THE MEASUREMENT OF BETA RADIATION DOSAGE WITH PHOTOGRAPHIC EMULSIONS

by

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Certified by

Thesis Supervisor

Chairman, Departmental Committee on Graduate Students
ABSTRACT

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The purpose of this thesis has been to investigate the feasibility of quantitative beta radiation dosage measurement with photographic emulsions, a technique expected to excel existing methods for high resolution dosimetry problems. The survey has proceeded along two complementary lines: First, I have studied the response of various films to mono-kinetic electrons, from a \( \beta \) ray spectrometer, covering the energy range 0.03 to 1.8 Mev. These measurements have given fundamental information regarding the dependence, on electron energy, of various basic film properties. Among these were the shape of the density vs. exposure curve and the absolute sensitivity for electrons striking the emulsion at several different angles of incidence. Second, I have investigated the response of these same films to continuous spectrum \( \beta \) rays, of graded maximum energies, from sources whose surface dosage rates had been accurately determined in an extrapolation ionization chamber. This approach yielded fundamental data regarding the effect of absorbers on the exposing efficiency of the continuous \( \beta \) ray spectrum, together with practical information of value in direct applications of the photographic dosimetry technique. From these two different types of measurement the general conclusion has been drawn that, with intelligent use, photographic emulsions will constitute a valuable addition to existing instrumentation for the measurement of \( \beta \) radiation dosage. Finally, two applications have been described which demonstrate the usefulness of this technique in problems of current medical interest.
ACKNOWLEDGEMENTS

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Others who have given scientific advice and assistance are innumerable. Dr. B.M. Dobyns, Profs. J.W. Irvine, M. Deutsch, and S.C. Brown, Dr. Van Dilla, Mr. Maletskos, Mr. Perry, and the entire Radioactivity Center staff, to mention only a few, would all find ideas of their own within this thesis.

Finally, three individuals have made special contributions. Dr. G.L. Brownell carried out a part of the development of the apparatus in connection with his own thesis; Dr. Randall S. Caswell was very generous in sharing his own extrapolation chamber and \(4\pi\) counter; and Miss Virginia White, legally or illegally, has been of invaluable assistance in making many of the measurements.
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CHAPTER 1: INTRODUCTION

The purpose of this investigation has been the evaluation of photographic film as an instrument for measuring the radiation dosage, or energy dissipation per unit mass of matter, resulting from the absorption of β rays.

The idea of using film for this dosimetry problem arose from the belief that film would be capable of measuring β radiation dosage over a smaller area than any other instrument; that is, it has high resolution. The need for high resolution β ray dosimetry grew out of a specific problem which confronted the Radioactivity Center in one of its cooperative research undertakings. A group of food technologists wished to use radioactive Ca\(^{45}\) as a tracer in studying certain phases of calcium metabolism in humans. The experiment was sound in principle, but could not be carried out immediately because of the possible radiation hazard to the subject of the experiment. The Ca\(^{45}\) tracer would be deposited in the bone, thereby endangering, if present in sufficient concentration, the blood forming organs and other adjacent tissues. At that time, the Atomic Energy Commission (A-2) specified that Ca\(^{45}\) could not be administered to humans unless it were reliably established that the radiation dosage rate would at no point in the body exceed 0.1 rep/day. One of the problems which the Radioactivity Center therefore set out to solve was the estimation of the maximum radiation dosage rate to which any point in the bone would be subjected after a given quantity of Ca\(^{45}\) was absorbed into the blood stream. As a step toward this estimation, it was decided actually to measure this dosage rate in the bones of dogs. Because it was known at least qualitatively from autoradiographic studies that the concentration of Ca\(^{45}\) varied markedly over small regions of bone, it was necessary to develop a technique capable of measuring dosage rates over a fraction of a
square millimeter. Quantitative autoradiographic measurements with film appeared to be the best method, and were consequently undertaken.

The research problem seemed worthy of detailed study not only to meet the needs of the calcium metabolism investigation, but also to provide a more generally applicable dosimetry tool in a field of rapidly expanding biological and medical interest: radioactive tracer studies and radiation therapy. The technique held promise of usefulness chiefly in evaluating the dosage rate from isotopes in tissue, such as Ca$^{45}$ in bone, or I$^{131}$ in thyroid. However, it could conceivably meet special needs also in such other problems as the dosimetry of external radiation, for example, β rays from β ray applicators, or electrons from high energy machines; both of these are currently finding increasing therapeutic use. The attempt to apply and evaluate the use of film as a dosimeter of β rays in certain of these problems has led to the research here reported.

The plan of procedure of this thesis is the following: We first consider in some detail the interaction of β rays with matter, in order to make clear the nature of β ray radiation dosage, to permit an understanding of the photographic effect of β rays, and to aid in the interpretation of experimental measurements later reported. Then, a brief survey is given of the various experimental methods of measuring dosage, to set the photographic method in its proper context. This is followed by a study of the general photographic process, whose basic principles lie at the roots of the succeeding investigations. We then proceed to an analysis of the more specific principles involved in the photographic effect of β rays, with the intention of delineating more clearly just what experimental knowledge is required for practical β ray dosage measurements with film. Having established this background, we survey the
present state of experimental knowledge on the detection of \( \beta \) rays with film, and follow this with a presentation of the experimental results obtained during this research. (Many of the details of the present experiments are assigned to appendices, in order not to interrupt too seriously the central chain of development of this study.) From these findings, conclusions are drawn as to the accuracy and practicality of \( \beta \) ray dosimetry with film. Finally, two specific applications are described, one of which is the biological problem which gave rise to this thesis.
CHAPTER 2: THE INTERACTION OF BETA RAYS WITH MATTER

Beta ray radiation dosage is defined as the energy absorbed, per unit mass of matter, from β rays traversing that matter. It is expressed, preferably, in absolute units of ergs/gm (I2), although other units, such as the rep (usually 93 ergs/gm), have lately found widespread use. Before attempting to study the measurement of β ray dosage, it is essential to understand both the nature of dosage and the nature of the effects of β rays on a variety of detecting instruments. For that reason, it has seemed advisable to begin with a somewhat detailed survey of the interaction of β rays with matter. The reader who is well acquainted with the subject might well use this chapter only as reference material at certain points later in the thesis. The reader who is not familiar with the subject would do well to study this section, perhaps having to accept on faith the fact that most of the material discussed will appear again in a more specific application further on.

Regarding terminology, it should be mentioned here that I have used the terms "β ray" and "electron" almost interchangeably, although technically β ray applies only to an electron originating in an atomic nucleus. Except when attention is focussed on the characteristic continuous energy spectrum of β rays, the distinction is of no significance, and statements made of β rays are equally true of electrons. Usually "electron" has been used as the more general term.

A. Scattering of Electrons by Matter

As electrons pass through matter, they lose kinetic energy to the matter and are scattered by the matter.
In this section, scattering is examined independently of energy loss, although in practice the two can never be completely separated.

1. Single scattering

The theory of elastic single scattering events caused by atomic nuclei was worked out on quantum mechanical principles by Mott (M8). For slow electrons it reduces to the classical Rutherford theory which shows that scattering varies as $Z^2/v^4$, where $Z$ is the atomic number of the nucleus and $v$ is the electron velocity. The predictions are not identical for negative electrons and positrons, negatrons being somewhat more highly scattered than positrons. At low $Z$ the difference is small (e.g., about 8% for 1 Mev particles in polystyrene), but at high $Z$ a considerable discrepancy exists (e.g., a factor of approximately 3 for 1 or 2 Mev particles in Pt).

For negatrons of 1 to 2 Mev, Mott's formula has recently received rather accurate experimental verification (B-24) over an atomic number range between Be and Au. Positron scattering experiments have not been carried out with equal accuracy, although results (L-6) in the energy region of 1 Mev are in moderately good agreement with theory. Despite the reasonably sound theoretical and experimental understanding of scattering, the single scattering formula is not of great value in practical applications because repeated scattering of the same electron takes place in all but the thinnest foils.

2. Multiple scattering

The next most elementary case is that known as multiple scattering. This case can be represented by electrons passing through a foil sufficiently thick as to produce many scattering events (most of which are small: less
than a few degrees), but sufficiently thin that the total deviation of most of the electrons is less than $10^\circ$ to $20^\circ$. An understanding of multiple scattering gives a more accurate picture of scattering in practical applications of $\beta$ ray detection and measurement, and is therefore considered here in more detail.

In Fig. 1, a beam of monokinetic electrons is incident perpendicularly on a foil, and a particular electron emerges on the other side scattered by an angle $\Theta$.

![Diagram](image)

The angular distribution of the emergent beam is characterized by the fraction $dF$ of the electrons contained in a solid angle segment $dw$ at angle $\Theta$ with the normal, i.e., by $\frac{dF(\Theta)}{dw}$. Small angle multiple scattering has been treated theoretically by several investigators. The following formulas are taken from a summary by Rossi and Greisen (R-1). These men express the angular distribution in the form

$$\frac{dG(\Theta_y)}{d\Theta_y} = k e^{-f(T,Z,x) \Theta_y^2}.$$  

$\Theta_y$ is the angle $\Theta$ projected onto a particular plane containing the incident beam (e.g., the plane of the paper in Fig. 1). $dG(\Theta)$, then, is the probability that an electron will emerge at some angle $\Theta$, which, when projected onto the
plane of the paper, lies between $\Theta_y$ and $\Theta_y + d\Theta_y$ degrees. When $f(T, Z, x)$ is put into workable form by reducing numerical constants, one finds that

$$f(T, Z, x) = \frac{T(T + 1) e^2 A}{2.01 \ln(183Z^{-1/3})Z^2 x}.$$

In this formula $T =$ kinetic energy of electron in Mev, $\beta =$ ratio of electron velocity to velocity of light, $A =$ atomic weight of element, $Z =$ atomic number of element, $x =$ foil thickness in mg/cm$^2$. It can be shown, if one wants the angular distribution in terms of $\Theta$ instead of $\Theta_y$, that

$$dP(\Theta) = k_2 \Theta e^{-f(T, Z, x) \Theta^2} d\Theta,$$

where $dP(\Theta)$ is the probability that the electron will emerge at an angle between $\Theta$ and $\Theta + d\Theta$. To get the fraction of electrons per solid angle segment, one divides by

$$dw = 2\pi \sin \Theta d\Theta \approx 2\pi \Theta d\Theta.$$  

Hence

$$\frac{dF(\Theta)}{dw} = k_2 e^{-f(T, Z, x) \Theta^2} \quad (1).$$

In Fig. 2 are shown some cloud chamber experimental data obtained by Slawsky and Crane (8-9) on the small angle multiple scattering of 0.9 Mev electrons in aluminum. The theoretical curve is plotted from the above equation (1). It can be seen that the observed scattering is considerably less than the predicted scattering, an observation reported by others also (W-9). If the error lies in the theory rather than the experiment (which is not certain), the theory underestimates the foil thickness required for a given angular distribution by a factor of about 2. It is most accurate for thin foils giving small scattering.

On the basis of this theory, which is seen to have at least reasonable validity, I have computed for
Figure 2
SCATTERING OF 0.9MeV. ELECTRONS IN ALUMINUM
(Slowsky and Crane)
various energies the number of $\text{mg/cm}^2$ of lucite required to reduce $\frac{\text{d}F(\theta)}{\text{d}w}$ to $1/2$ of its maximum at $15^\circ$. (The maximum occurs, of course, at $0^\circ$.) These calculations, shown in Table 1, give an idea of the dependence of scattering on energy in low Z materials.

Table 1

<table>
<thead>
<tr>
<th>$T$</th>
<th>$X$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0 Mev</td>
<td>56 $\text{mg/cm}^2$</td>
</tr>
<tr>
<td>1.0</td>
<td>17</td>
</tr>
<tr>
<td>0.50</td>
<td>5.4</td>
</tr>
<tr>
<td>0.20</td>
<td>1.12</td>
</tr>
<tr>
<td>0.10</td>
<td>0.32</td>
</tr>
<tr>
<td>0.050</td>
<td>0.087</td>
</tr>
</tbody>
</table>

As is shown later, these thicknesses are equal to a small fraction of the maximum range of the electrons, so that in even low Z materials scattering is seen to be considerable.

As foil thickness is increased in multiple scattering experiments, the angular distribution of the emergent electrons takes on a characteristic form which is rather independent of thickness in a particular material. The equilibrium distribution is too complicated for theoretical treatment; however, Slawsky and Crane experimentally found it to be rather similar to, but slightly more spread out than, the third graph of Fig. 2 for 0.9 Mev electrons in aluminum.

3. Angular distribution of $\beta$ rays from plane geometry source

Another type of scattering problem which will be of interest later is the angular distribution of $\beta$ rays
from plane geometry sources of various thicknesses and under various conditions of absorption. If a small area $\Delta A$ is delineated on the source or absorber surface, the angular distribution from the source is characterized by the same function used above, $\frac{dF(\theta)}{d\omega}$, for the $\beta$ rays emerging from $\Delta A$.

For an infinitesimally thin source the $\beta$ rays are emitted isotropically in all directions, just as they emerge from the parent nuclei. Therefore $\frac{dF(\theta)}{d\omega}$ is a constant independent of $\theta$.

For a thick source, of thickness greater than the maximum range of the $\beta$ rays (discussed later), a different angular distribution pattern exists at the source surface. The situation can be understood most clearly by focusing attention first on the angular relationships of the $\beta$ rays deep inside the source.

In Fig. 3 an elementary area $\Delta A$ is inscribed on an imaginary plane at an arbitrary orientation and at a depth inside the source greater than the maximum $\beta$ ray range. For concreteness, $\Delta A$ may be considered a circle seen edge on. The angular distribution of the $\beta$ rays
passing through $\Delta A$ is again characterized by the function $\frac{dF(\theta)}{d\omega}$, where $\theta$ is the angle made between the normal to $\Delta A$ and the direction in question. At this depth in the source, the $\beta$ rays know nothing about the source surfaces; hence there can be no preferred direction of motion and the angular distribution is completely random. Consequently, for any other orientation of $\Delta A$, the angular distribution $\frac{dF(\theta)}{d\omega}$ would of necessity be identical with the $\frac{dF(\theta)}{d\omega}$ of $\Delta A$ as oriented in Fig. 3 (always being measured with respect to the normal to the $\Delta A$ in question). In particular, the perpendicular component $\frac{dF(\theta)}{d\omega}$ is the same for all orientations of $\Delta A$. Let the number of $\beta$ particles (per unit solid angle) passing through $\Delta A$ in the direction $\theta$ be represented by $\frac{dN(\theta)}{d\omega}$. $\frac{dN(\theta)}{d\omega}$ is just proportional to the product of the area of $\Delta A$ and the fraction of the $\beta$'s passing through $\Delta A$ at the angle $\theta$; that is,

$$\frac{dN(\theta)}{d\omega} = k \Delta A \frac{dF(\theta)}{d\omega}$$

(2)

Now consider the number of $\beta$'s passing through $\Delta A$ in the two directions $\theta = 0$ and $\theta = 0_1$. We have

$$\frac{dN(0)}{d\omega} = k \Delta A \frac{dF(0)}{d\omega}$$

(3)

and

$$\frac{dN(0_1)}{d\omega} = k \Delta A \frac{dF(0_1)}{d\omega}$$

(4)

Eq. (4) can be expressed in another way. The number of $\beta$'s passing through $\Delta A$ in the direction $0_1$ is the same as the number passing in the same direction through $\Delta A \cos 0_1$, the area of $\Delta A$ projected normal to the $0_1$ direction. But the number of $\beta$'s passing perpendicularly through this new area element $\Delta A \cos 0_1$ could equally well be given by an expression analogous to eq. (3), namely
\[
\frac{dN(\theta_1)}{d\omega} = k(\Delta A \cos \theta_1) \frac{dF(0)}{d\omega} \quad (5)
\]

\(\theta = 0\) here represents the same direction as before but is measured with respect to (i.e., along) the perpendicular to the new area element. As already discussed, the magnitude of \(\frac{dF(0)}{d\omega}\) is invariant with changes in orientation of the area element. We therefore have 2 expressions for the number of \(\beta\)'s passing through \(\Delta A\) in the direction \(\theta_1\):

\[
\frac{dN(\theta_1)}{d\omega} = k \Delta A \frac{dF(\theta_1)}{d\omega} = k(\Delta A \cos \theta_1) \frac{dF(0)}{d\omega}.
\]

Since this holds for any value of \(\theta_1\), we have

\[
\frac{dN(\theta)}{d\omega} = \frac{dN(0)}{d\omega} \cos \theta.
\]

In other words, the number of \(\beta\)'s (per unit solid angle) passing through \(\Delta A\) in the direction \(\theta\) is proportional to \(\cos \theta\). In the remainder of this thesis, I refer to this angular pattern as a "\(\cos \theta\) angular distribution".

We now ask what angular distribution pattern exists at a plane surface of such a thick source. Imagine the source of Fig. 3 sliced in half along the plane of \(\Delta A\) (i.e., along the dotted line), so that \(\Delta A\) is now on the source - air interface. If \(\beta\) particles travelled in straight paths (for example, like \(\alpha\) particles), the angular distribution of the particles passing upward through \(\Delta A\) would be unchanged by the removal of the top half of the source; without scattering back and forth between the two halves, the upper half could have no effect on the angular distribution of the particles entering it from below. Hence the angular distribution at a thick source surface would be rigorously \(\cos \theta\) for such particles. When scattering is involved, as it always is with \(\beta\) particles, the situation is somewhat more complicated and a rigorous solution
of the problem is not possible. However, it is entirely plausible that the angular distribution is still $\cos \Theta$. We have seen that the fundamental condition producing a $\cos \Theta$ distribution is a complete randomness of angular distribution within the source. Removing the top half of the source does change the number of $\beta$ particles passing upward through $\Delta A$, for it removes $\beta$'s which were emitted or scattered from the top half into the bottom half, and then back through $\Delta A$. However, these $\beta$ rays, which have undergone this much scattering (and started from a random distribution anyway), must have an essentially random distribution in the bottom half of the source and therefore a $\cos \Theta$ distribution as they pass back through $\Delta A$. Hence by removing the top half, one subtracts a small number of $\beta$'s having a $\cos \Theta$ distribution from a larger number of $\beta$'s having a $\cos \Theta$ distribution; this leaves a $\cos \Theta$ distribution in the $\beta$'s emerging from $\Delta A$ at the source-air interface.

The important practical effect of scattering is that it establishes a random angular distribution of the $\beta$ rays in sources of thickness even less than the maximum $\beta$ ray range. Consequently, the angular distribution of $\beta$'s from even relatively thin sources may be $\cos \Theta$. An example of this is shown in an experiment of Elliot and coworkers (E-2). They found a good approximation to a $\cos \Theta$ distribution for the $\beta$ rays ($U_2X_2$) from a uranium metal source of thickness roughly equal to 1/5 the maximum range of the $\beta$ rays. For sources still thinner, the angular distribution is intermediate between isotropic and $\cos \Theta$.

From the preceding discussion of thick sources it can perhaps be anticipated that a similar situation exists when absorbers are placed over a thin source. The combined effects of absorption and scattering will tend to produce random angular distribution of the $\beta$ rays in the absorber,
and consequently a \( \cos \theta \) distribution at the surface. Brownell (B-23) (from whom much of this angular distribution analysis is taken) has shown experimentally that this is the case. Using \( \beta \) rays, he found that a fair approximation to the \( \cos \theta \) distribution is established in 24 mg/cm\(^2\) of lucite, and a very good approximation by 90 mg/cm\(^2\). When enough absorber is present to give considerable scattering but not much absorption, the newly established (approximately \( \cos \theta \)) angular distribution contains more electrons in the forward direction than did the no absorption distribution. This is known as self focusing, and has been observed by Brownell, among others.

In the discussion thus far, the angular distribution has been characterized in terms of the number of \( \beta \)'s contained in a solid angle increment at some particular direction. Sometimes it is more useful to consider the distribution in terms of the number of \( \beta \)'s contained in a plane angle increment at that direction; that is, one asks how many \( \beta \)'s leave the source surface at an angle between \( \theta \) and \( \theta + d\theta \) with respect to the normal. This distribution has already been represented by the symbolism \( \frac{dp(\theta)}{d\theta} \). By the reverse of the transformation used previously, one has simply:

\[
\frac{dp(\theta)}{d\theta} = 2\pi \sin \theta \frac{df(\theta)}{dw}.
\]

The important aspects of the angular distribution of \( \beta \) rays from various plane geometry sources are summarized in Table 2. \( \frac{df(\theta)}{dw} \) and \( \frac{dp(\theta)}{d\theta} \) are there given normalizing factors such that the integral between \( \theta = 0 \) and \( \theta = \pi/2 \) is unity. However, the dependence of these quantities on the angle is the significant feature.

A clear comprehension, both mathematical and intuitive, of the angular distribution phenomena just dis-
cussed is essential to an understanding of the remainder of this thesis. (Such an understanding on my part would also have hastened the completion of this thesis.)

Table 2.
Summary of Angular Distribution Patterns

<table>
<thead>
<tr>
<th>Source</th>
<th>( \frac{dF(\theta)}{d\omega} )</th>
<th>( \frac{d\Phi(\theta)}{d\omega} )</th>
<th>Name for Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thin</td>
<td>( \frac{1}{2\pi} )</td>
<td>( \sin\theta )</td>
<td>Isotropic</td>
</tr>
<tr>
<td>Thick</td>
<td>( \frac{1}{\pi} \cos\theta )</td>
<td>( 2 \sin\theta \cos\theta )</td>
<td>Diffuse or ( \cos\theta )</td>
</tr>
<tr>
<td>Thin, with thick absorber</td>
<td>( \frac{1}{\pi} \cos\theta )</td>
<td>( 2 \sin\theta \cos\theta )</td>
<td>Diffuse or ( \cos\theta )</td>
</tr>
</tbody>
</table>

4. Saturation backscatter

In the preceding section we considered the angular distribution of the electrons passing through an absorber. However, some of the electrons incident on a piece of matter are scattered back through the surface which they entered. They are said to be "backscattered". When the sheet of matter becomes thick enough (roughly \( \frac{1}{5} \) the maximum range of the electrons \( E-5 \)), the backscattering no longer increases with increasing thickness, and is said to have reached "saturation." In this section saturation backscatter is discussed.

There are at least 3 questions regarding the backscattered electrons which are pertinent to the problems of this investigation: (1) What is the angular distribution of the backscattered electrons? (2) What fraction of the incident electrons are backscattered? (3) What
is the dosage contributed in the surface layer by these electrons?

The angular distribution of backscattered electrons has not been investigated in much detail. However, a certain amount of experimental data is available. Neher (N-3) gave a brief report of his measurements with normally incident, monokinetic electrons. His conclusions were that for the "heavier elements" at energies above 100 kev the cosθ law held accurately. "Small deviations" were found for carbon and beryllium, especially below 100 kev. Brownell gives the angular distribution of electrons from a thin source in contact with the backscatterer. A large fraction of the β rays striking the backscatterer in these measurements therefore were incident on the material at grazing angles. For lucite the backscattered β rays had an almost isotropic angular distribution, although the intensity dropped beyond 60°. For higher Z backscattering materials (e.g., Pt) the radiation followed the cosθ law rather closely. It therefore appears that a cosθ function is the stable angular distribution pattern for backscattered electrons, and that this distribution tends to be approached whatever the initial angular distribution of the incident electrons.

