effect on the depth dose variation when the plaque source dimensions are large. Figure 3 illustrates the nearly exponential depth dose variation, as in Figs. 1 and 2, whose initial steep gradient does indeed become modified by the thickness and atomic composition of the source assembly.

In Fig. 4 the calculated curves are for infinite plaque sources; the curve \( E_1(b') \) represents the uncollided photon component; the total component \( G'(b') \) was calculated using the Moment Method results with relation (15). Almost identical results were obtained by using the simpler relations (17) and (19). The experimental curves A and D were derived from those in Fig. 3. By inspection we find these results in at least qualitative derivations with several of the finite source correction factors, as will be brought out in Table VI.

Figures 5 to 7 show the results of extensive depth dose measurements with the 40 inches x 40 inches plaque source and paraffin absorber. In this case the source assembly and paraffin absorbers were placed vertically. The paraffin was composed of 5/8 inch thick blocks in the region of the detector and 1-3/4 inch thick blocks elsewhere to form a slab 7 inches thick, 60 inches high and 90 inches long, the paraffin density being 0.88 gm/cm\(^3\).

The influence of air gaps between source and absorber in Fig. 5, of steel absorbers in Fig. 6, and of composite filters in Fig. 7, again
Fig. 5 Depth dose variation in paraffin with 40 inches x 40 inches plaque cesium-137 source, 1/16 inch steel filter, showing effect of air gap between source and absorbers.
Fig. 6  Depth dose variation in paraffin with 40 inches x 40 inches plaque cesium-137 source, with 2 inch air gap, showing filtration effect of steel absorbers.
Fig. 7 Depth dose variation in paraffin with 40 inches x 40 inches plaque cesium-137 source, with 2 inch air gap, showing filtration effects of composite filters between source and absorber.
provide both supporting and supplemental data for comparison with our theoretical expectations, and help directly for quantitative design of high level cesium-137 irradiators.

Off-axis depth dose variations were also examined, both for water and paraffin. Figure 8 illustrates two sets of results obtained with paraffin absorbers (the curves for water absorbers were very similar). It may be noted that the field drops off rapidly near the edge of the source, and as expected is approximately constant up to one relaxation length (about 13.5 cm or 5.2 inches for paraffin and cesium-137 gammas) from the edge of the source. Another feature of interest in Fig. 8 is that the presence of absorber B produces an increased dose rate at very shallow penetration, i.e., it acts as a reflector, while at greater depths it acts as a net absorber.

C. Photon Spectral Measurements

Since the complications in the design and interpretation of large gamma irradiators are so dependent on the scattered radiation components, it was thought desirable to examine the photon spectral distribution directly. From preliminary tests it was established that thin NaI crystals, up to 1/2 inch in thickness and 1-1/2 inches diameter, showed little Compton folding when irradiated with a small cesium-137 source in lucite. Under these conditions the photopeak due to 662 Kev cesium-137 gammas and barium K x-rays at 32 Kev stood out very clearly, with a negligibly small Compton background. By inspection of spectral intensity curves derived in the NBS-NDA Moment Method program, and shown by Rockwell, Goldstein and others, it was expected that the multiply-scattered radiation should show a pronounced peak in the region of 60 Kev for appreciable
Fig. 8 Off-axis dose rate variations at three depths in paraffin slab absorber with 40 inches x 40 inches - cesium-137 plaque source. In the set shown with open circles (o), the absorbers A and B were present on both sides of the detector. In the set shown crossed (x), only absorber A was present.
depths in water and paraffin absorbers.

In Fig. 9 the dashed curve shows the observed spectrum (without correction for variations in counting efficiency with photon energy) with the 40 inches x 40 inches cesium-137 plaque source before the paraffin absorber was introduced. Scattered radiation, peaking in the region of 200 Kev shows up very clearly. This is consistent with expectations of primarily single and plural scattering in the source assembly when no absorber is present. With the paraffin absorber present, the scattered radiation is seen to go through a transition, finally peaking at about 70 Kev. The 662 Kev photopeak is seen to decrease progressively. (The decrease with paraffin depth was found to be not as rapid as the exponential integral $E_1(b)$, probably due chiefly to the projected crystal area varying with distance from the source rather than changes in source solid angle.)