This is not surprising in view of the discussion of the preceding section. In the backscattering process, the scattering in the material tends to establish a more random angular distribution of the electrons than existed in the incident beam. Therefore, whatever the angular distribution of the incident electrons, one would expect a more nearly cosθ angular distribution of the backscattered electrons.

The fraction f of electrons backscattered has been studied by several investigators under a number of different conditions. Two well known experiments for f as a function of electron energy and Z of backscattering material are those of Schonland (S-3) and Eklund (E-1), who concerned themselves with normally incident, monokin-
etic electrons. Both found \( f \) to be substantially independent of energy in the energy range studied, but strongly dependent on \( Z \). Eklund's data, being the more complete, are shown in Table 3. Schonland's agree closely.

Table 3.

<table>
<thead>
<tr>
<th>Element</th>
<th>( Z )</th>
<th>( 50 )</th>
<th>( 100 )</th>
<th>( 200 )</th>
<th>( 300 )</th>
<th>( 370 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be</td>
<td>4</td>
<td>.03</td>
<td>.03</td>
<td>.03</td>
<td>.04</td>
<td>.05</td>
</tr>
<tr>
<td>C</td>
<td>6</td>
<td>.05</td>
<td>.06</td>
<td>.05</td>
<td>.07</td>
<td>.07</td>
</tr>
<tr>
<td>Ca</td>
<td>20</td>
<td>.19</td>
<td>.23</td>
<td>.21</td>
<td>.22</td>
<td>.23</td>
</tr>
<tr>
<td>Cu</td>
<td>29</td>
<td>.24</td>
<td>.28</td>
<td>.29</td>
<td>.30</td>
<td>.30</td>
</tr>
<tr>
<td>Mo</td>
<td>42</td>
<td>.29</td>
<td>.34</td>
<td>.36</td>
<td>.37</td>
<td>.38</td>
</tr>
<tr>
<td>Sn</td>
<td>50</td>
<td>.33</td>
<td>.37</td>
<td>.40</td>
<td>.42</td>
<td>.40</td>
</tr>
<tr>
<td>W</td>
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<td>.36</td>
<td>.44</td>
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</tbody>
</table>

Various workers have lately investigated the backscattering from thin sources of \( \beta \) ray emitting isotopes in close contact with the scattering material. Pertinent negative electron experiments have been brought together by Burtt (B-26). He and others found essentially no dependence of \( f \) on \( \beta \) ray energy between \( \text{P}^{32} \) (1.7 Mev) and \( \text{I}^{131} \) (0.6 Mev), for all \( Z \). There appears to be a 30% lower \( f \) in the case of \( \text{Co}^{60} \) (0.3 Mev), but this may be caused by window absorption of the very low energy back-scattered particles. A strong dependence of \( f \) on \( Z \) is observed. Burtt's results, which agree fairly closely with those of several other investigators, are shown in Table 4 for RaE \( \beta \) rays (1.17 Mev).

It can be seen that Burtt's values are considerably
higher than Eklund's. This is of course to be expected, because of the difference between the isotropic and perpendicular angular distributions, respectively, of the electrons incident on the backscattering material.

Table 4.
Burtt's Backscatter Measurements

<table>
<thead>
<tr>
<th>Backscattering Material</th>
<th>Z</th>
<th>f</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lucite</td>
<td>5.8</td>
<td>.15</td>
</tr>
<tr>
<td>Al</td>
<td>13</td>
<td>.29</td>
</tr>
<tr>
<td>Cu</td>
<td>29</td>
<td>.48</td>
</tr>
<tr>
<td>Mo</td>
<td>42</td>
<td>.60</td>
</tr>
<tr>
<td>Ta</td>
<td>73</td>
<td>.75</td>
</tr>
<tr>
<td>W</td>
<td>74</td>
<td>.79</td>
</tr>
<tr>
<td>Pt</td>
<td>78</td>
<td>.79</td>
</tr>
</tbody>
</table>

Brownell studied the backscattering of $^{32}$P rays from various Z materials in his spectrometer. He was thus able to determine both the energy and number of the backscattered $\beta$ rays. On integrating over energy (for the $\beta$ rays emitted perpendicular to the source) one finds the values for $f$ from Brownell's measurements, as listed in Table 5. Brownell's integrated results agree fairly well with the data of the others (e.g., Burtt), and consequently his differential data merit considerable confidence. The differential data are used later in evaluating the backscattered dosage.

Backscattering of positrons has been observed to be less than that of negatrons, in conformity with the difference between these types of particles in single scattering as already discussed. Seliger (3-5) observed, under experimental conditions very similar to Burtt's, that
backscattering of positrons from Na\(^{22}\) was 30% to 40% lower than backscattering of negatrons from I\(^{131}\), RaE, and P\(^{32}\), for Z between Be and Pb.

Table 5.
Brownell's Backscatter Measurements

<table>
<thead>
<tr>
<th>Backscattering Material</th>
<th>Z</th>
<th>f</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lucite</td>
<td>5.8</td>
<td>.14</td>
</tr>
<tr>
<td>Al</td>
<td>13</td>
<td>.26</td>
</tr>
<tr>
<td>Cu</td>
<td>29</td>
<td>.41</td>
</tr>
<tr>
<td>Sn</td>
<td>50</td>
<td>.55</td>
</tr>
<tr>
<td>Pt</td>
<td>78</td>
<td>.68</td>
</tr>
</tbody>
</table>

The dosage contributed by the backscattered electrons, to a thin surface layer of the backscattering material, was the third problem posed at the beginning of this section. Logically, perhaps, the analysis of this problem belongs here. However, since the details are a little lengthy and involve many of the ideas discussed in the next section, they are given in Appendix 1. These details are used in Chapter 7. The results are that approximately 20% of the total dosage at the surface is contributed by the backscattered electrons, at all energies of incident electrons below 1 to 2 Mev and at essentially all angular distributions of incident electrons.

B. Energy Loss from Swift Electrons

The dissipation of kinetic energy by electrons in matter is the fundamental physical problem with which this research is concerned. It takes 2 forms: ionization and
radiation. The elementary processes are discussed first, followed by the treatment of certain special problems pertinent to the energy loss from continuous $\beta$ ray spectra.

1. Elementary ionization process

The loss of energy from energetic electrons through ionization and excitation of the surrounding atoms has been treated theoretically by a number of investigators. The historical development of these theories is not discussed here. Instead, the best available theories are considered and compared where possible with the best experiments in order to estimate their validity.

The ionization process is a statistical one. Any one energetic electron will have wide deviations from the average rate of energy loss. In general, a large fraction of its energy is lost in little clumps of ionization rather than in a uniform distribution along the path. The statistical average process is considered first, and is followed by an analysis of the clumping.

a. Statistical average process

According to Evans (E-7), the rate of loss of energy from electrons by ionization and excitation, in increments of energy less than $W$, is given by the following currently preferred formula:

$$- \frac{dW}{ds} = \frac{2ne^4Nz}{m_0v^2} \left[ \ln \frac{2m_0v^2W}{J^2(1 - \beta^2)} - \beta^2 \right]$$

(6)

The symbols are defined as follows:

$W$ = upper limit considered for energy transferred in single collision
$T$ = kinetic energy of incident electron
$s$ = path length of electron trajectory
$e$ = electronic charge
\[ \frac{dT}{ds} = \frac{2\pi e^4 N Z}{m_0 v^2} \left[ \ln \frac{m_0 v^2 T}{J^2 (1 - \beta^2)} - \beta^2 \right] \]  

Eq. (7), in its several slight variations, is commonly known as the Bethe-Bloch formula. The low energy limit for its theoretical validity is reached when the electron has a velocity comparable with the Bohr orbital velocity of the atomic electrons (i.e., at a few keV for low Z absorbers). Approximations made in the derivation of the logarithmic term are thought to involve errors of only a few percent except at cosmic ray energies.

With the exception of the hydrogen atom, the theoretical evaluation of J cannot be carried out accurately. Consequently, J is commonly calculated from experimental particle data, where it is considered as an adjustable constant in making the best fit between theory and experi-
ment. Small errors in its magnitude are unimportant for the accuracy of the expression as a whole.

The experimental verification of this theory for electrons has proved a difficult problem, although there are good grounds for extrapolating its a particle validity to electrons. The experiments do not all agree, but probably the most careful were those of White and Millington (W-6), who studied the energy loss of the RaB and RaC conversion electrons in thin mica foils. Williams (W-8) has analyzed their data, and expressed their rate of energy loss results as $P_W$, with $P_W$ defined as

$$P_W = \frac{dW/ds}{2Ne^4N/m_0v^2}$$

Williams chose $W = 1500$ ev. The theoretical $P_W$, according to the Bethe-Bloch derivation, eq. (6), is given by

$$P_W = \ln \frac{2m_0v^2W}{J^2(1 - \beta^2)} - \beta^2$$

With $J$ taken as 150 ev from a particle measurements, I have compared the predictions of the Bethe-Bloch formula with Williams' analysis of White and Millington's data, as shown in Table 6. It can be seen that the two sets of values are in excellent agreement, considering the experimental difficulties. The theory can consequently be used with confidence that it is accurate to a few percent.

The Bethe-Bloch formula is more convenient to use when it is expressed in terms of energy dissipation per gm/cm$^2$ = $dW/\rho ds$, where $\rho$ is density. Absolute values for this function can be obtained from Fig. 4 and Table 7. The figure shows, as a function of energy, $dW/\rho ds$ in relative units normalized so that the minimum value = 1 for electrons in water and AgBr, respectively. As can be seen, these relative values of the function are very insensitive to the
Figure 4

RATE OF ENERGY LOSS BY IONIZATION vs β-RAY ENERGY
magnitude of \( J \) (listed below). If the value of the function for a variety of low Z materials is known in absolute units at the energy of minimum ionization, the graph for water permits one to determine its value at other energies with good accuracy. The same may be said for materials

Table 6.

Comparison of Theoretical and Experimental
Energy Loss by Electrons

<table>
<thead>
<tr>
<th>( T )</th>
<th>( P_W ) (theory)</th>
<th>( P_W ) (experiment)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.154 Mev</td>
<td>11.1</td>
<td>10.6</td>
</tr>
<tr>
<td>0.204</td>
<td>11.3</td>
<td>10.7</td>
</tr>
<tr>
<td>0.262</td>
<td>11.5</td>
<td>11.7</td>
</tr>
<tr>
<td>0.341</td>
<td>11.8</td>
<td>12.0</td>
</tr>
<tr>
<td>0.522</td>
<td>12.2</td>
<td>13.4</td>
</tr>
<tr>
<td>0.98</td>
<td>12.9</td>
<td>12.8</td>
</tr>
<tr>
<td>1.31</td>
<td>13.4</td>
<td>13.6</td>
</tr>
</tbody>
</table>

similar to AgBr. Table 7 shows the absolute minimum rate of energy loss of electrons in air, water, lucite, and AgBr. \( J \) has been taken in these computations as 18 ev in \( H_2 \), 82 in C, 98 in air, 111 in \( O_2 \), and 450 in AgBr (based on a set of a particle measurements as compiled by R.D.Evans). \( J \) for lucite \((C_5H_8O_2)\) is calculated as a weighted geometric mean of the above values, and is equal to 72 ev.

Table 7.

Minimum Rate of Energy Loss in Various Materials

<table>
<thead>
<tr>
<th>Absorber</th>
<th>Minimum rate of energy loss</th>
<th>( \frac{Mev}{gm/cm^2} )</th>
<th>( \frac{ergs}{gm/cm^2} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>1.76</td>
<td>2.82 x 10^{-6}</td>
<td></td>
</tr>
<tr>
<td>Water</td>
<td>1.81</td>
<td>2.90</td>
<td></td>
</tr>
<tr>
<td>Lucite</td>
<td>1.82</td>
<td>2.91</td>
<td></td>
</tr>
<tr>
<td>AgBr</td>
<td>1.21</td>
<td>1.93</td>
<td></td>
</tr>
</tbody>
</table>
One modification of the energy loss theory which has not been included in the Bethe-Bloch formula is the so-called polarization effect present in condensed (non gaseous) matter. When charged particles travel with almost the velocity of light, their electric field shape is distorted, according to the principles of relativity, into a disc shaped rather than spherical pattern. Under these conditions the field extends farther out from the path than it otherwise would. However, the polarization of the medium acts so as to reduce the field at a distance, thereby decreasing the rate of energy loss relative to the Bethe-Bloch theory. Halpern and Hall (H-1) have treated this effect in some detail, giving theoretical graphs for the correction to the rate of energy loss as a function of particle momentum. Their calculations indicate that for electrons in a low Z material such as lucite the correction is unimportant below 1 Mev, but rises to about 8 percent at 2 Mev and 18 percent at 20 Mev. (For the special case of graphite, they calculate a correction of 15% to 20% even at low energies, but this is almost certainly incorrect as demonstrated by the careful stopping power measurements of Gray (G-3).) Wick (W-7) has made another theoretical investigation of the polarization effect, and finds a similar but somewhat smaller correction. Experiments on this effect are rather inaccurate. The data of Pickup and Voyvodic (P-4) and of Crane, Oleson, and Chao (C-8) establish fairly clearly that the polarization effect is present in high energy electrons, but its magnitude is still in doubt. In this investigation the polarization correction to the Bethe-Bloch theory has not been applied. At the energies studied it is in any case not very large, although at higher energies the correction would probably be important.
b. Clumping of ionization

As stated earlier, the Bethe-Bloch formula gives the average rate of loss of energy, whereas any single electron loses much of its energy in clumps of ionization. This clumping effect is of significance in the exposure of photographic grains, and will consequently be briefly discussed here.

If the swift electron passes quite near to an atomic electron, it can transfer a sizeable fraction of its energy to the secondary electron. The secondary, known as a delta ray, will in turn ionize other atoms, giving a clump of ion pairs. The determination of the ionization clumping distribution therefore requires an analysis of the collision processes in which relatively large transfers of energy take place.

In summarizing formulas for collision processes, Bethe (B-8) gives the following expression for the cross section per (absorbing) electron for an energy transfer of amount between \( W \) and \( W + dW \):

\[
f(W) dW = \frac{2\pi e^4}{m_0 v^2} \left[ \frac{1}{W^2} - \frac{1}{W(T - W)} \cdot \frac{(2T + m_0 c^2)m_0 c^2}{(T + m_0 c^2)^2} \right. \\
\left. + \frac{1}{(T - W)^2} + \frac{1}{(T + m_0 c^2)^2} \right] dW
\]

\( T = \) kinetic energy of incident electron, as before. The energy transferred in these collisions = \( W f(W) \) dW, and the total amount of energy lost in collisions involving energy transfers in amounts between \( W_1 \) and \( W_2 \) is given by the integral:

\[
\int_{W_1}^{W_2} W f(W) dW
\]
The greatest possible energy transfer is considered to be $T/2$, as before, and the total cross section for the transfer of energy to delta rays of energy greater than $W_1$ is

$$\int_{W_1}^{T/2} W f(W) \, dW .$$

When one performs the integration, it is possible to compare the energy loss to delta rays with the total energy loss. In Fig. 5 I have plotted, for incident electrons of several energies in AgBr, the fraction of total energy loss which is contained in delta rays of energy greater than $W$, as a function of $W$. Actually, the curves are too high at low $W$ because the binding energy of the inner electrons in the AgBr prevents them from receiving such small energy contributions. (The Ag K edge, with 2 electrons, is 25 kev, and the L edge, with 8 electrons, is 4 kev.) A rough estimate of the delta ray production by a 0.2 Mev electron, taking into account atomic binding, is sketched in as the dotted line. The other curves would be reduced proportionately. The significance, for photographic exposures, of this clumping of ionization is discussed in a later chapter.

One further item regarding energy loss by ionization will be met in the following chapters. One of the most useful methods of detecting and studying $\beta$ rays consists in the measurement of the ionization which they produce in a volume of gas. Ionization can be measured rather simply even in absolute units. Now it is a very significant fact that for most gases, including air, the average energy dissipation $W$ per ion pair is essentially independent of the electron energy. (This $W$ is not related to the $W$ used in examining delta ray production.) Consequently, when $W$ is known one can convert ionization to energy dissipation (or dosage) without any detailed knowledge of the energy distribution of the electrons. This is, in fact, one of the
Figure 5
DELTA RAYS IN AgBr

FRACTION OF ENERGY LOSS CONTAINED IN DELTA RAYS OF ENERGY $> W$ vs. $W$.

- Atomic Electrons Considered Free
- Atomic Electrons Bound (Approx.)
best ways of measuring β ray dosage. The actual investigation of $W$ and its dependence on energy has been carried out by many workers. A critical review of the experiments on $W$ for air may be found in a paper by Gray (G-4), in which he concludes that $W = 32.5 \pm 0.5$ ev.

2. Elementary radiation process

Besides ionizing atoms in the absorbing medium, the electrons lose energy also through radiation, known as bremsstrahlung or continuous spectrum x rays. In practical problems of dosimetry, this bremsstrahlung is a complicating factor for the reason that such energy is absorbed at a distance from the place where it was given up by the electron. If the ionization dosage is highly localized, the bremsstrahlung contribution to the dosage in the local area is small. If the activity is uniformly distributed throughout a large volume, the full bremsstrahlung dosage must be added to the ionization dosage.

However, this problem is of little concern in this investigation, for bremsstrahlung represents a very small fraction of the total energy loss at the energies we deal with. Evans gives the ratio of radiation to ionization loss for electrons with velocity near that of light as:

$$\frac{(dT/ds)_{\text{rad.}}}{(dT/ds)_{\text{ion.}}} = 0.0012 ZT$$

$Z =$ atomic number of absorber, and $T =$ kinetic energy of electron in Mev. In lucite, for example, at 2 Mev, bremsstrahlung contains only a little over 1% of the energy lost by the electron. At the higher energies available with the new accelerators, however, the fraction of energy dissipation contained in bremsstrahlung cannot be ignored.
3. Path length and range of energetic electrons

The average path length of the electron trajectory can be computed by integrating the energy loss equation:

\[ \text{Path length} = pS = \int_{T_0}^{0} \frac{1}{dT/\rho ds} \, dT \quad \text{gm/cm}^2 \] (8)

There is, of course, much straggling in path length, so that wide deviations from the average are found for individual electron paths. Furthermore, the existence of scattering will make the range of the electron, or beeline distance traversed, considerably shorter than the path length. The average path length can in general be known only theoretically. A few measurements have been made, however, for low energy electrons in cloud chambers by Williams (W-8) and in photographic emulsions by Krudener (K-2). Krudener's measurements are compared with theory below. The average range can be determined only experimentally; however, it is not customary to measure this type of range, and in experiments reported, its value would depend greatly on geometry conditions (cf., for example, Rutherford, Chadwick, and Ellis (R-4), p. 411). The property usually measured in experimental absorption studies is the maximum range. (This is defined experimentally so as to permit an unambiguous measurement, but the intuitive meaning is sufficiently definite for present purposes.) It is found experimentally that the maximum range, in terms of mg/cm², is very nearly independent of the absorber Z over a wide range of Z. Measurements are usually made in aluminum.

In Fig. 6 are plotted the average path length vs. T for lucite and AgBr, as obtained from graphical integration of eq. (8). Also shown is a curve for maximum range vs. T, which is taken from a compilation by Glendenin (G-12) of experimental data for aluminum. (Most of Glendenin's points above 0.1 Mev are based on continuous \( \beta \) ray spectra absorp-
Figure 6

MAXIMUM RANGE AND AVERAGE PATH LENGTH
OF ELECTRONS

Energy (Mev.)

Path Length or Range - mg/cm²
tion measurements, but the difference between maximum range for monokinetic electrons and for continuous spectrum \( \beta \) rays is not large.

The theoretical average path length data of Fig. 6 may be compared with Krudener's experimental data. Krudener measured the path lengths of electrons in an Eastman NTB nuclear emulsion. At each of the energies 80, 102, 126, and 153 kev he examined approximately 50 tracks, and presented both the average path length and fractional standard deviation in path length at each energy. For comparison with theory, I have used Webb's data \( (W-5) \) on nuclear emulsion composition: percent weight of AgBr in emulsion = 82\%, percent weight of gelatin = 15\%, percent weight of water = 3\%, density of AgBr = 6.47 \( \text{gm/cm}^3 \), density of emulsion = 3.64 \( \text{gm/cm}^3 \). Since gelatin and water are similar to lucite, the path length in \( \text{mg/cm}^2 \) emulsion at a particular energy is given closely by: \( (0.82)(\text{path length in AgBr}) + (0.18)(\text{path length in lucite}) \). The path length in microns \( = \frac{1}{0.364} \times \text{path length in mg/cm}^2 \). Krudener's experimental data and the theoretical values taken from Fig. 6 are compared in Table 8.

<table>
<thead>
<tr>
<th>Energy</th>
<th>Observed average path length</th>
<th>Observed standard deviation</th>
<th>Theoretical average path length</th>
</tr>
</thead>
<tbody>
<tr>
<td>80 kev</td>
<td>45 microns</td>
<td>22%</td>
<td>43 microns</td>
</tr>
<tr>
<td>102</td>
<td>65</td>
<td>22</td>
<td>65</td>
</tr>
<tr>
<td>126</td>
<td>90</td>
<td>25</td>
<td>94</td>
</tr>
<tr>
<td>153</td>
<td>120</td>
<td>22</td>
<td>137</td>
</tr>
</tbody>
</table>

The agreement between theory and experiment is surprisingly good. It should be emphasized that the observed
standard deviation in path length is due mainly to actual straggling, rather than to experimental error.

4. Dosage from continuous spectrum $\beta$ rays

In practical applications of dosimetry with electrons, the continuous energy spectrum of $\beta$ rays is more frequently dealt with than are monokinetic electrons. Hence it is essential to understand the pertinent aspects of their interaction with matter in order to use film most efficiently in their dosimetry. Furthermore, as a byproduct of some of the experiments with film, later reported, I have obtained a certain amount of useful information on the spatial distribution of dosage around various types of $\beta$ ray sources. For these reasons, it will be advantageous to examine here some of the principles of continuous spectrum $\beta$ ray dosimetry.

a. Exponential absorption of $\beta$ rays

One of the earliest observed experimental facts, and also one of the most significant, was that in certain conditions of geometry (such as a thick plane source) $\beta$ rays are absorbed almost exponentially over a few half thicknesses. Furthermore, this type of absorption is observed in both ionization and counting measurements. Associated with each spectrum, and characteristic of its maximum energy, is a linear absorption coefficient $\mu$ cm$^{-1}$, or a mass absorption coefficient $\mu/\rho$ cm$^2$gm$^{-1}$ ($\rho$ being the density of the medium). The mass absorption coefficient has the convenient property of being relatively independent of the atomic number of the absorber.

The absorption coefficients are to be looked on somewhat in the nature of rules of thumb. They vary somewhat with absorber thickness (as is required by the fact of the finite range) and with geometry of detection and
source. They are sufficiently unambiguous, however, as to be very useful. Gleason, et al. (G-1) have recently made absorption studies on a large number of isotopes. They used thin sources, without backing, at a distance of 4 to 6 cm from a conventional end window counter; the Al absorbers were placed a few mm in front of the window. They observed the following relationship between $\mu/\rho$ and the maximum energy, $E_{\text{max}}$ Mev, of the particular isotope:

$$\frac{\mu}{\rho} = 17 E_{\text{max}}^{1.43} \text{ cm}^2/\text{gm}.$$ 

For $\beta^{35}$ (0.167 Mev) and $\beta^{32}$ (1.70 Mev), this formula predicts $\mu/\rho = 220 \text{ cm}^2/\text{gm}$ and $7.8 \text{ cm}^2/\text{gm}$, respectively. Recent measurements in our laboratory under roughly similar conditions, except for the use of thick sources (plaster of Paris, which is CaSO$_4 \cdot 2\text{H}_2\text{O}$), gave $\mu/\rho = 250 \text{ cm}^2/\text{gm}$ and $7.7 \text{ cm}^2/\text{gm}$ for $\beta^{35}$ and $\beta^{32}$, respectively.

It would probably be possible to imagine various sets of coincidental mechanisms of absorption which would account for the exponential character of absorption as detected either by ionization chambers or by counters, instruments which differ widely in their response as a function of electron energy. The following set is found experimentally to be most nearly correct: (1) the energy spectrum remains nearly uniform throughout a considerable range of absorption, and (2) the angular distribution pattern of the $\beta$ rays remains substantially constant under absorption. It must be emphasized that both of these conditions are essential; either one alone would not give the observed absorption properties. An example will make this more clear. Consider a parallel plate ionization chamber, one of whose plates consists of an absorber of variable thickness over a plane source of $\beta$ rays. A certain amount of energy (number of $\beta$ rays x average energy per $\beta$ ray) enters the air gap; a fraction is absorbed, while most passes on through. If (1) is true, the fraction of incident energy which is absorbed in the gap per unit path length of the $\beta$ rays in the gap, is independent
of the thickness of absorber over the source. The total fraction of energy absorbed in the gap is proportional to the total path length of the β rays in the gap. This total path length in the gap is independent of thickness of absorber if, and only if (for practical purposes), condition (2) is true. Therefore if (1) and (2) are both true, the fraction of the incident energy which is absorbed in the gap is independent of the thickness of absorber over the source. This is exponential absorption. If (2) were valid, but the average energy of the β rays decreased with absorption so as to make (1) invalid, then, in view of the higher specific ionization of lower energy electrons, the fraction of incident energy absorbed in the gap would increase with thickness of absorber and exponential absorption would not exist.

The experiments substantiating condition (2), to a rather good approximation, have already been referred to in the analysis of β ray scattering. In general, a \( \cos \theta \) distribution tends to be established, and is fairly stable under absorption. Brownell, from whom much of the foregoing reasoning is taken, has carried out an extended series of experiments to examine the validity of condition (1), the constancy of the β ray spectrum, for the case of \( \beta^{32} \).

Three of his graphs are presented here as Figs. 7, 8, and 9. Fig. 7 shows the number of electrons per unit energy interval, as a function of energy, in the initial \( \beta^{32} \) spectrum, in the initial spectrum plus saturation backscatter from lucite, and in the spectrum transmitted through various thicknesses of lucite absorber. (The results are for β rays emitted normal to the source or absorber surface, and are slightly different for other angles.) Figs. 8 and 9 show the average energy of the β rays in the spectrum transmitted through lucite and Al absorbers, respectively. These figures indicate that the constancy of the β ray spectrum under absorption is a reasonable assumption.
Fig. 7

$^{32}\text{P} \cdot \text{LUCITE} \cdot 0^\circ$

- o no absorber
- △ 23.9 mg/cm$^2$ absorber
- □ 89.9 " "
- × 237.5 " "
- ▽ 418.2 " "
- ⋄ 237.5 " backscatterer

$N(E)$ vs. $E$ (Mev)
**Fig. 8**
P$^{32}$ - AVERAGE ENERGY - LUCITE

- no absorber
- all angles

- $0^\circ$
- $30^\circ$
- $60^\circ$
- $70^\circ$

**Fig. 9**
P$^{32}$ - AVERAGE ENERGY - ALUMINUM

$E$ (Mev) vs. $mg/cm^2$ absorber.
There are at least two important conclusions to be drawn from the approximate constancy of angular distribution and energy spectrum of β rays during the absorption process. First, by making use of these conditions it is possible to derive a rather simple but general theory of β ray absorption, or dosimetry, based essentially on a diffusion type of analysis. This Brownell has done, and the reader is referred to his work for details. Second, and more pertinent to the problems of this thesis, is the simplification introduced by these conditions into the experimental measurement of β ray dosage. All β ray detectors have a response to β rays which, in varying degree, is a function of energy and angle of incidence that differs from the ideal for dosimetry measurements. However, to the extent that the angular distribution and spectral shape remain constant, it is possible to calibrate the instrument under one set of absorption conditions and use it with accuracy under other absorption conditions. This is especially useful in β ray dosimetry at the surface of thick sources (e.g., bones) in which the spatial distribution of the activity, and therefore the absorption conditions, are unknown. I have carried out considerable experimental work in the course of this thesis in order to investigate the extent to which these absorption conditions simplify β ray dosimetry with photographic film.

b. Dosage from plane source of β rays

There are at least 2 special case β ray dosimetry problems which are involved in the experimental work of this thesis. The first is dosage from a plane source of β rays, and the second is dosage from a point source. The first is considered now.

The goal is to describe the dosage produced by β rays, from a plane source, at any prescribed depth in an
absorbing medium covering the source. Several theories are in existence which can be applied to this situation. Brownell's, which is the simplest, is mentioned first.

Brownell assumed, as previously discussed, the constancy of spectral shape and angular distribution. However, it was pointed out earlier in this chapter that the angular distribution of the β rays is isotropic at the surface of a thin plane source, becoming \( \cos\theta \) only beyond some minimum distance into the absorber. As a result of this changing angular distribution, Brownell's theory cannot be applied to the initial absorption (perhaps about 50 mg/cm\(^2\) for \(^{32}\)P in lucite). Beyond this, however, it predicts, of course, an exponential decrease in dosage with depth in absorber.

During the course of this investigation I have actually measured experimentally the dosage resulting from plane sources of β rays. The measurements were carried out with a parallel plate ionization chamber of such small gap that the gap does not distort the dosage distribution. This is an extrapolation chamber, discussed in Chaps. 3 and 7 and in Appendix 10. It is therefore of interest to compare this and the following theory with the experimental results. These results apply, for our present purposes, to the condition of thin plane sources in an infinite lucite absorbing medium. The absorption of β rays from 4 isotopes (\(^{91}\)Y, 1.54 Mev; \(^{204}\)Tl, 0.78 Mev; \(^{185}\)W, 0.43 Mev; \(^{35}\)S, 0.167 Mev) is shown plotted in 2 different ways in Figs. 10 and 11. The abscissa gives the absorber thickness at which the dosage is measured; the ordinate gives the dosage rate in units of 100 ergs/gm hr (which is approximately 1 rep/hr). It can be seen in Fig. 10 that over the first part of the curves, where changes are occurring in the angular distribution, the absorption is far from exponential. At greater depths in the absorbing medium the curves show signs of
Figure 10

PLANE $\beta$ RAY SOURCES: DOSAGE RATE IN ABSORBER vs ABSORBER THICKNESS.
Figure 11

PLANE $\beta$-RAY SOURCES DOSAGE RATE IN ABSORBER vs ABSORBER THICKNESS.

- **Experimental**
- **Theoretical**

Absorber Thickness - mg/cm$^2$

Dosage Rate - 1000 ergs/gm hr.
becoming more nearly exponential. Loevinger (L-7) has made almost identical measurements to greater absorber depths, using $^{32}\text{P}$ ($1.70$ Mev) in polystyrene. He found almost identically the same curve as my $^{91}\text{Y}$ data for the same range of absorber thickness, followed by an exponential function to $350$ mg/cm$^2$. Toward the end of the $^{32}\text{P}$ range a more rapid decrease occurred, as of course is necessary to give a finite range. Hence, Brownell's theory gives a good description of the dosage from $\beta$ rays in the range where his own experiments showed the assumptions to be valid (beyond about $50$ mg/cm$^2$, but not near the end of the range). The high initial dosage rate is expected because the thin source isotropic angular distribution of $\beta$ rays relative to the $\cos\theta$ distribution gives an excessively large number of $\beta$ particles travelling almost parallel to the source, and therefore contributing a large dose near it.

A second type of treatment has been used by Evans (E-6), and can be seen in a rather general form in a discussion by Caswell (C-2). This treatment provides for changes in angular distribution as caused by absorption, but not by scattering, and it retains the assumption of exponential absorption. Specifically, it assumes a function for the distribution of dosage about a point source, namely

$$I(r) = k \frac{e^{-\lambda r}}{r^2},$$

which takes into account decrease of dosage $I(r)$ at distance $r$ through exponential absorption and inverse square geometry. This function is then integrated over the entire source to give the total dosage at any point in the absorbing medium. It must be pointed out that the angular distribution of $\beta$ rays about a point source is a rather strong function of $r$, in identically the same way as the angular distribution of electrons incident normally on an Al foil is a strong function of depth in the foil. Consequently,
the assumption of exponential absorption is rather unjusti-
ified (at least on the basis of the thick plane source ex-
periments). It is shown rather simply in Appendix 2 that
when the dosage from a thin plane source is calculated ac-
cording to the above assumptions, one finds that at a point
a distance $a$ inside the absorber the dosage is given by

$$D(a) = k_1 \int_{1}^{\infty} e^{-\mu a} \frac{dz}{z},$$

where $z$ can here be considered merely a dummy variable in
the definite integral. This is a logarithmic integral
$-E_1(-\mu a)$ whose values as a function of $\mu a$ can be found in
tables. By trial and error a value for $\mu$ (or $\mu/\rho$) has been
found which gives the theoretical curves shown in Fig. 11.
The $\mu/\rho$ values used were: $Y^{91}$, 2.5 cm$^2$/gm; $Tl^{204}$, 10.8;
$W^{185}$, 35.0; $S^{35}$, 175. It can be seen that by judicious ad-
justment of the parameter, a rather good fit of this theory
to the experimental data can be obtained. The conventional
absorption experiments give the following values for $\mu/\rho$
as taken from the equation $\mu/\rho = 17E_{\text{max}}^{-1.43}$: $Y^{91}$, 9.2
cm$^2$/gm; $Tl^{204}$, 24; $W^{185}$, 57; $S^{35}$, 220. It is apparent that,
especially for the higher energy spectra, the $\mu/\rho$ values
from my experimental data are too small by a rather large
factor. The reason for this discrepancy undoubtedly is to
be found, as already suggested, in the difference between
the angular distribution conditions of the 2 experiments.
Beta rays initially travel out radially from a point source,
contributing a minimum dosage in concentric shells of small
radius because the path length in these shells is a minimum.
As the particles are scattered at larger distances they
acquire a component of tangential velocity, so that their
path length in shells of specified thickness increases as
the radii of the shells increase. The increase of dosage
resulting from the greater path length in the outer shells
is evidently sufficient to counterbalance much of the absorp-
tion which of course has been taking place in the shells of
smaller radii.

A third, and apparently rather effective, means of treating the dosage from a plane source is to set the problem up as in the last method, except that the distribution function $I(r)$ for the dosage about the point source is given an experimentally determined form. This analysis should make possible a rigorously correct general solution for the dosage from any $\beta$ ray source distribution. The difficulties associated with this treatment are two: first, the experiment to determine $I(r)$ is a difficult one, and second, unless $I(r)$ can fortuitously be described by a simple analytical function, the mathematics is difficult or impossible. As far as I know, no accurate direct measurements of $I(r)$ for any isotope have yet been made, although Marinelli (M-2) is now setting up such an experiment using a wire grid ionization chamber in air. However, Loevinger (L-8) has made some very interesting indirect determinations of $I(r)$ for $P^{32}$, by using experimental plane source data of the type which we have just been trying to explain. His measurements cannot be used, therefore, in interpreting my plane source dosage curves, but instead my curves can be used as a check, and extension to other isotopes, of his point source function $I(r)$. This information would be a very useful byproduct of my plane source measurements, and is therefore treated in the next section.

c. Dosage from point source of $\beta$ rays

Loevinger has developed a theory in a rather general form for expressing $I(r)$ in terms of quantities measured in a parallel plate extrapolation ionization chamber. For details, the reader is referred to his papers. However, the fundamentals are simple enough. If it is accepted for the present that the extrapolation chamber can measure the dosage $D(a)$ which is produced at $P$ on a plane at distance $a$ into an absorber covering a plane source, the
following expressions can be derived, referring to Fig. 12.

\[ D(a) = k \int_{0}^{\infty} 2\pi r \, dr \, I(r) = k \int_{a}^{\infty} 2\pi r \, dr \, I(r) \]

\[ \frac{dD(a)}{da} = -k2\pi a \, I(a) \]

or \[ I(a) = -\frac{k1}{a} \frac{dD(a)}{da} \]  \hspace{1cm} (9)

\[ I(a) \] is simply the function we wished to find for the dosage distribution about a point source, namely \( I(r) \), evaluated at \( r = a \). \( I(r) \) is consequently expressed in terms of quantities experimentally measurable in a parallel plate extrapolation chamber. For \( P^{32} \) Loevinger found the following relationship for \( D(a) \):

\[ D(a) = A - B \ln a \]

\[ = C e^{-da} \]

\[ \rho a < 110 \text{ mg/cm}^2 \]

\[ 110 < \rho a < 350 \text{ mg/cm}^2 \]

\( A, B, \text{ and } C \) are constants. On solving for \( I(r) \), which is the same as \( I(a) \), one finds:
\[ I(r) = \frac{k_2}{r^2} \quad \rho r < 110 \text{ mg/cm}^2 \]

\[ = \frac{k_3}{r} e^{-\mu r} \quad 110 < \rho r < 350 \text{ mg/cm}^2 \]

In other words, for \( \rho r \) less than 110 mg/cm\(^2\), \( I(r) \) falls off only with the inverse square law, having no apparent absorption. Beyond this it falls off more rapidly than in inverse square (despite the first power term of \( r \) in the denominator), showing the presence of absorption. Both of these functions are fairly simple to handle analytically. Loevinger has calculated, for example, the dosage distribution near a thick plane source, and found it to agree very well with experiment.

My own data, taken for other purposes, do not carry the absorption through a sufficiently extensive range to make worthwhile the fitting of analytical functions to the curves. However, the following interesting conclusions can be drawn: When \( D(a) \) vs. log of absorber thickness, as in Fig. 11, gives a straight line, dosage falls off as pure inverse square. This is quite closely the case for \(^{91}\text{Y}\) (which is nearest \(^{32}\text{P}\) in energy); a straight line comes within 5% of each point. (The previous theory, which automatically put in absorption, led to a small absorption coefficient.) The other isotopes give curves with positive second derivatives, which indicate that dosage falls off somewhat more rapidly than \( 1/r^2 \). To see this, consider the function \( a^2 I(a) \); if it decreases with increasing \( a \), dosage decreases more rapidly than \( 1/a^2 \) (\( = 1/r^2 \)):

\[
a^2 I(a) = -a^2 \int_1^a \frac{dD(a)}{da} = -a^2 \int_1^a \frac{dD(a)}{d\ln a} \cdot x \frac{d \ln a}{da} = - \frac{dD(a)}{d \ln a}
\]
Therefore \[
\frac{d}{dr} \left[ a^2 I(a) \right] = \frac{d}{da} \left[ \frac{-D(a)}{d \ln a} \right] = \frac{1}{a} \frac{d}{d \ln a} \left[ \frac{dD(a)}{d \ln a} \right]
\]

\[= - \frac{1}{a} \left\{ \frac{d^2 D(a)}{d(\ln a)^2} \right\} \]

The quantity in brackets is the second derivative of the curves in Fig. 11. Therefore, if the second derivative is positive, dosage decreases more rapidly than \(1/a^2\), showing absorption.

At first sight, Loevinger's experimental method for determining \(I(r)\) appears to be a very powerful one. Actually, it is not that simple. Since at each measurement in the extrapolation chamber one detects \(\beta\)'s for all \(r\), it can be seen intuitively that very precise measurements are required to give \(I(r)\), for a particular \(r\), accurately. Formally, one must determine not just \(D(a)\), but rather the slope of \(D(a)\), with good accuracy.
Before proceeding further into the analysis of photographic film as a β ray dosimeter, it would be well to consider the various different types of β ray dosimetry instruments already in existence. After a brief survey of the instrumentation available, it will be possible to assess in better perspective both the merits and the limitations of photographic film as a dosimeter.

Any instrument which is capable of detecting β rays is capable also of measuring, although perhaps only under certain narrowly prescribed conditions, β radiation dosage. The practicality of an instrument is determined in general by the range of experimental conditions in which it is capable of reliable results. We now consider briefly several specific instruments for β ray detection, assessing the suitability of each for dosimetry measurements.

A. Ionization Chamber

The most generally useful β ray dosimetry instrument is the ionization chamber. It combines reasonable sensitivity with the direct measurement of the ionization produced by β rays. As has already been pointed out, the direct measurement of ionization gives an accurate measurement of dosage.

Many types of ionization chamber have been designed for the measurement of radiation dosage from β rays. The more conventional type consists simply of a small, enclosed, fixed volume of air between 2 electrodes that collect the ionization produced by β rays in the air. Examples of this type of chamber are those of Neary (N-2), and of Blomfield and Spiers (B-13). The latter, to be specific, was a small cylindrical chamber 4 mm in diameter and 1 mm
deep, with a 10 mg/cm² graphited cellophane window, electron metal sides, and amber insulator separating the electrodes. Voltage changes produced by the collected charge were measured with a Lindemann electrometer. With this chamber they attempted to evaluate the dosage rates at the surface of β ray plaques. These consisted of deposits of Radium (and its decay products) on such materials as brass, with several mg/cm² monel foil filtration. By taking a set of measurements under different distance and absorption conditions they were able to extrapolate to find the dosage rate at the surface of the plaque. The authors compared their data with independent measurements on the same applicator by Neary; they found a 15% discrepancy between the two values for surface dosage rate, but only a few percent discrepancy for the dosage rate a few mm from the applicator surface. The results of this comparison afford perhaps the best evaluation of the accuracy to be attributed to their measurements.

Crabtree and Gray (C-6) used a similar type of chamber for a similar purpose, but were attempting to measure dosage rates over a smaller area. Their chamber, a paper walled cylinder of only 5 mm³, may be considered near the practical limit of resolution in the dosimetry of β rays with this kind of ionization chamber. They admit a 15% possible error in their measurements.

The chief limitations on accuracy of dosimetry with the conventional type of ionization chamber are absorption of β rays in the window and air, and the distortion of the β ray spatial distribution by the air cavity. A more specialized type of ionization chamber has been developed by Failla and others in recent years which, in particular circumstances, effectively removes these two limitations. The basic modification which distinguishes this so-called extrapolation ionization chamber from the other types
is its variable volume. If ionization as a function of volume is determined by a set of measurements, the readings can be extrapolated to zero volume, thus eliminating the air cavity geometrical distortion and $\beta$ ray absorption. If furthermore, the chamber is so constructed that the source itself forms one electrode, the window with its attendant absorption of the $\beta$ rays can be eliminated. The extrapolation ionization chamber is probably the most precise $\beta$ ray dosimetry instrument. Its limitations lie in the specialized source construction necessary, its insensitivity at small volume, and its poor spatial resolution.

**B. Chemical Dosimetry Instruments**

Another instrument for $\beta$ ray dosimetry is the type which makes use of chemical changes produced in matter by the radiation. Every form of matter has chemical changes induced in it by the bombardment of $\beta$ rays. In fact, there is no doubt that it is these chemical changes which are responsible ultimately for the biological damage caused by $\beta$ rays. Day and Stein (D-1) have given a survey of chemical means of dosimetry of ionizing radiations, with emphasis on water systems consisting of liquids or solid gels. They attribute most chemical changes in water systems to oxidation and reduction reactions by H and OH radicals, which are believed to be produced shortly after ionization of water molecules. The chemical changes may be detected by a variety of techniques, such as titration, colorimetry, conductivity, etc.

In certain problems, these chemical methods have certain important advantages. The chief distinctive advantage is that the method is ideally suited to the determination of the total amount of energy dissipated throughout a large volume. The tissue-like composition of the detector is an important advantage in x ray dosimetry. Finally, the response of the detector, as a function of $\beta$ particle or
secondary electron energy, very nearly meets the ideal requirements for dosage measurement. The chief disadvantage of the method is its low sensitivity, for the lower limit of detectability is of the order of magnitude of several hundred reps.

C. Calorimeter

A third possible dosimeter of β rays is the calorimeter, an instrument which measures energy dissipation per gram very directly. It has actually been used in measurements related to dosimetry by Zumwalt and co-workers (Z-1). These men measured the rate of evolution of nitrogen gas from liquid nitrogen at its boiling point in order to determine the energy liberation from P\textsuperscript{32}. Although apparently very accurate results were achieved, the method is so insensitive and inconvenient to use that it is suitable only in certain very specialized applications.

D. Beta Ray Counters

Beta ray counters, whether Geiger-Mueller, proportional, or scintillation, are the most sensitive dosimeters of β rays. Geiger-Mueller counters are widely used for this purpose, although in a crude manner, in the form of β ray survey meters. The reason for this crudeness is that the GM counter measures number of particles, rather than dosage, and the relationship between the two depends markedly on β ray energy, as was seen in the previous chapter. If the energy dependence of the sensitivity of the GM counter (when it is considered as a β ray dosimeter) could be circumvented by calibration with a known source of β rays having an energy spectrum identical with that of the unknown source, the instrument would yield fairly reliable dosimetry data. The proportional counter, however, measures a quantity which is almost identical with ionization; as we
have seen, this would permit quite accurate β ray dosimetry with this type of counter. It would be more sensitive, but less adaptable and convenient than the ionization chamber. Scintillation counters are capable of measuring the number and energy of incident β particles, which could be converted to dosage at the crystal surface with fair accuracy. If the crystal could be made as thin, in terms of mg/cm$^2$, as can the gas thickness in proportional counters or ionization chambers, scintillation counters would measure essentially the same quantity as do these instruments.

E. Photographic Film

Finally, photographic film has already found some use in the dosimetry of β rays. The chief use has been in so-called film badges worn as survey instruments by people exposed to radiation. The β rays expose the film in the same manner as does light, and the degree of photographic blackening gives some measure of the absolute β radiation dosage. This dosimetry technique should be classed logically under "chemical methods", for the photographic action of β rays on film is chemical in nature. The difference between this technique and the other chemical techniques lies in the development process to which photographic film is subjected. This introduces what is essentially amplification of the initial chemical reaction by a factor of perhaps $10^8$, and raises photographic sensitivity to a very practical level. The greatest limitation of film as a dosimeter can be expected to lie in the dependence of its sensitivity on β ray energy, in the same way as the GM counter. Its greatest advantage is its high resolution. A second advantage, related to this, is a high sensitivity which in certain applications surpasses even that of β ray counters. To a first approximation, the lower limit of activity detectable with a counter depends on the total activity of the source; with film it depends more nearly on the activity/cm$^2$ of the source. Thus a source of 1 mm diameter may
have low total activity but a high activity/cm²; it may therefore be below counter background but above film background.