A similar set of spectral profiles is shown in Fig. 10. In this case the absorber was water in the steel tank placed directly on the horizontal plaque source. Again it is evident that degraded photons in the region of 70 Kev are dominant contributors to the radiation flux within the absorber.

It was found that inspection of the spectral profiles was also a convenient way to examine the effects of absorbers and filters between the source and main slab absorber. Figure 11 shows a set of such profiles, taken at the front surface of the paraffin absorber, when the air gap was set at 2 inches. It is worth noting that with lead filters there is a pronounced peaking at 75 Kev, due to the lead fluorescent K x-ray, and that this occurs at practically the same photon energy as the multiply-Compton scattered flux at deeper penetration.
Fig. 9 Observed photon spectral distribution at several depths in a paraffin absorber irradiated by a 40 inches x 40 inches cesium-137 plaque source. The dashed curve illustrates the observed spectrum in the absence of the absorber.
Fig. 10 Observed photon spectral distributions at several depths in water, irradiated by a 40 inches x 40 inches cesium-137 plaque source.
Fig. 11 Spectral profiles at front surface of paraffin absorber, showing the effects of various filters in the source assembly.
V. INTERPRETATION AND CONCLUSIONS

Quantitative results of both the experiments and calculations for water absorbers are illustrated in Table VI. The first three source arrangements (derived from Fig. 1) bring out the gradual increase in dose rate and improvement in dose uniformity as the lateral source dimensions are increased, (the source activity per unit area being kept constant). In the four tests A, B, C and D with the 40 inches x 40 inches source (based on Fig. 3), the increase in source assembly thickness, \( b_s \), by means of cadmium and lead filters is seen to bring about further improvements in depth dose uniformity but at some expense in dose rate.

The calculated performance of the above test source arrangements, as indicated at the bottom of Table VI, assumed negligible corrections for the uncollided flux component, i.e., \( K_0 \approx 1.0 \), and two separable components for the scattered radiation correction factor \( K_s = K_1 K_2 \). Here \( K_1 \) is an estimate of the effect of lateral source dimensions \( H_s \) and \( L_s \), and \( K_2 \) is designed to take into account the reduction in scattering due to high atomic number filter (or source) materials. The \( (s/\mu_a) \) values were estimated from Table I.

As seen in Table VI, the calculated values of the geometry factor \( G' \) and dose rate \( D \) are generally slightly lower than those observed, but within our estimated experimental uncertainty of 10 percent for the absolute absorbed dose rates in water. The observed depth dose uniformity values are seen to range from 1.54 to 1.11, whereas the calculated values show the same trend but are more conservative.

Based on Fig. 8, which indicates that the off-axis radiation field is sensibly constant to within one mean free path \( 1/\bar{\mu} \approx 4.6'' \) in water.
### TABLE VI