F. Summary

To summarize briefly the methods of β ray dosimetry: There are a wide variety of instruments capable of detecting β rays, and each of these can be used, under certain conditions, as a β ray dosimeter. The most widely used, and in special cases the most accurate instrument, is the ionization chamber. However, it does have limitations which preclude its applicability in certain problems, such as that which gave rise to this thesis. To fill these gaps, other types of instruments are used, the choice depending on the nature of the problem. The problem of resolution is best surmounted by the use of photographic emulsions as the dosimeter; hence an investigation of this method was necessary to carry out the biological study outlined above.
A knowledge of the fundamental principles of the photographic process is essential to an intelligent use of film in the dosimetry of β rays. In this chapter these principles will be discussed in a somewhat general manner, leaving the photographic action of β rays, specifically, to a later chapter.

Much of the following analysis is taken from the review by James and Higgins (J-1), to which the reader is referred as a convenient but more detailed survey of the subject. Supplementary material is taken from Mees' authoritative work (M-5), and other sources listed in the bibliography.

In briefest form, the photographic process can be described as follows. The sensitive material, known as the emulsion, consists of small silver halide crystals in gelatin. Under the action of the exposing agent (light, β rays, etc.), a so-called "latent image" is formed in the grains such that they can be reduced to silver by the chemical action called development. Certain other chemical steps are necessary in order to make the final image permanent. The emulsion and developer characteristics determine both the relationship between the quantity of exposure and the resulting photographic effect, and also the microscopic structure, or resolution properties, of the final image.

A. Emulsion Composition

The photographic emulsion is always used in the form of thin layers spread on film (cellulose acetate) or glass. The thickness ranges from a few microns to several hundred microns, with most common emulsions between about
10 and 25 microns. This thickness for common emulsions corresponds roughly to a mass of from 2 to 5 mg per cm$^2$. Over the emulsion surface there is usually a very thin protective gelatin layer, sometimes known as a T coat, of thickness about 1/2 micron = 0.05 mg/cm$^2$.

The primary purpose of the gelatin in the emulsion is to support the silver halide grains in a permanent manner, while still permitting ready access of the processing chemicals to the grains. Besides this passive role, however, the gelatin also plays an active part in the photographic process. First, it affects the exposing agents as they pass through the emulsion. Ultraviolet light is highly absorbed in the gelatin, making necessary special low gelatin content emulsions for this type of work. Electrons also, as is clear from the discussion of Chap. 2, are absorbed to some extent by the gelatin, which usually represents about half the mass of the emulsion (depending partly on the humidity). Second, the gelatin affects the photographic properties of the silver halide. The reaction of certain trace sulfur containing compounds in the gelatin with the silver halide grains forms on them "sensitivity centers", mentioned later, which enormously increase the photographic sensitivity of the emulsion. Sheppard was able, in fact, to find a relationship between the sensitivity of his emulsions and the quantity of mustard seed in the diet of the calves from whose skin the emulsion was made. Moreover, the moisture content of the emulsion affects the permanence of the latent image, high humidity tending to increase the rate of fading.

The silver halide grains in most emulsions are predominantly AgBr. However, they often contain a few percent of AgI, which increases sensitivity. AgCl is sometimes the primary ingredient of the crystals in such slow photographic materials as photographic paper. These silver halide crystals are precipitated from solution in the presence
of the gelatin. By careful attention to conditions, the size of the grains may be controlled with considerable reliability. Most often the grains are triangular or hexagonal tablets ranging from a small fraction of a micron to a few microns on a side. If one plots \( N(a) \), the number of grains having projection area (area as seen by a beam of electrons perpendicular to film) between \( a \) and \( a + da \), as a function of \( a \), one finds in certain special emulsions a rather symmetrical Gaussian form of size-frequency distribution. More often, however, a skewed distribution exists with a tail extending toward the large areas, as shown in Fig. 13.

![Gaussian distribution](image)

**Fig. 13**

A more significant type of size distribution curve is that in which one presents total projection area of all grains between \( a \) and \( a + da \), as a function of \( a \). In other words, one plots \( aN(a)\,da \) vs. \( a \). \( aN(a) \) is proportional to the amount of light absorbed from a beam by the grains of area \( a \), either in the undeveloped or developed emulsion. This latter curve can be represented, to a first approximation, by giving \( N(a) \) the exponential form

\[
N(a) = \frac{N}{\bar{a}} \, e^{-\frac{a}{\bar{a}}}
\]

where \( \bar{a} \) is the average grain area, and \( N \) is the total number of grains. If one defines the average grain size as the total projection area of all grains divided by the total number of grains, the average grain size of common emulsions ranges from about 0.3 \( \mu^2 \) in motion picture positive
film to about 2 $\mu^2$ in x-ray films. We shall see later that the grain size and size distribution are of practical importance in determining film sensitivity and other significant properties.

B. Latent Image Formation

The specific nature of the latent image, or attribute of the exposed grains which makes them susceptible to development, was for a long time a mystery. However, in the last decade or two enough experimental and theoretical work has been accomplished to give a very plausible explanation of the phenomenon. This generally accepted theory of latent image formation, together with supporting experimental evidence, is briefly described in this section.

The theory was first stated in essential completeness by Gurney and Mott (G-7); it asserts the following: The exposing agent transfers some of its energy to the AgBr (or other silver halide) crystal, thereby raising the energy of one or more electrons into the conductance energy band. These electrons then travel through the crystal until trapped in sensitivity centers, which might consist of an impurity (e.g., sulfur containing compound) or deformities in the crystal lattice. The electrostatic potential set up about these centers brings into action a second process. A certain very small fraction of silver ions in the crystal are free to migrate through the crystal, the fraction depending strongly on the temperature. These ions are attracted to the trapped electrons, which neutralize the ions to give silver atoms. Under continuing exposure these 2 processes continue until a group of silver clumps, containing one to several silver atoms each, are distributed throughout and on the surface of the crystal. These clumps constitute the latent image, and in some way not completely
understood catalyze, during the development process, the chemical reduction of the remainder of the ionic silver in the grain.

The experiments in support of each of the important features of this process are very convincing, but they can only be touched upon here. First of all, the latent image is almost certainly metallic silver. The chemical reactions of the latent image are approximately the same as macroscopic amounts of silver; furthermore, when the exposure is great enough, analytically determinable amounts of silver are found in the crystal bearing a direct proportion to the quantity of exposure. Second, it can be demonstrated, from the dependence of the electrical conductivity of a silver halide crystal on illumination (i.e., from photoconductance), that electrons are raised to the conductance energy band by the absorption of light in the crystal. Moreover, the dependence of photoconductivity on wavelength of illuminating light parallels, to some extent, the dependence of photographic sensitivity on wavelength. Third, the dependence of photoconductance current on applied voltage indicates that the electrons are readily trapped, especially in crystals containing impurities. Fourth, the fact that the primary electronic process does not give the whole picture can be demonstrated by experiments at low temperatures: photographic sensitivity is very low at the temperature of liquid air, whereas photoconductance is practically as high as at room temperature. Fifth, the variation of photographic sensitivity with temperature does, however, rather closely parallel the variation of ionic (dark) conductivity with temperature. Finally, the fact that the secondary ionic process by itself is not the complete picture can be demonstrated by repeated exposures at low temperatures alternated with periods of warming to room temperature. In these experiments it is evident that a part of the latent image formation process does take
place at low temperatures, and is "remembered" until room temperatures permit completion of the process. Thus, almost every step of the Gurney-Mott theory has been shown by experiment to be both a physically possible and a physically necessary phase of latent image formation.

There are a number of effects that take place under certain conditions of exposure in which the photographic action of a given exposure is dependent on other properties than the mere quantity of exposure. The only one of these which will be of primary concern in our problems is the so-called reciprocity law failure. The reciprocity law states that the photographic effect of a given continuous exposure is a function only of the quantity $I_t$, where $I$ is the intensity of the exposing agent and $t$ is time of exposure. This law is known to fail for most emulsions in exposures to light; there is a certain $I$ for which the photographic action of a given exposure, $I_t$, is a maximum. The quantitative relationship between intensity of exposing agent and photographic effectiveness of exposure can be expressed in many ways. One of the most common is to express the photographically effective exposure, $E_{eff}$, by the equation $E_{eff} = I^p t$, where $p$ is the so-called Schwarzchild constant. If $p = 1$, the reciprocity law is valid. It is now well established that this simple equation cannot express the true dependence of $E_{eff}$ on $I$ and $t$ for most films; in other words, $p$ is usually a function of $I$ or $t$ rather than being a constant. However, when $p$ is nearly unity, the expression is a useful approximation and in this case the deviation of $p$ from unity is an adequate measure of reciprocity law failure.

Reciprocity law failure can be explained in terms of the Gurney-Mott theory in the following way. At high intensities, electrons are raised into the conduction band, and enter traps, so rapidly that the secondary and
slower ionic conduction process can not keep up. In this case, a particular electron caught in a trap, until finally combining with a silver ion, will repel other approaching electrons. These electrons will then be forced to start other silver clumps at other traps. The net result is that more silver clumps are formed than at lower intensities, but they are smaller and are thought to be much less effective in catalyzing development. At low intensities, the silver clumps grow very slowly. While still small, they are thought to be rather unstable, with the result that they may disintegrate before growing to stable size. We shall use these ideas later in discussing the reciprocity law validity for β ray exposures.

C. Film Processing

The exposed film is developed to bring out the image from the latent image, soaked briefly in the "stop bath" to give an abrupt end to development, fixed to remove unexposed silver halide, and washed to remove the processing solutions.

There are 2 types of development, described, rather inappropriately, as chemical and physical. In chemical development each grain is reduced as a unit to silver, the developer serving merely as the reducing agent. In physical development, each silver clump in the latent image acts as a unit, silver being deposited on each clump from solution in the developer. Certain developers combine both processes, but the first is of much the greater practical importance.

It has been reported (V-1, C-12) that physical development produces an image more nearly a function of radiation dosage in the emulsion, at least in the case of heavily ionizing particles such as α's, than does chemical development. This would of course be a useful property if
it were true in the case of β rays. However, the physical development procedure is rather tricky, and in the expectation that more would be lost by irreproducibility than would be gained by improved dosage response, I have not seriously attempted physical development in this investigation.

Chemical development, as stated above, is an oxidation-reduction reaction. The developer contains a reducing agent, usually an aromatic organic compound, which has such properties that it will reduce the silver ions to silver in the presence of the latent image as catalyst, but will attack unexposed silver halide much less rapidly. In a general way, individual grains act as units in the development process. However, their size and shape change somewhat, and a certain amount of clumping takes place.

Besides the specific developing agent, the developer contains various other chemicals which fulfill special needs. An alkali is present to adjust the developer pH to that at which the developing agent behaves most satisfactorily. Sodium sulfite is usually added to help preserve the developer and to remove the stain producing oxidation products of the developing agent. Potassium bromide is often present as a fog restrainer; it inhibits reduction of unexposed silver halide more than exposed.

The action of the developer on photographic film is determined by many factors other than original developer composition. Like any chemical process, the reaction rate increases rapidly with increasing temperature. Development is usually carried out at about 20°C. The rate is also influenced by local developer concentration. If development is carried out with no agitation, variations in developer concentration result from variations in the oxidation of the developer between points on the emulsion which have received different exposures (W-10). These effects, called adjacency effects, are found, despite agitation, at sharp
boundaries between exposed and unexposed regions. The completeness of development varies with time in a complicated manner. The simplest law, although only an approximation, states that $D = D_\infty (1 - e^{-kt})$, where $D$ is the density (discussed later) to which the image is developed in time $t$, and $D_\infty$ and $k$ are constants.

Following development, the film is rinsed briefly in the short stop bath, a dilute acetic acid solution which stops development quickly by lowering the pH. The undeveloped silver halide is then dissolved out in the fixer or "hypo", a sodium thiosulfate solution, to prevent future fogging of the film. Washing removes the fixer to prevent future staining. These last steps have little effect on the developed image, although if large temperature differences exist in the solutions the emulsion may become reticulated.

D. Sensitometry

Sensitometry is the science of measuring the sensitivity of photographic materials. To make the science precise, a number of concepts have been defined, and the relationships between them quantitatively expressed. The concept of sensitivity, intuitively clear enough, is defined rigorously a little later. Exposure ($E$) in the case of light, can be defined as the number of lumens or ergs incident on the emulsion per unit area. The spectral quality and exposure rate must be specified to make the information complete. In the case of β rays, exposure can be defined as the number of β particles incident per unit area, or as the energy dissipation per unit mass at the film surface. Again, the energy spectrum of the β rays is one of the significant attributes of the exposure, and the rate of exposure may be.

The photographic effect produced by a given exposure, followed by development, is usually expressed in terms of density ($D$). Density is defined as
\[ D = \log_{10} \frac{I_o}{I}, \]

where \( I_o \) is light intensity incident on the blackened area and \( I \) is light intensity transmitted. Three types of density are in use: (1) specular density, in which light is normally incident on the image, and only the normally transmitted light intensity is measured, (2) diffuse density, in which light is normally incident on the image but all (diffusely) transmitted light intensity is measured, and (3) doubly diffuse density, in which light is diffusely incident, and all transmitted light intensity is measured.

The photographic effect produced could also be defined as the number of developed grains or the number of particle tracks per unit area. A simple approximate relationship between density and number of developed grains per unit area can be easily derived. (A more rigorous treatment leads to the same result (3-8).) If \( \bar{a} \) is the average projection area of the grains, \( n \) is the number of developed grains per unit area, \( T \) is emulsion thickness, and \( x \) is distance measured into the emulsion, then the quantity of light absorbed by the grains in a thin layer of emulsion is

\[ dI = -I\bar{a} \frac{n}{T} \, dx. \]

If this is integrated over the emulsion thickness, one gets

\[ \int I \log_{10} \frac{I_o}{I} \, dx = D = 0.4343\bar{a}n. \]

Experimentally, density is measured directly or as the ratio \( \frac{I_o}{I} \) by means of a densitometer. This is merely an instrument for passing light through a photographic image, and measuring what fraction of the incident light is transmitted.

The relationship between density and exposure is the film property of primary significance in quantitative
photographic measurements. This relationship is usually expressed graphically, sometimes as a plot of D vs. E, but more commonly as D vs. logE. This latter curve is called the characteristic curve, or H and D curve (after its originators, Hurter and Driffield). Simple derivations of representative H and D curve shapes, under various conditions, will be made in the next chapter.

With the H and D curve in mind, one can make clear certain useful concepts. Sensitivity can be defined in various ways, depending on the nature of the particular problem. For our work, it will be most useful to define it as the reciprocal of the exposure required to produce a specified net D (D above background). Two definitions of sensitivity are used in the following work: (1) particle number sensitivity, defined as the reciprocal of the number of electrons/cm\(^2\) required to produce a net D = 0.3, and (2) dosage sensitivity, defined as the reciprocal of the dosage (in units of \(\frac{100 \text{ ergs}}{\text{gm}} \approx 1 \text{ rep}\)) necessary to give a net D equal to 0.3. The gamma (\(\gamma\)) of the film is defined as the slope of the central straight line portion of the H and D curve. Contrast is defined, for our present purposes, as the slope of the curve at the D in question. The \(\gamma\) is therefore identical with the maximum contrast. High contrast is usually desired in quantitative \(\beta\) detection with film in order that the difference in density produced by 2 sources of almost equal activity be as large as possible. Latitude is defined as the logE interval corresponding to the straight line segment of the curve. These concepts will all be used later.

E. Resolving Power

It has previously been stated that high resolving power is one of the important attributes of film in \(\beta\) ray dosimetry measurements. Although it has not been investigated in this research under the assumption that it is suf-
sufficiently high for most purposes, resolution is briefly discussed here. Like most of the other photographic concepts, resolving power is defined in several ways, depending on both the type of radiation detected and the nature of the image. For light it is often defined rather unambiguously as the number of lines per mm which can be distinguished on the film. For β ray detection, the film resolving power might be characterized in terms of the size of the image formed by a point source of activity at the emulsion surface. However, in practice one is always concerned with the resolving power of the source-film system, which cannot take full advantage of the resolving power of the film itself because of a gap between source and film. This is considered in a little more detail in the next chapter.

Limitations are set on the resolving power of the film itself by granularity and by adjacency effects. Granularity refers to the discrete character of the silver deposit in the image, caused both by clumping during development and by the initial distribution of the silver halide in microscopic grains. Adjacency effects of development cause an effective overlapping of one part of the image by another. They can extend in distance as much as a few tenths of a millimeter in some emulsions.
In the second chapter the basic interactions of β rays with matter were discussed, and in the fourth chapter the fundamental principles of the photographic process were briefly surveyed. In this chapter we will make use of many of these ideas to treat quantitatively certain pertinent problems in the photographic action of β rays.

A. H and D Curve

The nature of the H and D curve is of much importance in the quantitative detection of β rays, as it determines the fundamental properties of sensitivity and contrast. This curve depends in part on development time, as has been mentioned, and on other development conditions. In this section we shall investigate how it may depend also on grain size and size distribution, on grain sensitivity and the amount of energy dissipation per unit path length along the electron track, and on absorption of the β rays by the emulsion. curves of D vs. E will also be discussed. In the literature may be found many attempts to derive theoretically the shape of H and D curves. Webb (W-2), for example, lists 20 papers treating the subject from a variety of approaches. Some of these are referred to in the following discussion.

1. Dependence of H and D curve on grain sensitivity and size

It is possible to derive several simple but instructive relationships between density and exposure under certain limiting assumptions of grain sensitivity and size. We have already seen that under simplifying conditions,

\[ D = 0.434 \bar{a} n \]

where \( n \) is number of developed grains per unit area and \( \bar{a} \) is average projection area per grain. The
remaining problem is to find \( n \) as a function of exposure. In deriving these relationships, the following symbols will be used:

\[
\begin{align*}
E &= \text{exposure in terms of number of electrons incident per cm}^2.
N &= \text{number of grains/unit area in undeveloped film}
n &= \text{number of developable or developed grains}
n_0 &= \text{number of grains not hit at all}
n_1 &= \text{number of grains hit once and only once}
\bar{a} &= \text{average projection area/grain}
\end{align*}
\]

The general assumptions are first made that the electrons pass through the emulsion perpendicular to its surface without absorption or deviation, and that the grains do not change size or shape during development. The first of these two assumptions is later removed.

\( \text{a. Uniform grain sensitivity and size} \)

It is assumed throughout this section that the grains have uniform sensitivity and size. Curves are derived under the three conditions of 1 hit, 2 hits, and 40 hits, respectively, required to make a grain developable.

When it is further assumed that the grains have high enough sensitivity so that a single hit is sufficient to make each one developable, the following expressions show the relationship between \( D \) and \( E \):

\[
\begin{align*}
dn &= (N - n) a dE \\
D &= 0.434aN(1 - e^{-aE}) = D_m(1 - e^{-aE}) \\
D &= 0.434aN_{\alpha}E = D_{m\alpha}E \text{ for low } D.
\end{align*}
\]

Equations (10) or (11) are often referred to as Silberstein's formula. They were originally derived by Silberstein (S-7) for light exposures, but were soon found experimentally to
be rather inaccurate. $D_m$ is seen in (11) to be the maximum density obtainable with the particular emulsion.

Eq. (11) is plotted in Fig. 16 with $D/D_m$ as a function of $\log aE$ (giving the $H$ and $D$ curve) and in Fig. 17 with $D/D_m$ as a function of $aE$. The sensitivity is approximately equal to $3.3\ D_a$. The contrast, $dD/\text{d}\log E$, is given by $2.3D_m aE^{-aE_m}$, or at low $D$, by $2.3D$. The maximum contrast is $0.84D_m$.

If it is assumed that 2 hits are necessary and sufficient to render a grain developable, one finds a different relationship between $D$ and $E$:

$$dn_0 = -an_0 \text{d}E \quad (12)$$
$$dn_1 = an_0 \text{d}E - an_1 \text{d}E \quad (13)$$
$$dn_2 = an_1 \text{d}E \quad (14)$$

On solving (12) and (13) for $n_1$, and making use of the initial condition $n_1 = 0$ at $E = 0$, one finds

$$n_1 = aNE e^{-aE} .$$

When this is substituted into (14) and the integration is performed, it is found that

$$D = 0.434aN\left[1 - (1 + aE)e^{-aE}\right]$$
$$= D_m\left[1 - (1 + aE)e^{-aE}\right]$$

$$D = 0.434aN(aE)^2 = D_m(aE)^2 \text{ for low } D \quad (15)$$

$D/D_m$ from eq. (15) is plotted in Fig. 16 as a function of log $aE$ and in Fig. 17 as a function of $aE$. The shape of these curves differs from those for the one hit theory both in the higher maximum contrast and in the initial bend in the $D/D_m$ vs. $aE$ curve. The sensitivity is approximately equal to $\sqrt{D_m/0.3}$. It should be borne in mind, when comparing this with the sensitivity of the single hit emulsion, that sensitivity has been defined in terms of the ex-
Theoretical H & D Curves

Figure 14
Single Hit
Single Grain Size
Exponential Absorption in Emulsion

\[ \alpha = 1.000 \]
\[ \alpha = 0.398 \]
\[ \alpha = 0.158 \]
\[ \alpha = 0.100 \]

Figure 15
Single Hit
Single Grain Size
Exponential Grain Size
Exponential Absorption in Emulsion

\[ \alpha = 1.000 \]
\[ \alpha = 0.398 \]
\[ \alpha = 0.158 \]
\[ \alpha = 0.100 \]

Figure 16
Single Hit
Exponential Grain Size
Single Hit
Uniform Grain Size
Double Hit
Uniform Grain Size

40 Hit, Exponential Grain Size
40 Hit, Uniform Grain Size
posure required for net $D = 0.3$. If one were interested in some other density (as one usually is), the relative sensitivities of the two emulsions would be different. The contrast is given by $2.3D_m(aE)^2e^{-aE}$, having a maximum value of $1.24D_m$.

It would be possible, if desired, to derive the density-exposure relationship for any specified number of hits required to make a grain developable. However, we will now consider just one other special case, namely, an emulsion requiring 40 hits per grain to produce developability. This might correspond to the number of light quanta hits necessary in the case of a typical emulsion (W-4).

A rigorous analytical solution for the dependence of $D$ on $E$ would of course be rather complicated. Therefore a simpler approximate method is used which is sufficiently accurate for present purposes. This involves a statistical treatment of the problem, in which from probability theory one can compute the number of grains which will receive 40 or more hits for any specified number of average hits per grain ($= aE$). The probability law used has been the simple normal distribution law (with $\sigma^2 = aE$), rather than the Poisson law, an approximation of little significance here. The results are shown, in the same form as the previous curves, in Figs. 16 and 18. Although analytic expressions are not available for the sensitivity and contrast, it can be seen from the figures that the sensitivity is much lower and the contrast much higher than for the previous theoretical $H$ and $D$ curves. The maximum contrast, $\gamma$, as determined from the graph, is $5.2D_m$.