**RESULTS FROM CESIUM-137 PLAQUE SOURCES AND SIX INCH WATER ABSORBERS**

<table>
<thead>
<tr>
<th>Test Source</th>
<th>b'</th>
<th>( \overline{D} )</th>
<th>( \overline{G} )</th>
<th>Ud</th>
<th>( \overline{G} )</th>
<th>( \overline{D} )</th>
<th>Ud</th>
<th>( F_1 )</th>
<th>( (F_1)_{max} )</th>
<th>( (F_2)_{max} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>20&quot; x 20&quot;</td>
<td>0.24</td>
<td>3.1</td>
<td>0.76</td>
<td>1.54</td>
<td>0.82</td>
<td>3.3</td>
<td>1.57</td>
<td>0.22</td>
<td>0.38</td>
<td>0.48</td>
</tr>
<tr>
<td>20&quot; x 60&quot;</td>
<td>0.24</td>
<td>3.6</td>
<td>0.90</td>
<td>1.45</td>
<td>0.94</td>
<td>3.7</td>
<td>1.54</td>
<td>0.25</td>
<td>0.45</td>
<td>0.57</td>
</tr>
<tr>
<td>40&quot; x 40&quot; A</td>
<td>0.24</td>
<td>4.2</td>
<td>1.05</td>
<td>1.33</td>
<td>0.99</td>
<td>4.0</td>
<td>1.50</td>
<td>0.38</td>
<td>0.52</td>
<td>0.66</td>
</tr>
<tr>
<td>40&quot; x 40&quot; B</td>
<td>0.36</td>
<td>3.6</td>
<td>0.90</td>
<td>1.21</td>
<td>0.85</td>
<td>3.4</td>
<td>1.38</td>
<td>0.33</td>
<td>0.45</td>
<td>0.57</td>
</tr>
<tr>
<td>40&quot; x 40&quot; C</td>
<td>0.45</td>
<td>3.0</td>
<td>0.75</td>
<td>1.23</td>
<td>0.72</td>
<td>2.9</td>
<td>1.31</td>
<td>0.28</td>
<td>0.37</td>
<td>0.47</td>
</tr>
<tr>
<td>40&quot; x 40&quot; D</td>
<td>0.58</td>
<td>2.8</td>
<td>0.70</td>
<td>1.11</td>
<td>0.62</td>
<td>2.5</td>
<td>1.27</td>
<td>0.24</td>
<td>0.35</td>
<td>0.44</td>
</tr>
</tbody>
</table>

**Calculations Based On:**

\[
G'(b') = E_1(b') + K_\xi K_\beta e^{-b'}
\]

\[
b' = b + \mu q ; b = u_b h/2 + \mu_c t_c + \mu_f t_f
\]

\[
\beta = (A_1 a_1 + A_2 a_2) = 2.34 \text{ for cesium-137 and water}
\]

\[
K_\xi = (1 - 1/\mu_H_b)(1 - 1/\mu L_b)
\]

\[
K_\beta = \frac{1 + 2 \Sigma (G_j/\mu_c) b_i}{1 + 2 \Sigma (G_j/\mu_c) b' i}
\]

\[
F_1 = \mu_a \overline{G'} (1 - 2/\mu_H_b) \quad (F_1)_{max} = \mu_a \overline{G'} \overline{abs} \quad (F_2)_{max} = (1 + e^{-\mu_f}) (F_1)_{max}
\]
or 5.2" in paraffin) of the edge of a large uniform plaque source, the efficiency values $F_1$ in Table VI were calculated under the condition of uniform dose in the vertical direction by means of source overlap ($H_s \approx H_t + 2/\mu$). The column headed $(F_1)_{\text{max}}$ was based on the experimentally determined value of $G'$, from $\bar{D} = 2\pi k_a h S_m G'$, assuming sufficient package overlap to integrate the fringing field. The column marked $(F_2)_{\text{max}}$ refers to the same conditions, with the addition of a second pass of the six inch water absorber on either side of the source.

No direct experiments were made in this study on the radiation from thick cesium chloride source material. However, there are strong indications from theory that the resultant behavior of plaque sources with increasing source thickness $h$ should be very similar to our observation in Fig. 3 and Table VI on thick source assemblies using high atomic number filters. Thus, to a first approximation curve $D$, with 1/2 inch of aqueous cesium-137 source followed by brass, lead, cadmium and steel absorbers each 1/16 inch in thickness, has a total source assembly attenuation length of $b_s \approx 0.58$. As far as the uncollided flux is concerned this should be very nearly equivalent to a cesium chloride source with a thickness $h$ and cladding of 1/16 inch stainless steel, such that $\mu_s h \sim 1$ mean free path, or $h \sim 1/\mu_s \sim 13.5 \text{ gm/cm}^2$, or $\sim 4.5 \text{ cm, } \sim 1.8 \text{ inches cesium chloride}$. This approximates the condition of cesium chloride source material in 2 inch diameter pipes. As for the scattered radiation, this combination of lead, cadmium and steel leads to comparable estimates of the composition correction factor, $K_z$, as for the cesium chloride, when $\sigma_s/\mu_s \approx 1.0$ is used for the latter.
Using the values shown in Section II for the radiation properties of available cesium-137 source material, the above considerations lead to expected dose rates for high level irradiators and six inch water equivalent slab absorbers, in the form