The 3 sets of curves just derived show how sensitivity and contrast depend on the emulsion properties under the very restrictive conditions of uniform grain size and sensitivity. The results are useful in illus-
trating basic principles, but they are not so useful in
practice for the reason that commercial emulsions contain
grains of widely varying sensitivity and size. In the lit-
erature many theoretical derivations of H and D curves have
followed reasoning similar to the above. Often it has been
attempted to deduce from these theories, combined with ex-
perimental H and D curves, the number of light quanta hits
required for developability of a grain. The results usu-
ally indicate a small number (1 to 4), although it may be
known that many more light quanta fall on the average
grain. As Webb (W-3) points out, the calculation of a
small number is always found if one assumes a uniform grain
size and sensitivity, since in practical emulsions contrast
is not sufficiently high to fit a curve based on a large
number of hits per grain. We see in the next section how
grain size variation decreases the contrast.

b. Exponential grain size

In order to make the theory somewhat more accur-
ate, we may investigate the next most general case of an
emulsion having uniform grain sensitivity but a grain size
distribution more similar to that of \[ N(a) = \frac{N}{e^{-a/E}} \]
discussed in Chapter 3. We now derive the relationship be-
tween D and E under 2 different conditions of (uniform)

grain sensitivity: 1 hit per grain and 40 hits per grain,
respectively, required to render a grain developable.

In the single hit case, it can be considered that
the problem is one of summing over an infinite set of grain
size classes, each populated according to the exponential
function of \( a \) just referred to. The number of grains in
each class, \( N(a)da \), contributes when completely developed,
a density \( dD \) taken from eq. (11):

\[
dD = 0.434aN(a)da(1 - e^{-\frac{aE}{E}}) .\]
On integrating over the grain size, one gets:

\[
D = 0.434 \alpha E \left[ 1 - \frac{1}{(1 + \alpha E)^2} \right] = D_m \left[ 1 - \frac{1}{(1 + \alpha E)^2} \right]
\]

\[
D = D_m \alpha E \quad \text{for low } D.
\]

The \(D/D_m\) vs. \(aE\) curve, and the \(H\) and \(D\) curve, are shown in Figs. 17 and 16. The sensitivity is given approximately by \(6.6D_m\). The contrast equals

\[
\frac{4.6D_m \alpha E}{(1 + \alpha E)^3},
\]

the maximum contrast being \(0.69D_m\). The significant features of this theory are that the density is still proportional to exposure at low exposure, as in the single grain size theory, but that the contrast is now lower.

In the 40 hit, exponential grain size theory, the method for finding \(D\) as a function of \(E\) is the same in principle as above, except that the solution must be carried out graphically. My results are shown in Figs. 16 and 18. The outstanding conclusions are that the contrast is lower and more constant over a wide range of \(D\) than in the single grain size theory, and that the initial bend in the \(D\) vs. \(E\) curve is not nearly as pronounced. It is apparent that the introduction of grain size variation into the theory has made the \(H\) and \(D\) curves of the 40 hit and the single hit theories much more similar.

To make these theories more general still, we should now introduce grain sensitivity variation into the treatment. Webb (W-3) has done this by graphical methods, making use of actual grain size and sensitivity data made available to him by Eastman for 3 different emulsions. With one adjustable constant for sensitivity variation, he was
able to get very close agreement with experimental curves over the whole density range. For his treatment the reader is referred to his paper. To make this type of analysis worthwhile here, however, we would require more explicit information on sensitivity and size variation than the emulsion manufacturers care to divulge. A qualitative picture of the effect of grain sensitivity variation can be obtained by imagining an H and D curve which is the sum of several uniform sensitivity H and D curves, based on 1 hit, 2 hit, etc. sensitivity, respectively. The effect on contrast would be the same as the effect of nonuniformity in grain size: contrast would be decreased. In quantitative β ray detection with film, the effect of grain sensitivity on H and D curve shape is of particular interest, as is discussed later.

To summarize the results of the foregoing analyses, the important derivations, together with those from the next section, are brought together in Table 9.

2. Dependence of H and D curve on absorption in emulsion

One of the assumptions in all of the preceding theories was that there was no absorption of the β rays in the emulsion. However, it has been pointed out in Chap. 2 that low energy β rays are absorbed appreciably in thicknesses of matter corresponding to that of typical emulsions (a few mg/cm²). In this section we consider the effect on the H and D curve shape of absorption in the emulsion.

This effect can be qualitatively understood by means of the following simplified example. If incident electrons were to penetrate just half way through the emulsion, but no further, the maximum density obtainable with the emulsion would be just half as great as for full penetration. Consequently, sensitivity and contrast would
also be lower, as is evident from the dependence of these expressions on $D_m$. Charlesby (C-3) has developed a detailed theory of the effect of exponential absorption on the H and D curve shape in the single hit, variable grain size, situation. (We have already seen that the exponential law is frequently an accurate description of the absorption of $\beta$ rays.) Charlesby's method is similar to the approach used for the single hit theory in the preceding section, except for the further complication brought in by the absorption. The final H and D curves are obtained by a rather simple form of graphical integration. For a complete study of his theory, the reader is referred to the original paper.

Using Charlesby's theory, I have computed several H and D curves for representative degrees of absorption in both single grain size and exponential grain size emulsions. The degree of absorption is characterized by the transmission, $a$, defined as the fraction of the incident number of electrons which penetrates completely through the emulsion.

The single grain size, single hit theory predictions are shown in Fig. 14, which contains four H and D curves having $a = 1.000$, $0.398$, $0.158$, and $0.100$, respectively. The outstanding information from these curves is that both the sensitivity and the contrast decrease with $a$, although the contrast decreases by a surprisingly small amount. This means that on grounds of absorption alone, one would not expect an important dependence of H and D curve shape on electron energy until the electrons have such low energy as to be almost completely absorbed in the emulsion. The $D/D_m$ vs. $AE$ curves, if plotted, would be similar to the single hit curve of Fig. 17, having an initial slope of $\frac{a - 1}{2.3 \log a}$. Table 9 gives the sensitivity and maximum contrast ($\gamma$) for these curves.
The exponential grain size, single hit theory, is illustrated by the H and D curves of Fig. 15 for the same values of \( a \). A similar dependence of contrast and sensitivity on \( a \) is observed as in the case of the single grain size theory, although contrast is affected less. The \( D/D_m \) vs. \( \bar{E} \) curves would be similar to that of Fig. 17 for the same conditions without absorption, but with an initial slope of \( 2(\frac{a-1}{2.3 \log a}) \). Table 9 brings together a summary of all these facts.

Table 9.

Theoretical Density-Exposure Relationships

<table>
<thead>
<tr>
<th>Number of hits per grain</th>
<th>Grain size distribution</th>
<th>Transmission</th>
<th>Initial slope of ( D/D_m ) vs. ( \bar{E} )</th>
<th>Sensitivity</th>
<th>Maximum contrast (( \gamma ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Uniform</td>
<td>1.000</td>
<td>1.000</td>
<td>3.33(D_m\alpha)</td>
<td>0.84(D_m)</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.398</td>
<td>0.655</td>
<td>2.18(D_m\alpha)</td>
<td>0.75(D_m)</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.158</td>
<td>0.458</td>
<td>1.53(D_m\alpha)</td>
<td>0.70(D_m)</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.100</td>
<td>0.391</td>
<td>1.30(D_m\alpha)</td>
<td>0.65(D_m)</td>
</tr>
<tr>
<td>1</td>
<td>Exponential</td>
<td>1.000</td>
<td>2.000</td>
<td>6.67(D_m\alpha)</td>
<td>0.69(D_m)</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.398</td>
<td>1.31</td>
<td>4.37(D_m\alpha)</td>
<td>0.69(D_m)</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.158</td>
<td>0.916</td>
<td>3.05(D_m\alpha)</td>
<td>0.61(D_m)</td>
</tr>
<tr>
<td>1</td>
<td></td>
<td>0.100</td>
<td>0.782</td>
<td>2.61(D_m\alpha)</td>
<td>0.59(D_m)</td>
</tr>
<tr>
<td>2</td>
<td>Uniform</td>
<td>1.000</td>
<td>0</td>
<td>3.3(D_m\alpha)</td>
<td>1.24(D_m)</td>
</tr>
<tr>
<td>40</td>
<td></td>
<td>1.000</td>
<td>0</td>
<td>Low</td>
<td>5.2 (D_m)</td>
</tr>
<tr>
<td>40</td>
<td>Exponential</td>
<td>1.000</td>
<td>0</td>
<td>Low</td>
<td>1.2 (D_m)</td>
</tr>
</tbody>
</table>

The foregoing analyses point up the effect of many emulsion variables on the relationship between \( D \) and \( E \). To some extent the theoretical results may be used to deduce information from experimental data on the number of hits, and hence on the energy dissipation, required to make
a single grain developable. In particular, one may be rather confident that a sizable fraction of single hit events occur if the $D$ vs. $E$ curve is a straight line, at low $D$, passing through the origin. However, because of the large number of variables, most conclusions drawn from the $H$ and $D$ curve shape alone are to be considered somewhat tenuous.

B. Energy Dissipation per Photographic Grain Traversed

Curves have been shown in Chap. 2 of the dependence of rate of energy loss on the energy of the electron. In the preceding section of this chapter it has been demonstrated that the $H$ and $D$ curve shape is a function of the number of hits per grain required for developability, i.e., of grain sensitivity. Putting these 2 considerations together, one draws the conclusion that, in general, the $H$ and $D$ curve shape should be a function of the energy of the exposing electrons. In this situation, the relative sensitivity (reciprocal of number of electrons/cm$^2$ required for a specified density) of a particular emulsion for electrons of different energies would be a function of density. Hence the quantitative description of the photographic effect of $\beta$ rays could become quite complicated. To be sure, we have seen that the conclusion regarding the dependence of $H$ and $D$ curve shape on electron energy is modified somewhat by certain considerations. In the first place, the $H$ and $D$ curve shape would be independent of energy for an emulsion having grains so sensitive that a single hit of even a minimum specific ionization electron would produce developability. Also, it has been shown how the presence of grain size variation makes the $H$ and $D$ curve of a many hit emulsion quite similar to that of a single hit emulsion, except at low densities. However, the fact remains that some differences in $H$ and $D$ curve shape are to be anticipated with different energy electrons.
It becomes useful, therefore, to consider the quantity of energy dissipated by an electron traversing a silver halide grain. The pertinent information is readily available from Chaps. 2 and 4. The size of most AgBr grains lies in the range between perhaps 0.1 to 2 microns diameter. Using Table 7 and a density of AgBr = 6.5 gm/cm$^3$, it can be seen that a fairly large fraction of an electron's energy is dissipated in delta rays of energy greater than say 5 kev. In the case of the 1 Mev electron, for example, this fraction is almost 30%. If it is assumed from Fig. 5 that all delta rays of energy less than roughly 20 kev expend their energy in a single $\mu$m grain, then again for the 1 Mev electron, about 10% of its total dosage would occur in delta rays of 5 to 20 kev within a single $\mu$m grain. These figures show in a semi-quantitative way how the energy dissipation can be clumped in individual grains, and therefore how many single hit exposures might occur even when average energy dissipation per grain is insufficient for exposure.

C. Resolution

Calculations have been made by several investigators on the resolution obtainable under various $\beta$ ray autoradiographic conditions. Two useful papers to which the reader is referred for full details are those of Doniach and Pelc (D-3) and Gross, et al. (G-5). These calculations are very useful in estimating the relative importance, for good resolution, of certain geometrical parameters in the source-film system, namely, emulsion and source thickness, and width of gap between source and film. They do not, however, deal with $\beta$ ray scattering nor with granularity of image. A representative set of results obtained by Gross and co-workers is given in Table 10. The calculations were for exposures by a source having the form of a line perpendicular to the surface of the tissue sec-
tion being autoradiographed. Resolution in this calculation is defined as the radius of the circle on the film which is the locus of points receiving \( \frac{1}{2} \) the exposure given to the center of the exposed area.

Table 10

Theoretical Resolution Conditions

<table>
<thead>
<tr>
<th>Emulsion thickness (( \mu ))</th>
<th>Gap (( \mu ))</th>
<th>Section thickness (( \mu ))</th>
<th>Resolution (( \mu ))</th>
<th>Maximum exposure</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>0.1</td>
<td>5</td>
<td>2.3</td>
<td>3.2</td>
</tr>
<tr>
<td>5</td>
<td>0.1</td>
<td>10</td>
<td>2.8</td>
<td>3.5</td>
</tr>
<tr>
<td>10</td>
<td>0.1</td>
<td>5</td>
<td>2.7</td>
<td>3.5</td>
</tr>
<tr>
<td>10</td>
<td>0.1</td>
<td>10</td>
<td>3.7</td>
<td>3.9</td>
</tr>
<tr>
<td>15</td>
<td>0.1</td>
<td>5</td>
<td>3.3</td>
<td>3.7</td>
</tr>
<tr>
<td>15</td>
<td>0.1</td>
<td>10</td>
<td>4.4</td>
<td>4.1</td>
</tr>
<tr>
<td>5</td>
<td>0.5</td>
<td>5</td>
<td>5.7</td>
<td>1.8</td>
</tr>
</tbody>
</table>

Two features of this table might be emphasized. First, the width of the gap between film and section is perhaps the most important single parameter affecting resolution. Second, a decrease in resolution automatically results in a decrease in maximum exposure, so that quantitative measurements may be limited in their accuracy by poor resolution.

As is true with all physical measurements, resolution and sensitivity are mutually exclusive in the photographic detection of electrons. This is the case for 2 reasons. The first is that high resolution requires small grains, and these are in general less sensitive than larger grains due to the lower energy dissipation per incident electron during its shorter path within the smaller grain.
The second reason is more fundamental, as it results from the statistical character of the electron exposure. Suppose it is desired to measure quantitatively an exposure of $10^6$ electrons/cm$^2$ received by a small area of film of $10^{-4}$ cm$^2$. (Instruments and films exist which would permit this except for the statistical limitation being discussed.) On the average, a $10^{-4}$ cm$^2$ area would receive only 100 hits, with a probable error of $67/\sqrt{100} = 6.7\%$. One could get greater accuracy either by decreasing the resolution requirements or by increasing the exposure. If by sensitivity one meant the exposure required for a given statistical accuracy, sensitivity would decrease in proportion to the decrease in area being densitometered. This shows the fundamental antagonism between sensitivity and resolution which even a "perfect" film could not circumvent.
CHAPTER 6: PRESENT KNOWLEDGE ON ELECTRON DETECTION WITH FILM

Having examined the nature of the interaction of β rays with matter, and of the response of photographic film to β rays, we are in a position to determine just what kind of experimental information is needed in order to permit the practical use of film in β ray dosimetry. Then, in the light of these conclusions, the present state of experimental knowledge on the subject of electron detection with film is reviewed.

A. Experimental Information Required

As in any quantitative work with photographic film, it is essential to establish experimentally the relation between density and exposure, in terms either of the H and D curve or of an equivalent function. This permits the translation of density, which is read from the film, into exposure in relative units, which is either the quantity sought or else is a halfway step toward the final quantity, absolute exposure. When one is detecting electrons of more than a single energy, as is usually the case, it is essential to know specifically how the shape of the H and D curve depends on energy. Should it be the case that the H and D curve is substantially independent of energy, the relative sensitivity of the film for electrons of different energies is independent of density, a considerable simplification.

One must also know how the sensitivity of film depends on a variety of exposure and processing conditions. (When the term "sensitivity" is used throughout the remainder of this chapter, it will mean, unless otherwise specified, the particle number sensitivity, i.e., the reciprocal of the number of electrons required per cm² to give a net D = 0.3. Particle number sensitivity is more convenient to use than dosage sensitivity when referring to experimental
work reported in the literature.) One of the chief ques-
tions is the dependence of sensitivity on $\beta$ ray energy, for
$\beta$ ray energy is often an uncontrollable (and perhaps even
unknown) variable in practical dosimetry. This problem,
and that of $H$ and $D$ curve shape, could be understood in
somewhat more absolute terms if one knew the energy dissi-
pation per grain required to bring about developability;
therefore this becomes a question of interest. Another
problem is the determination of the effect of the angle of
incidence of the $\beta$ rays on sensitivity. Because of finite
emulsion thickness, with attendant $\beta$ ray absorption and scat-
tering, one can expect this effect to be a function of ener-
gy. It is also essential to establish the dependence of
sensitivity on rate of exposure, i.e., to determine whether
the reciprocity law is invalid, and whether latent image
fading is important over long periods of exposure. If nei-
ther of these is the case, comparison of the photographic
effect of exposures carried on at different rates (e.g.,
especially in the use of calibration sources) is greatly
simplified. It is much easier to vary exposure time quant-
itatively than it is to vary intensity. It can be expected,
in view of the chemical and physical processes that take
place in latent image formation, that sensitivity will be
affected by the temperature and moisture content of the
emulsion at the time of exposure. If so, knowledge regard-
ing these effects would be advantageous. The composition
and temperature of developer, and the time of development,
will of course be significant variables influencing sensi-
tivity.

Another problem of much practical importance is
the determination of the effect of absorption by matter on
the exposing efficiency of the continuous $\beta$ ray energy spec-
trum. The effect of absorption on the continuous spectrum
has already been discussed in the chapter on the interac-
tion of $\beta$ rays with matter. In the light of that discus-
sion, one can expect that the sensitivity of a particular film (for the $\beta$ rays which reach the film surface) will be substantially independent of the amount of absorption which has been imposed upon them. If this were found to be true, it would be of importance, as has been mentioned, in the accurate calibration, with known sources, of $\beta$ ray exposures from sources having unknown amounts of self absorption.

Resolution has been mentioned as one of the prime assets of film as a dosimeter. Wherever there is a question as to the sufficiency of the resolving power of a specific film in a specific application, the resolving power should of course be known.

Artifacts in the photographic process will lead to the misinterpretation of dosimetry measurements with film. Their presence and cause should be known and understood.

Finally, one is interested in the overall reproducibility of the photographic dosimetry technique under practical conditions.

B. Survey of Previous $\beta$ ray and Electron Detection with Film

In this section we review previous work on the detection of $\beta$ rays and electrons with film. After a brief historical survey, we take up the questions posed in the preceding section and attempt to systematize the currently available information bearing on them. It seems advisable to examine the field in considerable detail; therefore the reader interested in only the cardinal points might well skip to the summary at the end of the chapter. On the other hand, I need hardly state that this literature survey contains many omissions; some of these are intentional, while others, no doubt, are inadvertent.
1. Historical

Perhaps the first detection of $\beta$ rays with photographic emulsions was carried out, unknowingly, by Niepce de Saint-Victor (N-4) in 1867. In studying fluorescence, he shone light through various colored glasses onto many materials, then exposed them to AgI or AgCl impregnated paper. Among these materials was a uranium compound, which, however, behaved like the other materials in that it was activated by blue, indigo, or violet light, but not by red, orange, or yellow. The uranium compound, as well as certain non-radioactive compounds, maintained its photographic action for several months when sealed in a tin box. Whether or not Niepce de Saint-Victor was himself guilty, I have herewith had fair warning against the "interpretation" of data in the remainder of this investigation.

Becquerel (B-4, B-5), as everyone knows, was either more honest or more lucky in 1896. While performing essentially the same experiment, he observed that no light at all was required to activate his uranium mineral. We know, in retrospect, that it was primarily $\beta$ rays which left the shadow of the thin metal plates on his photographic film.

Following Becquerel's discovery, many applications were made of film in the detection of radioactivity in various substances. Mme. Curie (G-11) studied a large number of minerals and metals, detecting activity in U, Th, and K compounds. Crookes (C-10) went through his entire mineral collection, and found that only U and Th ores were capable of exposing film. Others made similar investigations for the presence of naturally occurring radioactivity in minerals. Lacassagne and Lattes (L-1), who studied the deposition of Po in rabbit tissue, were probably the first to make biological applications of autoradiography. In 1938 Groven, Govaerts, and Gueben (G-6) with some pride
laid claim to the discovery of the photographic action of artificial β emitters, namely neutron irradiated iridium and P32. From this time on, especially since the coming of piles, autoradiography has found rapidly expanding use in many sciences, particularly biology and medicine. Reviews of biological autoradiography have been compiled by several investigators (G-2, G-5).

The study of cathode rays and x ray ejected electrons was also aided from the beginning of the century by taking advantage of their photographic action. Innes (I-1), for example, reported a typical study of the velocity of x ray photoelectrons with a magnetic spectrometer and film. Mark and Wierl (M-3), another example among many, used film in investigating electron diffraction. The advent of the electron microscope (R-3) greatly increased the amount of work being done in the photographic detection of electrons.

Along with these many scientific applications of photographic film in the detection of β rays and electrons came a need for a deeper understanding of the photographic action of these particles. Studies were therefore started on the reciprocity law (B-20), the H and D curve shape (B-20), the variation of sensitivity with particle energy both at low (S-4) and high (E-4) monokinetic energies and at various maximum energies of continuous spectra (C-4). This deeper understanding, in turn, led to quantitative applications of photographic emulsions. Among these were the determination of shape of β ray energy spectra (E-4), and more important for the present research, measurement of the concentration of radioactive isotopes in biological tissues (A-3, D-4, S-11, B-22).

2. Scientific findings

A great many of the findings reported in the literature are of limited value for the reason that they apply
to films not currently available or to incompletely specified processing conditions. In the following review, greatest detail is given to the results which I consider, for one of several reasons, to be most pertinent to present day needs.

a. Shape of $H$ and $D$ curve, and its dependence on energy

A great many men have studied the relationship between density and exposure, as this is the simplest to investigate and perhaps the most useful of all the photographic properties. In general, a very wide variety of $H$ and $D$ curve shapes has been reported. This is not in the least surprising, in view of the large number of variable emulsion properties which determine the shape. However, one widely observed property is that the $D$ vs. $E$ plot has a slope which is finite at the origin, and decreases at higher exposures. Out of more than a dozen papers giving information on this point, to choose somewhat at random, (B-2, B-3, B-15, B-20, C-3, C-7, D-4, E-3, L-2, N-1, S-1, S-4, T-1), all agree within the experimental error (which is often rather large). It is somewhat surprising that Weidner and Nacken should find this, because at the low energies these workers used, the chief exposing agent is thought to be fluorescent radiation, as mentioned later. This should give the $D$ vs. $E$ relationship of light, which has essentially zero slope at the origin (multi-hit theory). Experimental error would mask a certain amount of deviation from the observed finite slope.

At low energies (less than about 50 kev), there is general agreement that the shape of the $H$ and $D$ curve depends on energy. Seitz and Harig, Nacken, and Schaffer (S-2), for example, found that at a certain density, whose magnitude is roughly proportional to energy, the $H$ and $D$ curve reaches a plateau, while at considerably greater exposures
it begins to rise again. They attribute the plateau to the fact that at these low energies the electrons are capable of exposing only the front of the emulsion, without penetrating all the way through it. Therefore as energy increases, penetration increases and consequently plateau height increases. At high exposures, they suggest that fluorescence or bremsstrahlung excited in the front of the emulsion by the electrons is capable of exposing the remainder of the grains in the rear. Others have found the same plateau height dependence on energy without carrying exposures high enough to observe the subsequent rise.

Above about 50 kev there is general agreement that the shape of the H and D curves is the same, at least up to rather high densities, independent of energy. This is reported by Baker, Ramberg, and Hillier, by Ellis and Wooster, and by Langendijk and Ornstein. However, Rudenberg, Cowing and Spalding claim to have found rather different H and D curves for I\textsuperscript{131} (0.6 Mev, max.) and P\textsuperscript{32} (1.7 Mev, max.). The accuracy of this anomalous result is open to doubt.

b. Sensitivity of film as a function of electron energy

The dependence of relative sensitivity on electron energy has been investigated by many workers. Most of the currently available data are for energies below 100 to 200 kev, a range of particular interest for electron microscopists, and for perpendicularly incident electrons. In the following discussion, perpendicular incidence is to be assumed unless otherwise specified.

The lower energy limit of detectability of electrons is below 50 volts. At these low energies the penetration of the electrons into the emulsion is of course negligible (or non-existent if a "T coat" covers the emulsion), so that one would expect to account for photographic action in terms of fluorescence excited by the electrons. Good
evidence for this has been deduced by Cole (C-5), Carr (C-1), and Burroughs (B-25), among others. They showed that the photographic plates were just about as sensitive when the electrons were incident on the glass backing as on the emulsion. Also, the lower limit of detectability depended on the kind of vapor in the region of the emulsion surface. Finally, sensitivity greatly increased when the film surface was smeared with grease, and under these conditions the fluorescence was visible. The sensitivity was so low that roughly $10^{12}$ electrons/cm$^2$ were necessary to give a practical density.

Above the lower energy limit the sensitivity increases roughly in proportion to energy up to 50 or 100 kev where it reaches a peak. Among the many papers illustrating this (B-2, B-3, B-15, C-7, L-2, N-1, S-2, S-4, A-1, B-14, C-9), one might cite that of Borries which was a study of 11 different films and showed good proportionality between sensitivity and energy for at least some of them. Becker and Kiphan found sensitivity proportional to energy only above 25 kev. Granberg and Halpern in a careful study of Ilford B2 and Eastman No Screen found for the Ilford emulsion, between 4 and 45 kev, that sensitivity $= KU^{1.45}$, where $K$ is a constant and $U$ is energy. There was no T coat on the Ilford emulsion. The 1p T coat over the No Screen emulsion complicated its behavior, making it less sensitive, relative to the Ilford emulsion, at the low energies. In all films this initial increase of sensitivity with energy is to be ascribed to the increasing penetration of the emulsion by the electrons.

At higher energies of 50 to 100 kev, a peak sensitivity is reached. The decrease in sensitivity beyond this energy occurs for two reasons. First, the higher energy particles have lower specific ionization; second, their path length in the emulsion is shorter than at the energy
of peak sensitivity because they pass all the way through the emulsion with less scattering. Borries (B-15) has shown a graph illustrating the influence of emulsion thickness on the energy of peak sensitivity. He plots this energy as a function of emulsion thickness, and compares it with a curve for the minimum energy required for the complete penetration of the emulsion as a function of emulsion thickness. The experimental points lie roughly 50% above the energy-range curve, as a result of the fact that most electrons, due to scattering, do not penetrate a distance equal to the maximum range for that energy. However, both the experimental points and the energy-range curve vary with energy in the same way.

The sensitivity of film for monokinetic electrons of high energies (above 0.1 or 0.2 Mev) has been investigated relatively little. Ellis and Aston (E-4), in an early study using monokinetic \( \beta \) rays from a spectrometer, examined an X-ray emulsion in the range between 0.25 and 2 Mev. They found that the sensitivity decreased continuously with increasing energy over this interval, at a rate somewhat greater than the specific ionization. The overall sensitivity decrease amounted to a factor of 3, as compared with about half that for specific ionization decrease. Blatz (B-10) studied the dosage sensitivity of film badges when exposed through about 20 mg/cm\(^2\) of absorber to monokinetic electrons. Under these conditions he found dosage sensitivity to be uniform in the range studied between 0.4 and 2 Mev. Arnoult (A-1), Langendijk and Ornstein (L-2), and Baker, et al. (B-2) have reported data up to about 0.25 Mev. They all show a decrease in sensitivity between the peak and 0.25 Mev amounting roughly to a factor of 2, whereas the specific ionization decrease in the same range is somewhat less.

In recent years a considerable amount of attention has been given to the sensitivity of various films
when exposed to continuous $\beta$ spectra of different energies, usually under conditions giving approximately diffuse incidence. The most inclusive paper is probably that of Steinberg and Solomon (S-12), who studied the response of 4 commercially available emulsions to the $\beta$ rays of $^{14}C$ (0.16 Mev), $^{45}Ca$ (0.25 Mev), $^{131}I$ (0.60 Mev), and $^{32}P$ (1.70 Mev). The angular distribution of the incident electrons was somewhere between perpendicular and diffuse. In Eastman No Screen X ray Film, an example among the emulsions studied, they reported that sensitivity decreased with increasing energy from $^{14}C$ to $^{32}P$ by a factor of about 5. However, this figure is quite out of harmony with the pertinent data described above (and with my own reported in this thesis). Inasmuch as the $^{32}P$ is capable of exposing both sides of the double emulsion film, while the $^{14}C$ radiation does not penetrate to the second side, one would expect the sensitivity ratio to be much smaller. Marinelli and Hill (M-1) found $^{32}P$ and $^{131}I$ sensitivity to be approximately the same for both Ansco No Screen X ray Film and Eastman Medium Lantern Slide Plates. The angular distribution of the $\beta$ rays was between diffuse and isotropic. Berriman, Herz, and Stevens (B-7) found an NT type nuclear emulsion stripping film to be 2.5 times as sensitive for $^{131}I$ as for $^{32}P$, under similar angular distribution conditions. Rudenberg, et al. (R-2) report that the sensitivity of duPont Dental Film No. 552-2, with probably 20 mg/cm$^2$ absorber present, was less than 1/3 as high for $^{131}I$ as for $^{32}P$. In their experiment the $\beta$ rays were incident approximately perpendicularly on the absorber surface. Various other fragmentary studies of this type have been made, but the results cannot be compared in any systematic fashion and are therefore relatively useless for our present purposes.

The preceding two problems would be considerably clarified if one knew the energy dissipation per grain required to render the grain developable. This information
could be combined with data on \( \beta \) ray absorption in matter to give a semi-absolute picture of the exposure process and its dependence on energy. Unfortunately, very little information exists on this subject, and most that does is out of date today. As an example, Borries (B-17) found for a rather widely differing group of emulsions that at the electron energy associated with peak sensitivity the energy incident on the film per developed grain ranged from 10 to 50 kev. If only half the mass of the emulsion is silver halide, approximately 5 to 25 kev energy dissipation in each grain was necessary for developability. If the effective grain diameter were a little over 1 \( \mu \), the grains of these various emulsions would require for developability, on the average, 5 to 25 hits from a minimum specific ionization electron. Delta rays and extra sensitive grains would contribute toward the existence of some single hit exposures.

In the last few years nuclear track plates have been produced that are capable of revealing individual tracks of electrons. Berriman (B-6) reported the development of an NT2a plate in which tracks of minimum specific ionization electrons could be detected. This indicates an appreciable fraction of single hit exposures in grains even as small as approximately 0.3 \( \mu \) diameter. The existence of single hit events, especially at lower energies, would account for the generally accepted finite slope of the \( D \) vs. \( E \) curve at low \( D \), discussed in the preceding section.

c. Dependence of sensitivity on angle of incidence

Almost no information is available on the variation of film sensitivity with the angle of incidence of the \( \beta \) rays. The only report I have found in the literature is that of Bothe (B-20). He exposed a piece of film wrapped around a glass rod to RaE \( \beta \) rays from a source several cm
from the rod. The β rays were incident perpendicular to the axis of the rod. Bothe found that the film blackening was quite uniform up to angles of incidence of about 50°, but fell off greatly thereafter. (He points out that if there were no scattering or absorption the blackening would be uniform all the way up to 90° (grazing) incidence.)

d. Dependence of sensitivity on rate of exposure

As a result of latent image fading or reciprocity law failure, the sensitivity of film may depend on the rate at which an exposure is made. A number of investigations have been made into the validity of the reciprocity law for β ray exposures. Most of these have been rather inaccurate, and apply only to limited conditions. However, there is rather wide agreement that the reciprocity law does hold with considerable accuracy over a rather wide range of exposure rates.

Bothe, using RaE, varied intensities by changing the distance between source and film in an evacuated chamber. In varying the exposure time between 4 minutes and 14 hours he found the Schwarzchild constant $p = 1.00 \pm 0.01$. Nacken (N-1) found approximately the same results for electrons of a few kev exposed over times ranging from a fraction of a second to hundreds of seconds. Becker and Kiphan (B-3) found $p = 1.00$ to about the same accuracy in the exposure time interval between a few and hundreds of seconds. Others reported $p = 1$ within experimental error at lower precision. At least two workers reported discrepancies from the reciprocity law. Salbach (S-1) claims to have found $p$ between 1.1 and 1.2 for sources of activity ratios in the neighborhood of 2. She concluded that the deviation from 1.0 were real although some of her results were not very consistent. Kovner (K-1) states that in several films investigated he has found the reciprocity law to hold at "low" and "high" intensities, but not at intermediate values. (I have been able to obtain only an abstract of this paper.)
Morgan (M-7) investigated the reciprocity law behavior for x rays, which in this consideration are identical with low energy electrons. In studying 4 x ray films, he found that \( p = 1.00 \), within the experimental error of less than 1%, for exposure times between 0.1 and 1000 seconds.

These results indicating the validity of the reciprocity law for electron exposures are not out of keeping with expectations. One would expect the law to be more nearly valid for electron exposures than for light exposures, because fewer hits per grain are required. In the limit of 1 hit per grain, the reciprocity law must be valid; in this case the formation of a latent image in any particular grain takes place instantaneously rather than throughout the whole time of exposure, and the length of the exposure time is of no significance.

Latent image fading during a prolonged exposure has the same effect as reciprocity law failure at low intensities, but is physically a somewhat different phenomenon. Little information seems to exist on the fading of latent images which have been produced by \( \beta \) particles. The best I have found is some unpublished data of Webb's (W-1) on nuclear track emulsions. These data show that fading of 190 kev x ray exposures in NTA, NTB, and NTB-3 (when stored at 20\(^0\)C and a relative humidity of less than 20%) can amount to a few percent per day initially and a fraction of a percent per day after a few weeks. Alpha particle exposures faded much less rapidly. The rate of fading increased with relative humidity, especially above 50%. In fact, storage of plates in an atmosphere of high relative humidity has been used by some (B-11) as a means of eradicating background before use. It can be expected that the rate of fading will increase also with temperature. It has been reported (L-3) that NTA plates (at unspecified humidity conditions) lose 10% of heavy particle tracks in 50 days when stored at 0\(^0\)C, 30% at 20\(^0\)C. Spence (S-10) believes that
humidity is a more significant factor in fading than is temperature. Yagoda (Y-1) has reviewed the literature on the fading of a particle exposures. He states that the fading is greatest in high gelatin content emulsions, and that it is worse for a particle exposures than for light exposures. From these facts he reasons that the fading may be caused by the action on the exposed grains of $H_2O_2$ which is produced by radiation in water.

e. Dependence of sensitivity on temperature and humidity at exposure

The physical condition of an emulsion at the time of exposure is known to have an effect on its sensitivity. Probably the most useful information comes again from Webb's unpublished data. In the case of $\beta$ rays on NTB-3, his figures show that sensitivity is rather constant below about 25 % relative humidity, but that at 50 % relative humidity it may be as much as 20 percent lower, and at 90 % relative humidity a factor of 5 lower. The effect, on sensitivity, of exposure temperature has been studied very little for ionizing particles. Dollman (D-2), however, reports that the grain density of proton tracks in Ilford C-2 (at 67 % R.H.) increases by about 30 % when the emulsion temperature rises from -66° C to 20° C, followed by a decrease of several percent at 70° C.

f. Dependence of sensitivity on development conditions

Development conditions which may affect the sensitivity of an emulsion are the following: type of developer, concentration, time, and temperature. There are many chemical varieties of developers, and their composition tends to influence film sensitivity in a rather unpredictable manner. An example of this may be found in a series of tests run by Borries (B-18), who used 5 different developers on each of 5 different films. The action of specific developers varied somewhat from film to film. In general,
developer concentration and development time bear an approximately reciprocal relationship toward each other. If the product, concentration x time, is constant, the degree of development will be rather similar. Both high concentration and long time tend to increase sensitivity, up to a limit when fog formation becomes excessive. The somewhat idealized density-time relationship of Chap. 4 is indicative of this: \( D = D_0 (1 - e^{-kt}) \). Elevated temperature has the same effect as increased concentration. However, the properties of the gelatin are such that development temperature should not be increased much above the normal 20° C.

It is conceivable that the effect of these different parameters could depend on the energy of the exposing \( \beta \) rays. The latent image is presumably more pronounced for a low energy \( \beta \) ray than for a high energy particle, and consequently \( k \) might be a larger number. Also, the diffusion time of the developer into the emulsion is appreciable in the case of thick emulsions; since low energy \( \beta \) ray exposures would be nearer the emulsion surface than high energy exposures, the effective \( k \) might again be a larger number.

e. Effect of absorption on \( \beta \) ray exposing efficiency

Little investigation has been directed toward studying the effect of the absorption of continuous energy spectrum \( \beta \) rays on their exposing efficiency (per unit dosage at the film surface). The only measurements of this sort that I have learned about are those of Blatz (B-9). He studied the surface dosage rate and the photographic exposure rate of a uranium metal source under various conditions of absorption, and found them to be closely proportional, independent of amount of absorption.
f. Resolving power

As has been previously mentioned, the degree of resolution which can be attained in autoradiography depends on many details besides emulsion properties. Chief among these are closeness of contact, source thickness, and β ray energy. In regard to the limiting resolving power of the film itself, it can be said, with certain limitations, that the behavior of the film with respect to light is a good indication of its behavior with respect to electrons. The resolving power of most photographic films for light, under typical processing conditions, is measured by the manufacturers. The resolving power of film for electrons, specifically, has been studied most extensively by electron microscopists, such as Borries (B-18). He showed that resolving power decreases slightly with increasing energy (at least below 100 kev), and with increasing development time (due to clumping of the grains). It also depends in a rather unpredictable manner on developer and development time. He found for the emulsions investigated that the minimum separation of 2 just resolvable points was in the range of roughly 20 to 100 μ. Pelc (F-2) prepared autoradiographs of rat thyroids containing $^{131}$I, and concluded that exposures from regions separated by 10 μ could be distinguished. Stevens (S-13) tested the resolution of the autoradiographic process under conditions giving approximately the maximum obtainable resolving power. Using specially prepared $^{131}$I sources in close contact with a very thin (4 μ) fine grained stripping film, he concluded that active points separated by 2 to 3 μ could be resolved. The data of Cobb and Solomon give a rough idea of the relative resolving power of a wide variety of emulsions commonly used today in autoradiography.

g. Reproducibility

The overall reproducibility found in the many
literature references varies a great deal from paper to paper, depending on the nature of the investigation and the care with which the worker performed the experiments. It might be said, however, that one is not surprised at absolute discrepancies between the results of different men which amount to 25% to 50%, nor at relative discrepancies in the work of a single investigator which amount to 10% to 15%. As generally used, film has not been a precision tool in quantitative measurements. However, one has a right to expect that the frequent use of calibration exposures, together with careful attention to experimental details, will permit somewhat more accurate results than has usually been reported.

h. Artifacts

Several sources of artifacts are known to be capable of giving misleading data both on the localization of exposures and on the quantity of exposure. First are the various development and adjacency effects mentioned in the chapter on photographic principles. Second, apparent exposures are produced by pressure on, or abrasion of, the film surface. These have been rather clearly demonstrated to be a result of point heating of the emulsion (M-4). They are most serious in those emulsions which do not have a protective T coat. Chemical actions of various types are known to produce photographic exposures. According to Yagoda, wood, for example, is capable of fogging certain emulsions, perhaps through the action of \( H_2O_2 \). Its use as a source mounting medium or as a film holder is undesirable. Boyd and Board (B-21) found both exposure and desensitization of NTB emulsions by the chemicals in various tissue sections.

1. Summary of previous knowledge and experiments required

The pertinent findings of previous workers in-
vestigating the photographic detection of electrons are summarized here. Conclusions are simultaneously drawn as to further work which can profitably be performed to permit more accurate or convenient use of photographic film in β ray dosimetry.

The shape of the H and D curve is substantially independent of electron energy above about 75 kev. However, it is a function of so many other conditions that it should be experimentally determined in each application.

The particle number sensitivity of film for perpendicularly incident electrons is zero at zero electron energy, rises rather linearly with energy to a peak at about 50 to 100 kev, and thereupon drops to a value considerably (factor of several) lower at energies greater than 0.5 or 1 Mev. The dosage sensitivity starts at zero, rises to a less pronounced peak at about the same energy, and falls off less at higher energies. More precise information on currently available film is greatly needed here.

The relative sensitivity of film as a function of the angle of incidence of the electrons has scarcely been studied at all. More data are needed, for almost all practical applications are made with diffusely incident β rays.

The energy input, from exposing electron, which is necessary to make a single grain developable is probably in the region of several kev, depending considerably on the emulsion. This means that for minimum specific ionization electrons at least a few hits per grain are required, on the average, to render a single grain developable. Information on this point would be useful for general understanding of the photographic action of electrons. It is not, however, a piece of data required in specific applications.
Reciprocity law failure is almost certainly not a serious problem in β ray exposures. Fading, however, is more serious in exposures of the order of days, and should be kept in mind in practical applications.

Sensitivity is probably not strongly dependent on emulsion temperature, in the range of normal room temperatures, but can be affected rather strongly by humidity conditions. An attempt should be made to control humidity.

Sensitivity is strongly dependent on development conditions. Consequently, these conditions should be standardized, or, as is probably more effective, they should be calibrated in each development batch.

The dependence of β ray exposing efficiency on the amount of absorption imposed on the β rays, as reported by a single investigator, is apparently not great, at least for high energy β rays. However, more extensive data on this question would be most useful in evaluating the reliability of data made quantitative by the use of calibration sources.

The resolving power of photographic film is sufficiently high for most β ray dosimetry measurements.

Artifacts do exist in photographic exposures. Their presence can often be detected by non-radioactive control exposures.

Reproducibility in the quantitative photographic measurement of electron exposures is not as high as could be desired. This emphasizes the need for care in making the measurements and for the wider use of calibration techniques.
CHAPTER 7: EXPERIMENTAL INVESTIGATION OF FUNDAMENTAL PROCESSES

The experimental data on fundamental processes obtained during the course of this thesis are to be described according to the following plan. The standardized film handling and processing techniques used generally throughout the experimental work are discussed first. Next, the study of the properties of several films for monokinetio electron exposures is presented. This is followed by an investigation of these same films under exposure to calibrated dosage sources of continuous spectrum \( \beta \) rays of several energies; emphasis is placed on the effect of absorbers on the exposing efficiencies of these \( \beta \) rays. The two sets of independent data on monokinetio and continuous energy spectrum \( \beta \) rays are then compared, as a check on these experiments. Finally, 2 short experiments on the reciprocity law and the accuracy of microdensitometry are described. A summary is given, at the end, of the major conclusions on the validity and practicality of the photographic \( \beta \) ray dosimetry technique. Much of the technical data on apparatus and techniques is given in appendices, where they do not disrupt the central chain of presentation of the results of the investigation.

A. Emulsion Composition

It has already been shown that many photographic properties depend on the emulsion thickness \((\text{mg/cm}^2)\) and on the silver halide content of the emulsion. Among these are two of the most fundamental properties, absolute sensitivity and variation of sensitivity with electron energy. It therefore appeared desirable to work out and apply to each film, or at least to each type of film, techniques for determining these quantities. The information so gained would serve two purposes: first, it would check one set of causes for possible fluctuations in data from one exposure.
run to another, and second, it would provide experimental
data of value to a general correlation of all factors in-
fluencing the response of film to $\beta$ rays. Accordingly,
techniques for measuring both mg emulsion/cm$^2$ and mg AgBr/cm$^2$
were developed, accurate to about 5 or 10%. These are de-
scribed in Appendix 3.

B. Film Processing Techniques

The importance of standardizing film development
and other processing techniques, if different development
runs are to be compared, has already been pointed out. Con-
sequently, standard processing conditions were set up and
tested. An even more effective means of preventing fluc-
tuations in results due to processing variations is to car-
ry through each processing batch a set of standard calibra-
tion exposures. These can later be used to normalize all
development runs, in a particular study, to the same con-
ditions. Both uniform processing and calibration exposures
were used in the experimental investigations to be reported.

The first requirement for testing the uniformity
of a film processing procedure is the achie vement of methods
for obtaining reliably reproducible exposures. The first
development tests which we conducted were on films exposed
in the light sensitometer of the M.I.T. optics laboratory.
The graded exposures were produced by a standard light
source screened from the film by a rotating wheel. This
wheel had segments of variable depth and arc length cut
in its rim so that in 1 revolution a series of exposures
graded by factors of 2 could be obtained. The instrument
was not operating perfectly, however, and it soon became
evident that exposures were not sufficiently reproducible.
Consequently, 2 "radiators" consisting of 6 $\beta$ ray sources
each were prepared using 20 year half life Sr$^{90}$ (0.61 Mev)
with its daughter, Y$^{90}$ (2.3 Mev), in plaster of Paris. The
activity of these 6 sources was graded by factors of about
2.2 (accurately known), and was sufficiently high that conveniently short exposures (order of a few minutes) could be used. In practice, 2 exposures of the radiator were made in the time ratio of 3:2. This procedure gave a conventional "gray scale" of 12 (relatively) known exposures. An example of one of these gray scales is shown in Fig. 19.

![Gray Scale Image](image)

**Fig. 19**

These radiators were later used in the same way to give the standard calibration exposures in each development batch. Their use was extremely convenient and extremely valuable throughout the whole course of this research, for it permitted an accurate comparison of data taken many months apart. These radiators are described in greater detail in Appendix 4.

A second essential in development testing, and of course in all other quantitative photographic work, is a densitometer. The instrument used throughout most of this research was an Ansco Color Densitometer. It is a fairly reliable and stable instrument, and is sufficiently sensitive to be capable of reading diffuse densities between 0 and 6 for aperture sizes less than 1/16" diameter. The manufacturers claim that when calibrated by a standard gray scale it has an accuracy of approximately ± 0.02 units of density over the whole density range. All films were analyzed with filters for black and white, rather than for color.

The processing procedure finally adopted provided
for development of 2 to 4 film strips (1" x 3 1/2") in a 4 oz. bottle of fresh developer at 20 ± 0.5°C, with rather uniform continuous agitation. Development was always followed by stop bath, fixing, and washing, as recommended by the manufacturer, all at 20°C. Details of the processing procedure are described in Appendix 5.