$$D = 2\pi k_a h S_m \bar{G}^t = 0.42 h \bar{G}^t \text{ Megarads/hour}.$$ 

For a thick source assembly, comparable to curve D of Fig. 3 and Table VI, dose rates up to 5 Megarads/hour can then be expected, together with very good dose uniformity. In such a single pass irradiator the efficiency is unfortunately quite low, in the region of 20 percent as indicated by Table VI.

Because of the economic considerations which suggest the need for high irradiator efficiency, high level cesium chloride source thicknesses of $h \sim 4 \text{ gm/cm}^2$, with 1/16 inch steel cladding ($b_s \sim 0.25$), are indicated from this study, together with source lateral dimensions $H_s$ and $L_s$ which are a large multiple of the mean free path ($1/\mu$). Under such conditions we estimate from the data in Table VI that $\bar{G}^t \sim 1.0$ for 6 inch water absorber, so that the dose rate $D \sim 1.5 \text{ Megarads/hour}$, the depth dose uniformity $U_d \sim 1.25$, and efficiencies of over 50 percent may be achieved with several absorber passes in a simple one plaque irradiator. To a close approximation, very similar performance is to be expected from $\sim 3 \text{ gm/cm}^2$ of cobalt-60 which has a specific activity in the region of 5 curies/gm.

The question of lateral source dimensions is perhaps the most critical design feature for high level gamma irradiators. The indications from this study are that the source height $\mu H_s$ and length $\mu L_s$, when
expressed in mean free paths for the absorber, need to approach 10 mean free paths in order that the scattered radiation builds up to nearly its infinite source value, and that the off-axis field remains sensibly constant over the absorber.

From Table II it may be found that the mean free path in water, $1/\bar{\mu}$, for iridium-192, cesium-137, cobalt-60 and sodium-24 are approximately 3.6, 4.6, 6.3 and 8.0 inches, respectively. For comparable performances to the 40 inches square cesium-137 sources used in this study, the corresponding source dimensions for iridium-192 would be only 31 inches, for cobalt-60 it would be 46 inches, and for sodium-24 the figure is 69 inches. Thus the lower photon energy sources should have considerable advantages in compactness and shielding. The situation is particularly relevant to the design of irradiators for low density materials such as medical supplies. If the effective material density is $\sim 0.25$ gm/cm$^3$, the indicated source dimensions for good dose rate, dose uniformity and irradiator efficiency are then indicated to be $\sim 10$ feet for iridium-192, $\sim 13$ feet for cesium-137, $\sim 16$ feet for cobalt-60 and $\sim 23$ feet for sodium-24 plaque source irradiators.

These calculations and experiments indicate that high efficiency designs of irradiators utilizing cesium-137 are realizable. Further it can be seen that such irradiators would be competitive with cobalt-60 units for a cobalt to cesium activity cost ratio of 3:1 with the advantage of minimum size and minimum shielding for a cesium irradiator.

Further, it would appear that the calculational approach developed in the course of this program permits extrapolation to the engineering design of plaque irradiators. The precepts outlined in this report
on cladding, source size and dose calculation should carry over to other source configurations with suitable consideration of the effects of geometric transformation.

If projected costs for large amounts of cesium-137 can be expected to approach 25 cents per curie we feel that it affords attractive advantages as a high level irradiator. If such costs are ultimately feasible additional studies should be undertaken to elucidate the features and efficiencies of other geometrical source configurations such as cylinders, hollow cylinders, etc., which appear attractive for chemical processing applications.
REFERENCES