After the final procedure was established, we conducted the following set of tests to determine the reproducibility of the method. Each of four Contrast Process Ortho films was cut into 4 strips, and an exposure of the radiator was made on each strip. Four strips, one from each film, were developed simultaneously and composed one development run. For each of the 4 development runs, an H and D curve was obtained based on the average of the 4 exposed strips. In this way, I hoped to eliminate the variation of emulsion sensitivity from film to film, leaving only the effects of development condition differences. This whole procedure was repeated, giving 2 sets of 4 development runs. The 4 H and D curves of each set were averaged, and the mean deviation from average determined. From the 6 independent pieces of data (2 degrees of freedom lost in the 2 averaging processes), it was concluded that development conditions were constant to within a mean fluctuation of 2.5%, interpreted in terms of exposure. This means that if identical exposures were given to identical strips of Contrast Process Ortho, and if these strips were developed independently, one would interpret the observed density variations as an average of 2.5% variation in exposure. This development variation appeared to be somewhat smaller than that resulting from differences in sensitivity of different pieces of film from the same box.

In exposing the film involved in the actual experiments later reported, precautions were consistently taken to rule out fluctuations caused by humidity and temperature variations at the time of exposure and fading be-
between exposure and development. Almost all films were kept in a CaCl₂ desiccator at room temperature for 12 hours minimum (and usually for several days) before exposure. The shorter conditioning periods may well have been too short for equilibrium humidity to have been reached, although it seems unlikely that the disparity would be large. Exposures were made at room temperature, and in view of the relative independence of sensitivity from temperature at time of exposure, this degree of control would certainly be adequate. The time lapse between exposure and development was in most cases less than 24 hours in order to minimize fading. Wherever fading appeared to be a real danger, simultaneous tests were made with the calibration sources so as to determine the magnitude of the effect. It was never greater than a few percent.

C. Monokinetic Electron Exposures

1. Properties investigated

Somewhat more than half the experimental work of this thesis has been directed toward the study of the response of film to monokinetic electrons. The information so obtained is not as directly applicable to practical dosimetry problems as the continuous spectrum exposures. However, it has been possible in the monokinetic exposures to separate some of the many variables, thereby revealing certain of the basic processes more clearly than could be done with continuous spectrum exposures. Five different types of film, of widely varying properties, have been investigated in the energy range between 0.030 and 1.8 Mev.

The measurements made on these films include, in each case, one or more of the items listed below. (The significance of all these items for β ray dosimetry with film, together with an evaluation of the present state of knowledge regarding them, has been discussed in the earlier
chapters, and is not considered in detail here.) First, the dependence of H and D curve shape on electron energy has been investigated. Second, a study was made of the dependence of sensitivity (expressed both as particle number sensitivity and as dosage sensitivity) on the electron energy for 0° angle of incidence. Third, the variation of sensitivity with the angle of incidence has been examined in sufficient detail to predict the sensitivity relative to perpendicular incidence for an arbitrary angular distribution of incident electrons in a wide energy range. Fourth, the effect of variations in development time upon sensitivity has been studied for exposures produced at several different energies.

2. Apparatus

A 180°, variable field magnetic β ray spectrometer was designed as a monokinetic electron source. The β ray source in the spectrometer consisted of about 17 mc of Sr90 and 17 mc of Y90, which provided an electron beam of adequate intensity at the exit window of the spectrometer between 0.03 and 1.8 Mev. The slit system of the spectrometer was so designed that half the β particles incident on the area of film of interest were within a momentum spread of 2%, or an energy spread of 2 to 4%. This resolution was entirely adequate for the problems under investigation. The mean energy of the electrons incident on the film was determined by measurement of the field strength of the electromagnet and a knowledge of the spectrometer slit system. The mean energy was known to within 2 or 3% for each particular exposure. The number of electrons incident per cm² of film was measured with a proportional counter which could be located over the exit window in place of the film. It seems probable that the measured number of electrons/cm² was accurate within 5 or 10% over the whole energy range of 0.03 to 1.8 Mev. The radiation
dosage to which the film was subjected was determined theoretically from the known energy and number/cm² of the incident electrons, using the curves computed in Chap. 2 and Appendix 1 for the incident and backscattered dosage, respectively. The film, mounted on a bakelite or lucite support, could be exposed to the electrons at any one of 5 angles of incidence: 0°, 17.5°, 35°, 52.5°, and 70°. Exposures could be timed accurate to the second, or to better than 1% in almost all cases. The electromagnet and associated equipment are discussed more fully in Appendix 6. The spectrograph is described in detail in Appendix 7. The proportional counter and auxiliary apparatus, together with the determination of the β ray spectrum at the exit window of the spectrograph, are considered in Appendix 8. This apparatus may be seen in its operating arrangement in Fig. 20. The gas and vacuum system is located at the right. The electromagnet, with the spectrograph between its pole faces and the counter in position at the spectrograph exit window, is on the left.

The magnet, and the circuitry and gas system for the counter, were shared with Dr. Gordon L. Brownell. A part of the development of these elements of the apparatus was done by Dr. Brownell, particularly in the equipment associated with the counter.

3. Experimental results

The bulk of the information obtained on the response of films to monokinetic electrons is contained in Figs. 21 to 42. These figures are to be considered as the primary experimental results. The remainder of this section is given to an amplification of these results, having as its purpose the clarification of the nature of some of the measurements and their analysis, the synthesis of general conclusions from the data on the 5 films, and the emphasis of certain features of the results on individual films.
a. Films and processing conditions

The 5 films investigated are listed below, together with certain of their significant features and the processing conditions which, unless otherwise specifically noted later, were used in each measurement.

Eastman No Screen X-ray Film

Physical properties:
Emulsion thickness: 5.8 mg/cm\(^2\) on each side of film base. Approximately 0.1 mg/cm\(^2\) gelatin T coat on emulsion surface
Silver halide content: 3.5 mg/cm\(^2\), each side of film base
Film base: cellulose acetate, 26 mg/cm\(^2\) thick

Data shown are for both double and single emulsion film (emulsion removed from back surface of film base to give the single emulsion film)

Processing:
Development: 7.0 minutes at 20° C in Kodak X-ray Developer (powder)
Stop Bath: 1/2 minute in 2.5% acetic acid
Fixation: 15 minutes in Kodak X-ray Fixer
Wash: approximately 1/2 hour

Eastman Industrial X-ray Film, Type A:

Physical properties:
Emulsion thickness: 3.8 mg/cm\(^2\), each side of film base. Approximately 0.1 mg/cm\(^2\) T coat on emulsion surface
Silver halide content: 1.8 mg/cm\(^2\), each side of film base
Film base: cellulose acetate, 21 mg/cm\(^2\) thick

Data shown are for both double and single emulsion film (emulsion removed from back surface of film base to give the single emulsion film)
Processing:
Development: 7.0 minutes at 20°C in Kodak X-ray Developer (powder)
Stop bath: 1/2 minute in 2.5% acetic acid
Fixation: 10 minutes in Kodak X-ray Fixer
Wash: approximately 1/2 hour

Cine Positive # 5302
Physical properties:
Emulsion thickness: 2.1 mg/cm². Approximately 0.1 mg/cm² T coat on emulsion surface
Silver halide content: 0.72 mg/cm²

Processing:
Development: 15.0 minutes at 20°C in 1/4 concentration D-19
Stop bath: 1/2 minute in 2.5% acetic acid
Fixation: 6 minutes in Kodak Acid Fixer
Wash: approximately 1/2 hour

NTB Stripping Film
Physical properties:
Emulsion thickness: 4.5 mg/cm² (10μ). Approximately 0.1 mg/cm² T coat on emulsion surface
Silver halide content: 3.7 mg/cm²

Processing:
Development: 30 minutes at 20°C in full concentration D-19
Stop bath: 1/2 minute in 2.5% acetic acid
Fixation: 7 minutes in Kodak Acid Fixer
Wash: approximately 1/2 hour

SWR
Physical properties:
Emulsion thickness: 0.17 mg/cm². No T coat
Silver halide content: 0.16 mg/cm²

Processing:
Development: 4 minutes at 20°C in 1/2 concentration D-19
Stop bath: 1/2 minute in 2.5% acetic acid
Fixation: 3 minutes in Kodak Acid Fixer
Wash: approximately 15 minutes

All films manufactured by the Eastman Kodak Co., Rochester, N.Y.

a. Analysis of measurements

(1). Criteria of acceptability of data

Most perpendicular incidence spectrograph exposures were made in duplicate or triplicate for the first films investigated, using a circular exit window of diameter described in Appendix 7. The only exceptions were the single exposures used in determining the monokinetic electron H and D curve shapes. Whenever inconsistencies existed in the data obtained from these supposedly identical exposures, the measurements were repeated. The later films were investigated using single exposures through the rectangular exit windows of length about 1/2", as described in Appendix 7. Since the number of electrons/cm²/sec was tested, by repeated photographic exposures, and found to be uniform over the length of the rectangle, several density measurements could be made on these films at different points which were identically exposed. Therefore the later films, also, in effect received multiple exposures. All exposures for angles of incidence other than 0° were done in duplicate.

Besides the check on faulty measurements as made by multiple exposures, a second check was available through the Sr⁹⁰ - Y⁹⁰ calibration exposures which were placed on the side of each film strip, where possible, or at least on 1 strip in each development batch. The primary purpose of these calibration exposures, as already mentioned, was to permit intercomparison of results obtained in different development runs. Beyond this, however, any excessively
erratic behavior of the calibration exposures on a single strip would serve to indicate the presence of errors on that strip (e.g., nonuniformity of emulsion thickness). Such erratic behavior was very seldom observed.

(2). Density-exposure relationship

I have chosen to show the relationship between density and exposure in an unconventional plot of logD vs. logE. This function was chosen simply because it permits the presentation of the experimental data with equal accuracy over the whole density range investigated. In the conventional H and D curve, if the scales are adjusted so as to give a reasonable slope (i.e., not near infinity) at the higher densities, a small fractional change in D at low D would be represented by such a small displacement on the graph as to be undetectable. In the case of a D vs. E curve, essentially the same difficulty would exist. However, the log D vs. log E curve gives no particular advantage in practical applications where two different exposures are to be compared via the graph, and it has the disadvantage of not making the contrast as readily apparent as the H and D curve.

We have seen that the nature of the initial increase of D with E gives at least a small amount of information regarding the number of hits per grain required for exposure. This problem has been discussed with reference to a more conventional D vs. E plot. For the log D vs. log E graphs shown, the situation is this: If $D = kE$, the curve would be a straight line of slope = 1 (i.e., 45° with the axes of the graph). For each film this relationship is sketched in as a dashed line normalized to the experimental curve at $D = 0.3$. If $D = kE^x$, the curve would be a straight line of slope = $x$. The intercept on the axes is of no significance.
In the log D vs. log E curves shown, I have been concerned only with the relative shapes of the curves. Therefore I have expressed exposure in relative, rather than absolute, units, and have adjusted the units for each set of points at a particular energy so that in any one graph each set falls as nearly as possible onto the same curve.

(3). Angle of incidence measurements

Certain features of the method of measurement and analysis of the angle of incidence data should be briefly discussed. In connection with the method of measurement, the arrangement of the film during exposure is described in Appendix 7. One problem which arose as a result of this arrangement was the following: In a 180° spectrograph, the β ray beam is brought to a focus at the exit window plane and diverges beyond that plane. The counter window and the film in perpendicular incidence measurements were positioned in that plane by the design of the apparatus. However, only the lower edge of the film strips mounted at an angle were in that plane, the rest of these strips being above the plane. Since the strips had to be densitometered at least a small distance in from their edge, the area densitometered was located above the exit window plane during exposure. As a result of the divergence of the β ray beam above the plane of focus, the number of β's/cm² incident on the film was lower than it would have been had the film been in that plane of focus. The situation was aggravated by the fact that at high energies, and especially at the larger angles of incidence, many of the β particles incident on the central (0°) film strip were scattered in the film holder and passed out of the holder through one of its side faces, i.e., through the film strips mounted at an angle. This process gave an extra spurious exposure near the lower edge of the angle film strips, and made it necessary to densitometer these films
several mm from their lower edges. To estimate the decrease, resulting from β ray beam divergence, in the exposure to which the film strips were actually subjected about 3 mm above the exit window plane, a special film holder was constructed which permitted 0° incidence exposures in that position. It was found by these quantitative photographic measurements that for film in this position, the exposure was approximately 20% less than in the exit window plane. This correction was applied where appropriate to the angle of incidence data. The film strips exposed to lower energy electrons were densitometered nearer to, but not at, their edges. A beam divergence correction of a few percent was estimated from the measured correction at 3 mm above the plane. Because of the need for these corrections, and of difficulties in positioning the film accurately, the angle of incidence data do not have as great an accuracy as the other spectrograph measurements.

The notation $S'_\theta / S_0$ on the ordinate of Figs. 25, 33, and 37 signifies the ratio of the particle number sensitivity of the film at the angle in question to the particle number sensitivity at 0°. (The angle in question is hereafter referred to as $\theta$.) The particle number sensitivity, $S'_\theta$, for the measurements at angles other than 0°, is based on a somewhat special definition of particle number exposure, $E'$. $E'$ is defined as the number of electrons incident on the film, per cm$^2$ in a plane normal to the beam, not per cm$^2$ film. Had this exposure been given in terms of electrons per cm$^2$ film ($= \cos \theta \times$ number of electrons per cm$^2$ normal to the beam), the ratio $S'_\theta / S_0$ would be greater than with the definition actually used by the factor $1/\cos \theta = \sec \theta$.

My reason for presenting the data as I have is the following: Suppose the electrons passed completely through the emulsion without scattering or absorption. If a piece of film were given 2 exposures of equal duration in
a given constant electron beam, first at $0^\circ$ incidence and then at $\theta^\circ$ incidence, the density produced in the 2 exposures would be the same. In the $\theta$ exposure the number of electrons per cm$^2$ film would be reduced, relative to the $0^\circ$ exposure, by the factor $\cos \theta$. However, the path length of each electron in the emulsion would be increased by $\sec \theta$, so that the total path length in the emulsion of all the electrons incident on 1 cm$^2$ of film would be the same. Therefore the density would be independent of the angle of incidence. If exposure is defined as it has been in Figs. 25, 35, and 27, i.e., in terms of number of electrons/cm$^2$ normal to the beam, the sensitivity of the above hypothetical film would be independent of the angle of incidence, and a plot of $S'_\theta/S_0$ would be a horizontal line having an ordinate value of unity for all angles. The deviation of the experimental curves for $S'_\theta/S_0$ from horizontal straight lines at unity is therefore an indication of the effects of absorption and scattering. In all other figures but these three, particle number exposure refers to the number of electrons/cm$^2$ film.

In Figs. 23, 31, and 35, the sensitivity of Eastman No Screen, Eastman Type A, and Cine Positive #5302 under diffuse incidence of monokinetic electrons is shown. These curves were calculated from the data of Figs. 25, 33, and 37, making use of the fact that in diffuse incidence the fraction of the electrons incident per cm$^2$ film at angle of incidence between $\theta$ and $\theta + d\theta = \frac{dP(\theta)}{d\theta}$ $d\theta = 2 \sin \theta \cos \theta \, d\theta$ (see Table 2). The calculation was carried out as follows: At a set of particular energies, $S'_\theta/S_0$ vs. angle of incidence was plotted, with experimental points at $\theta = 0^\circ$, $17.5^\circ$, $35^\circ$, $52.5^\circ$, and $70^\circ$. A smooth curve was drawn through these points. This curve was then multiplied by $2 \sin \theta \, S_0 \, d\theta$ and graphically integrated between $\theta = 0$ and $\theta = \pi/2$. It can be seen that the following relationships hold:
\[ \int_{0}^{\pi/2} \frac{S_0'}{S_0} 2 \sin \theta \, S_0 \, d\theta = \int_{0}^{\pi/2} \frac{S_0 \cos \theta}{S_0} 2 \sin \theta \, S_0 \, d\theta = \int_{0}^{\pi/2} S_0 2 \sin \theta \cos \theta \, d\theta \]

The last expression is an integration of the sensitivity as a function of angle weighted appropriately for diffuse incidence. Hence the calculation gives the diffuse sensitivity $S_{\text{Diff}}$. An analogous method could of course be used for any other selected angular distribution.

(4). Calculation of absolute sensitivity

For the calculation of absolute sensitivity at various energies, the raw data consisted of a measured density for a known number of electrons/cm$^2$. From this, the number of electrons/cm$^2$ required for $D = 0.3$ was determined as follows: The relationship between $D$ and $E$ was plotted (usually as log $D$ vs. log $E$) for the Sr$^{90}$-Y$^{90}$ calibration exposure associated with the spectrograph film strip in question. Using this curve, it was possible to find immediately the absolute exposure required for $D = 0.3$, knowing the absolute exposure for the measured $D$. Since several film strips were required in a particular sensitivity curve determination, several calibration curves were involved. The absolute exposures were therefore finally normalized (a matter of not more than a few percent) with respect to the mean calibration curve, so that the whole set of data would be self-consistent.

The dosage sensitivity curves were calculated from the particle number sensitivity curves by converting number of electrons/cm$^2$ to dosage via the data of Fig. 4 and Table 7. One is in general interested not in the dosage which exists in the film itself, but rather in the dosage which would exist in a piece of tissue were it in the
position of the film. Therefore the dosage sensitivity curves shown are based on this dosage, where for concreteness tissue is considered to be identical, for present purposes, with lucite. In Appendix 1 it was shown that to a reasonably good approximation, electrons backscattered from lucite, for any energy or angle of incidence of the incident electrons, contribute 25% to 30% of the surface dosage given by the incident electrons. Therefore in all the curves, 25% was added to the straight theoretical calculation of dosage to allow for this backscatter contribution.

(5). Relative energy dissipation per grain as a function of electron energy

We have seen in Chap. 4 that it would be useful to know the energy dissipation required for the exposure of an individual grain. In the absence of these data, related information on the relative amount of energy dissipation per grain exposed, as a function of electron energy, would be valuable. As previously pointed out, this information would not of itself permit a complete prediction of the dependence of the sensitivity of the film on electron energy, because of the complicating effects of absorption and scattering. However, it helps give an understanding of one of the variables in the process.

From the perpendicular incidence particle number sensitivity curves one can extract fairly reliable data on the relative energy dissipation per exposed grain at high and low electron energies. At high energies, when the electrons pass perpendicularly through the emulsion without scattering, the energy dissipation per electron in the emulsion can be computed by combining a knowledge of the mg/cm$^2$ AgBr and gelatin in the emulsion with the theoretical rate of energy loss of the electrons, together with a 25% extra dosage added for the contribution of the electrons backscattered from the emulsion backing. At low energies (less
than enough to permit complete penetration of the emulsion) the energy dissipation in the emulsion is given simply by the total energy of the electron minus a small correction for the electrons backscattered out of the emulsion. This correction may be estimated with sufficient accuracy for present purposes from the following rough analysis. Using Eklund's backscatter data of Table 3, one sees that about 30% of the low energy electrons incident on solid AgBr would be backscattered, and about 6% of those incident on gelatin. Since emulsion is about half AgBr, half gelatin, we will just average the above two values and say that 18% of the electrons are backscattered from emulsion. However, even these lose a fraction of their energy to the emulsion, as can be seen from Brownell's curves on the $P_{32}$ spectral shape with and without backscattering present.

We may estimate that very roughly half the energy of the electrons which are backscattered is given to the emulsion, and therefore take a value of 10% as the correction for the backscattered energy. Using these methods of analysis, one may then estimate the ratio, at a high and a low electron energy, of the energy dissipation in the emulsion for a given D. If the size of a developed grain is independent of the energy of the exposing electron, this calculation also gives the ratio of energy dissipation required for the exposure of an "average" grain. Of course, the energy associated with the high energy electron during its exposing action has an essentially constant value, whereas the energy associated with the soft electron during its exposing action is anywhere between its initial energy and zero. The results of these computations are given later under the discussion of the specific films.

c. General conclusions applicable to all films

From the figures giving the monokinetic spectrograph data, certain conclusions can be drawn which are pertinent, within fairly wide limits, to all the films investi-
gated. These conclusions are considered first.

(1). Independence of H and D curve shape from energy

For all the films except SWR, the density-exposure relationship is plotted in the form log D vs. log E, for exposures at several different energies between about 1 Mev and 0.1 Mev or less. (Figs. 21, 22, 29, 30, 34, and 38). It can be seen that over the entire range of energies the log D vs. log E curves have almost identical shapes. Below D = 1 the deviation of the various curves from each other (for a particular emulsion) are undetectable within the experimental error of a few percent. The only serious exception is the case of the double emulsion films at densities of about 2 or greater and at the lowest energies, an effect undoubtedly resulting from absorption of the electrons in the emulsion as discussed in Chap. 6. The significance and convenience, for quantitative β ray measurements, of this independence of H and D curve shape from energy was discussed in Chap. 6.

The extent of this independence is somewhat surprising in all the films except No Screen. The curves for these films show that many of the grains are definitely not exposed in single hit events. One would therefore expect that at the lower energies where specific ionization is higher, the curves would begin to approach a single hit shape. With reference to Cine Positive, for example, at 0.071 Mev the specific ionization in AgBr is about 2.5 times as great as at 1 Mev, so that one might well expect to find more single hit events at the low energy, relative to the high energy, than apparently exist.

(2). Effect of angle of incidence on sensitivity

The effect of the angle of incidence on sensitivity is in large part a property only of the emulsion thickness, rather than of such variables as grain sensitivity.
To this extent it can be estimated for any emulsion if only the simple property of emulsion thickness is known. Although as has already been stated, these data are not as reliable as the perpendicular incidence data, the following generalizations can be made from the curves of Figs. 25, 33, and 37. (One must bear in mind the definition of $S'_\theta$, and the fact that $S'_\theta/S_\theta$ would equal 1 at all energies and all angles of incidence if no absorption or scattering of the electrons by the emulsion occurred.)

At low energies and high angles of incidence, $S'_\theta/S_\theta$ is less than 1. This is to be understood in terms of absorption in the emulsion. When the energy is so low that in actuality even the $0^0$ electron cannot completely penetrate the emulsion (e.g., less than 0.06 Mev in the case of 5.8 mg/cm$^2$ No Screen), then the following situation obtains: If no absorption or scattering did exist, the path length of the $\theta$ electron would be greater than that of the $0^0$ electron by the factor $\sec \theta$, and as we have seen, $S'_\theta/S_\theta$ would equal 1. However, as a result of the absorption which does take place, the path lengths are actually equal (neglecting for the moment the backscattering out of the emulsion) instead of in the ratio $\sec \theta$: 1. Hence the sensitivity is lower for the $\theta$ electron than it otherwise would be by the factor $1/\sec \theta = \cos \theta$. Because backscattering out of the emulsion is somewhat more likely for the $\theta$ electron than for the $0^0$ electron, $S'_\theta/S_\theta$ will actually be somewhat less than $\cos \theta$. This relationship is found experimentally, as can be seen in the figures. (cos $70^\circ = 0.34$, cos $52.5^\circ = 0.61$, cos $35^\circ = 0.82$, cos $17.5^\circ = 0.95$).

As the energy increases, part of the path length of the $0^0$ electron is "wasted" in the emulsion backing, whereas none of the $\theta$ electron path is wasted in the backing. (At least, although some electrons are scattered perpendicularly into the backing, they are fewer in number.)
Therefore $S'_\theta / S_0$ increases with energy. At some intermediate value of energy, the $0^\circ$ electron will pass completely through the emulsion with little scattering, having a path length equal to the emulsion thickness $t$, whereas the $\theta$ electron will have a path length in the emulsion greater than $ts\theta$ because of scattering at the end of its longer oblique path. Therefore in this energy range $S'_\theta / S_0$ is greater than 1.

At energies sufficiently high that little scattering or absorption exists either for the $\theta$ or the $0^\circ$ electron, one expects, as previously seen, that $S'_\theta / S_0 = 1$. One might suggest that the effect of backscattering from the emulsion backing would affect the ratio in this case, since more of the $\theta$ electrons are scattered back into the emulsion than are the $0^\circ$ electrons. However, this case now resembles the situation discussed in Appendix 1, where it was shown that the fraction of the dosage in a thin surface layer, contributed by backscattered electrons, was essentially independent of the angle of incidence. Therefore we are still justified in expecting $S'_\theta / S_0$ to be approximately equal to 1 at high energies. The experimental curves indicate that $S'_\theta / S_0$ is somewhat greater than 1 at high energies, although it is less than the peak at an intermediate energy. However, the accuracy of the high energy high angle of incidence curves is low.

The calculated curves for diffuse incidence (shown in the case of dosage sensitivity in Fig. 25), indicated the same trends discussed for the case of a discrete angle of incidence $\theta$. The curves could not be carried to higher energies because at these energies an excessively large fraction of the sensitivity is contributed by electrons of $\theta$ greater than $70^\circ$, for which no experimental data existed. (The curves may have been carried farther than justified as it is.)
(3). Variation of sensitivity with energy

Certain aspects of the variation of sensitivity with energy are strongly influenced by the emulsion thickness, and therefore can be understood for any emulsion if only the simple property of emulsion thickness is known. We have already examined this at some length in Chap. 6 in evaluating experiments already reported in the literature. Furthermore, out of the 5 types of film examined in this research, the 3 which I investigated in most detail behave so similarly in the variation of their sensitivity with energy that information obtained from them has a certain degree of generality.

The effect of emulsion thickness on particle number sensitivity as a function of energy in the case of perpendicular incidence has been sufficiently covered in Chap. 6. It was shown there why one would expect an initial rise of sensitivity with energy, a peak at about 0.1 Mev, and a decline in sensitivity at higher energies. The double hump is to be expected in the double emulsion film by the same reasoning. The chief new contribution of my data (aside from establishing the magnitude of these variations for the particular films), is to show that particle number sensitivity is essentially constant beyond 0.5 or 0.7 Mev. Little good data were previously available at these energies and higher, and the most extensive, those of Ellis and Aston, showed a continuing decrease of sensitivity with increase in energy. At high energies, the double emulsion films are expected to be just twice as sensitive as the single emulsion films, and this is observed.

The diffuse incidence data help to clarify question in particular, namely, to what extent is the hump in the perpendicular incidence particle number sensitivity curve caused by scattering of the electrons, so that they may have a path length in the emulsion greater than the
emulsion thickness, and to what extent is it caused by the higher specific ionization that exists at 0.1 Mev relative to higher energies. Diffuse incidence exists when the electrons are incident with a \( \cos \theta \) distribution. We have seen in Chap. 2 that a \( \cos \theta \) distribution is the stable angular distribution for electrons as they are absorbed in matter. Therefore, to a first approximation scattering does not greatly increase the path length of 0.1 Mev electrons in the emulsion relative to 1 Mev electrons, since the angular distribution of both is initially, and remains, roughly the same. (This argument is not conclusive, however, since angular distribution is not the sole criterion determining path length in the finite emulsion thickness. For example, absorption of the soft electrons tends to decrease their path length in the emulsion, while on the other hand, higher scattering of the soft electrons tends to increase their path length in the emulsion even if their angular distribution remains \( \cos \theta \).) If the effect of scattering on the relative path lengths, in the emulsion, of 0.1 and 1 Mev electrons, respectively, is actually small when the electrons are diffusely incident, then one would expect the observed hump in the diffuse sensitivity curves to be caused by specific ionization effects. Further evidence that specific ionization plays some part in these sensitivity humps is deduced more fully in the later discussion of the individual emulsions. There it is shown that, although the energy dissipation per grain exposed is higher at low energies than at high energies, this ratio is not as great as the ratio of specific ionization at these 2 energies. Hence part of the increased specific ionization of lower energy electrons is actually photographically effective in all of these films.

The diffuse sensitivity curves were calculated by the method described, for the energy region between 0.06 and 0.3 to 0.8 Mev (depending on the film). They were extrapolated to 0 at the low energy end, and extended
in the high energy region under the assumption that diffuse sensitivity, like perpendicular sensitivity, is flat at high energies. As previously discussed, these curves quite likely indicate sensitivities that are too high at high energies, particularly in the case of Cine Positive.

The dosage sensitivity curves show humps at low energies which are much less pronounced than the particle number sensitivity curves. This is of course expected since they were calculated from the particle number sensitivity curves by dividing by the specific ionization, which is larger at low than at high energies. The extent to which these dosage sensitivity curves conform to the ideal of independence from energy can best be seen from the graphs. The significance for β ray dosimetry of the degree of this conformity has already been emphasized in earlier chapters.

d. Conclusions applicable to specific films

In this section, certain special features of the spectrograph data, pertinent to particular films only, are discussed. Again, only the highlights are mentioned; the primary source of information remains the curves themselves.

(i). Eastman No Screen

(a). Low exposure D vs. E curve

The most significant feature of the density-exposure relationship is that the initial D vs. E curve, which can be visualized on the basis of Figs. 21 and 22, is very accurately a straight line. This has been found to be the case in all other tests with No Screen, as well as with the single set of data shown. It must indicate that the exposure of most grains takes place in single hit events.
(b). Relative energy dissipation per grain exposed, at high and low energies

When the calculation is performed for Eastman No Screen, single emulsion, in the manner described under "Analysis of measurements", one finds the ratio of energy dissipation in the emulsion at 1 mev to that at 0.04 Mev, for a given density, to be 0.56. One can see therefore, that without the complicating effects of scattering and absorption, the dosage sensitivity at 0.04 Mev would be about 1/2 that of high energy electrons. (For dosage measurements, to emphasize the point again, one wants the dosage sensitivity to be the same at all energies.) However, since the specific ionization of a 0.04 Mev electron is about 4 times that of a 1 Mev electron, the density produced per incident particle, rather than per unit dosage, would be about \((0.56)/0.25\) = twice as great at 0.04 Mev as at 1 Mev. Hence one can see that the hump on the particle number sensitivity curve is caused in part by specific ionization considerations rather than by the effects of scattering alone. In other words, per incident particle, high specific ionization electrons are more effective in exposing No Screen than low specific ionization electrons.

We are now presented with something of a dilemma. How can this last statement be reconciled with a previous conclusion that a large fraction of the grains are exposed in single hit events, even at high energies? There would seem to be 2 possible explanations of this dilemma. The first is that the straight line D vs. E curve may result merely from a skew grain sensitivity distribution, rather than from single hit exposure events. It seems improbable, however, that this is the whole explanation, since the relationship is so accurately a straight line. The second possible explanation is that the individual grains do not behave entirely as single units, but instead a larger developed grain is associated with a larger latent image.
A situation similar to this does exist in physical development, and since some physical development does take place under practical development conditions, this explanation may be part of the solution of this dilemma.

(c). Variation of sensitivity with development

Figs. 26 and 27 give information on the variation of perpendicular sensitivity (either particle number or dosage) with development time, for various energies of incident electrons. Fig. 26 shows the sensitivity at development time $T$, relative to a development time of 7.0 minutes, vs. electron energy. It is apparent that for all energies sensitivity increases with development time. The other significant feature is that a latent image formed by low energy electrons is developed more rapidly than one produced by high energy electrons. This may be a result of the fact that the larger latent image associated with the high specific ionization of the low energy electrons catalyzes development more rapidly than the smaller latent image associated with the high energy electrons. Or, it may be merely a result of the fact that the low energy exposure lies nearer the surface of the emulsion than the high energy exposure, and is reached more quickly by the developer. Fig. 27 shows this second feature somewhat more clearly. In this case the development constant $k$ is shown as a function of electron energy, assuming the equation $D = D_0 (1 - e^{-kt})$ for the relationship between density and development time $t$. The value of $k$ was determined by treating $k$ as an adjustable constant in fitting the equation to the data. Actually, the equation in this case was not a very accurate representation of the relationship, but the values of $k$ taken from the data on the basis of this equation still have some significance.
(2). Eastman Type A

(a). Low exposure D vs. E curve

The single emulsion Type A D vs. E curve is slightly sigmoid at low exposures. The slope of the log D vs. log E curve is 1.2, indicating that at low exposures $D = kE^{1.2}$. This relationship, which would exist for a hypothetical uniform grain size, uniform grain sensitivity emulsion requiring 1.2 hits per grain, shows that at least many of the Type A grains are not exposed by a single electron.

(b). Relative energy dissipation per grain exposed, at high and low energies

By the same method of computation as was used for No Screen, it was found that the ratio of the energy dissipation for a given density, at 1 Mev relative to 0.04 Mev, was 0.43.

(3). Cine Positive #5302

(a). Low exposure D vs. E curve

The data of Fig. 34 show that at low exposures $D = kE^{1.2}$.

(b). Relative energy dissipation per grain exposed, at high and low energies

At 1 Mev and 0.03 Mev the relative energy dissipation in the emulsion for a given D is 0.58.
(4). NTB Stripping Film

(a). Diffuse incidence sensitivity

It was not possible to make a study on NTB of the variation of sensitivity with angle of incidence, for the reason that the emulsion peeled excessively under the conditions of exposure at angles of incidence other than zero. However, as already pointed out, this property is determined almost exclusively by emulsion thickness rather than by any of the more esoteric emulsion properties. Since NTB thickness is between that of Type A and No Screen, diffuse sensitivities may be estimated by the data on these two. When one uses the average of these two films, one finds that a pronounced peak still exists in the diffuse incidence particle number sensitivity curve, but that the dosage sensitivity curve is more nearly flat than any of the other films. On these grounds NTB would seem to be the best of the 5 films tested for purposes of diffuse incidence dosimetry measurements.

(b). Low exposure D vs. E curve

The data of Fig. 38 show that at low exposures $D = kE^{1.1}$.

(c). Relative energy dissipation per grain exposed, at high and low energies

An energy backscatter of 15%, instead of the 10% associated with the other films, was assumed for NTB exposures at low energies because of its high AgBr content. On this basis it is found that at 1 Mev and 0.04 Mev the relative energy dissipation in the emulsion for a given $D$ is 0.95. This result, also, suggests the excellence of NTB for dosimetry purposes.
Figure 21
EASTMAN NO SCREEN SINGLE EMULSION
NET DENSITY vs RELATIVE EXPOSURE

Net Density

Relative Exposure

- $E = 1.19$ Mev
- $E = 0.228$ "
- $E = 0.088$ "
- $E = 0.043$ "
- $E = 0.033$ "
- Calibration Exposure ($\text{Sr}^{90}_{-90}$)

$D = kE$
Figure 2.2
EASTMAN NO SCREEN DOUBLE EMULSION
NET DENSITY vs RELATIVE EXPOSURE

- $E = 1.19$ Mev
- $E = .228$
- $E = .088$
- $E = .043$
- Calibration Exposure ($\text{Sr}^{90} - \text{Y}^{90}$)
- $D = kE$
Figure 23
EASTMAN NO SCREEN
PARTICLE NUMBER SENSITIVITY vs ELECTRON ENERGY

- Single Emulsion, Perpendicular Incidence
- " " " " " "
- Double " " " " "
- Single Emulsion, Diffuse Incidence

Particle Number Sensitivity (Electrons/cm²) x 10⁷ for D = 0.3

Electron Energy (Mev)
Figure 24  Eastman No Screen
Dosage Sensitivity vs
Monokinetic Electron Energy

Dosage Sensitivity: \((100 \times \text{ergs}) / \text{gm}\) for \(D = 0.3\)

- **Double Emulsion, Perpendicular Incidence**
- **Single Emulsion, Perpendicular Incidence**
- **Single Emulsion, Perpendicular Incidence**
- **Single Emulsion, Diffuse Incidence**

Monokinetic Electron Energy (Mev)
Figure 25

EASTMAN NO SCREEN

$S_L/S_{0^\circ}$ vs ELECTRON ENERGY

Electron Energy (Mev)
**Figure 26** EASTMAN NO SCREEN (Single Emulsion)
EFFECT OF DEVELOPMENT TIME ON SENSITIVITY
$S_t/S_0$ vs ELECTRON ENERGY

**Figure 27** EASTMAN NO SCREEN (Single Emulsion)
DEVELOPMENT CONSTANT $k$
vs ELECTRON ENERGY
Figure 28
DOSAGE SENSITIVITY AT DIFFUSE INCIDENCE RELATIVE TO PERPENDICULAR INCIDENCE

$S_{\text{Dif}}/S_0^\circ$ vs ELECTRON ENERGY

- o Eastman No Screen, Single Emulsion
- • Eastman Type A, Single Emulsion
- □ Cine Positive *5302
Figure 29
EASTMAN TYPE A (Single Emulsion)

NET DENSITY vs RELATIVE EXPOSURE

- 1.19 Mev
- 1.172 "
- 1.071 "
- Calibration Exposure (Sr^{90}, Y^{90})
- \(D = kE\), norm. at \(D = 0.3\)
Figure 30
EASTMAN TYPE A, DOUBLE EMULSION
NET DENSITY vs RELATIVE EXPOSURE

Net Density

Relative Exposure
Figure 31
EASTMAN TYPE A
PARTICLE NUMBER SENSITIVITY vs ELECTRON ENERGY

Particle Number Sensitivity (Electrons/cm²) for D=0.3 x 10⁻⁶

- Single Emulsion, Perpendicular Incidence
- Single Emulsion, Diffuse Incidence
- Double Emulsion, Perpendicular Incidence

Electron Energy (Mev)
Figure 32
Eastman Type A
Dosage Sensitivity vs
Monokinetic Electron Energy

Dosage Sensitivity (100 ergs/gm) for D = 0.3

Monokinetic Electron Energy (Mev)
Figure 33
EASTMAN TYPE A, SINGLE EMULSION
$S_L/S_0^\circ$ ELECTRON ENERGY

Electron Energy (Mev)
Figure 35
CINE POSITIVE "5302
PARTICLE NUMBER SENSITIVITY vs ELECTRON ENERGY

Particle Number Sensitivity (Electrons/cm²) x 10⁹ for D = 0.3

Diffuse Incidence

Perpendicular Incidence

Electron Energy (Mev)
Figure 36
Cine Positive 5302
Dosage Sensitivity vs Monokinetic Electron Energy

Dosage Sensitivity: \( \frac{100 \text{ ergs}}{\text{cm}^2} \) for \( D = 0.5 \left( \frac{\text{C}}{\text{cm}} \right)^2 \)

- ○ Perpendicular Incidence
- ● Perpendicular Incidence
- — Diffuse Incidence

Monokinetic Electron Energy (MeV)
Figure 37
CINE POSITIVE *5302
$S_\theta/S_\theta^0$ vs. ELECTRON ENERGY

Electron Energy (Mev)
Figure 38

NTB STRIPPING FILM

NET DENSITY vs RELATIVE EXPOSURE

- □ 0.11 Mev
- ○ 1.19
- ● Calibration Exposure (Sr$^{90}$-Y$^{90}$)
- --- $D = kE$

Net Density

Relative Exposure
Figure 39
NTB STRIPPING FILM

PARTICLE NUMBER SENSITIVITY
vs ELECTRON ENERGY

Particle Number Sensitivity (electrons/cm²) x 10^5 vs D = 0.3

Electron Energy (Mev.)
Figure 40

NTB Stripping Film

Dosage Sensitivity vs Monokinetic Electron Energy (perpendicular Incidence)
Figure 41

SWR

PARTICLE NUMBER SENSITIVITY vs ELECTRON ENERGY
(Perpendicular Incidence)
Figure 42

SWR

Dosage Sensitivity
vs
Monokinetic Electron Energy
(Perpendicular Incidence)
(d). Reproducibility

For reasons never clearly understood, I found the reproducibility of NTB measurements to be somewhat poorer than other films (except SWR). The emulsion thickness is not expected to be less uniform than other films (S-10), and humidity and other variables were controlled as accurately as for the others. It is conceivable, but not likely, that an insufficient number of measurements were made on NTB to permit a reliable estimate of reproducibility.

(5). SWR

SWR is a very fine grain, low gelatin content (less than 5% by weight) thin emulsion produced for ultraviolet exposures. It was included in this investigation primarily because of its thinness. Had it turned out that absorption of β rays in the emulsion of films was the main limitation on dosage measurement accuracy, SWR was expected to be a useful film. Few measurements were made on SWR because abrasion exposures were a serious effect and prohibited high accuracy. Reproducibility in general was poor.

The most significant feature of Fig. 41 is that the peak particle number sensitivity was not reached at the lowest energy exposure. This is to be expected because of the emulsion thinness.

The density-exposure relationship, which has not been presented in a graph, showed that D was approximately proportional to E at low densities.

D. Continuous Spectrum β-ray Exposures

As previously pointed out, practical problems involving radiation dosage by electrons are most often concerned with continuous spectrum β rays. Although the re-
sponse of film to a particular β ray spectrum can be computed with moderate accuracy from the data on monokinetic electrons, two particular advantages are to be obtained from a direct investigation of this response using β ray sources with calibrated surface dosage rates. First, the information so obtained is subject to less error than the indirect information. Second, it is possible to investigate, with such sources, the effect of absorption of the β rays on their exposing efficiency. This problem has already been considered in Chaps. 2 and 3, where it was shown that if the exposing efficiency of a β ray spectrum were not changed by absorption (i.e., if the dosage sensitivity of the film for β rays incident on its surface were independent of the amount of absorber through which these β rays had penetrated before reaching its surface), then reliable dosage calibration of most β ray exposures would be possible. However, it was also suggested that exposing efficiency might not be independent of absorption, as a result of changes introduced by the absorption into the angular distribution and spectral shape of the β rays. For these reasons, I have made an investigation of the response of the 5 films previously considered to calibrated dosage sources of 4 isotopes under various conditions of absorption. These measurements are reported in this section.

1. Equipment

The equipment used in this group of experiments consisted of a thin source of each of the isotopes S\textsuperscript{35} (0.167 Mev), W\textsuperscript{185} (0.43 Mev), Tl\textsuperscript{204} (0.78 Mev), and Y\textsuperscript{91} (1.54 Mev), together with a set of absorbers of various appropriate thicknesses for each source, and an extrapolation ionization chamber and associated apparatus for calibrating the dosage rates produced by the sources at the surfaces of the absorbers.

The sources and their method of preparation are
described in detail in Appendix 9. In brief, the sources consisted of rather uniform deposits, very roughly 1 mg/cm$^2$ thick, of these 4 isotopes on the surface of lucite discs 2 7/8" in diameter and 1/4" thick. For each source there were 3 or 4 cellulose acetate absorbers (cellulose nitrate in the case of the thinnest absorbers for all but the Y$^{91}$ source), whose thicknesses ranged in each set from about 0.1 to 1 half thickness of the associated β rays. The thickness of these absorbers is shown in Table 11, in terms of mg/cm$^2$ and in terms of the number of half thicknesses for the β ray spectrum in question. (One half the thickness of the source itself has been added to the absorber thickness in each case.) The half thickness calculations are based on a value of the absorption coefficient given by the expression $\mu/\rho = 17E^{-1.43}$ cm$^2$/gm, as taken from Chap. 2.

The dosage rates produced by the 4 sources at the surfaces of the several absorbers were accurately measured in an extrapolation ionization chamber. This instrument was very generously made available in operating condition by Dr. Randall S. Caswell. For a complete analysis of the instrument the reader is referred to Caswell's thesis (C-2), although its basic features and the nature of the measurements which I made with it are described in some detail in Appendix 10. Basically, this extrapolation chamber is a parallel plate ionization chamber, of variable plate separation $d$. The source, or the surface of the absorber over the source, forms one electrode. Measurements were made, with a vibrating reed electrometer, of the ionization current in the air gap as a function of $d$ between $d = \text{about } 4 \text{ mm to about } 1 \text{ mm}$. From these data, ionization current per unit volume was plotted as a function of $d$ and extrapolated to $d = 0$. This extrapolated ionization current per unit volume was then converted so as to give, in units of 100 ergs/gm hr, the dosage rate (averaged over a central
area of 1" diameter) which would have been experienced at the surface of a piece of lucite in contact with the absorber surface.

Table 11

Absorbers for Thin Sources

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Absorber thickness</th>
<th>Number of half thicknesses</th>
</tr>
</thead>
<tbody>
<tr>
<td>S\textsuperscript{35}</td>
<td>1.28 mg/cm\textsuperscript{2}</td>
<td>0.41</td>
</tr>
<tr>
<td></td>
<td>3.0</td>
<td>0.96</td>
</tr>
<tr>
<td></td>
<td>6.5</td>
<td>2.07</td>
</tr>
<tr>
<td>W\textsuperscript{185}</td>
<td>1.6</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>3.8</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td>7.0</td>
<td>0.57</td>
</tr>
<tr>
<td></td>
<td>9.9</td>
<td>0.81</td>
</tr>
<tr>
<td>Tl\textsuperscript{204}</td>
<td>2.2</td>
<td>0.077</td>
</tr>
<tr>
<td></td>
<td>7.5</td>
<td>0.26</td>
</tr>
<tr>
<td></td>
<td>17.9</td>
<td>0.63</td>
</tr>
<tr>
<td></td>
<td>31.5</td>
<td>1.10</td>
</tr>
<tr>
<td>Y\textsuperscript{91}</td>
<td>3.1</td>
<td>0.041</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>0.23</td>
</tr>
<tr>
<td></td>
<td>48</td>
<td>0.64</td>
</tr>
<tr>
<td></td>
<td>96</td>
<td>1.27</td>
</tr>
</tbody>
</table>

2. Photographic measurements

After calibration, the sources, with associated absorbers, were laid on film for appropriate exposure times (ranging from about 1 hour to about 2 days). The films were calibrated with the Sr\textsuperscript{90} - Y\textsuperscript{90} radiator for the usual stand-
ardization purposes, and for fading as well, which never amounted to more than a few percent. Processing of the films was carried out in a manner identical with the mono-kinetic electron exposures. The developed film strips were then densitometered over a systematic grid of 23 points covering the central 1" diameter area of exposure, in order to average the exposure over the same area whose surface dosage rate was averaged by the extrapolation chamber. From these readings, and the Sr$^{90}$ - Y$^{90}$ H and D curve, the exposure required for D = 0.3 was found, and the dosage sensitivity computed.

3. Results of continuous spectrum exposures

Two of the films, Eastman No Screen and Cine Positive #5302, were exposed to each source through the whole set of 3 or 4 different absorber thicknesses per source. The other 3 were exposed to each source through the thinnest absorber only. The 2 films on which the most data were taken are considered first. These results aid in the interpretation of the more limited data obtained on the other 3 films.

a. Eastman No Screen

The measurements made on Eastman No Screen gave the information shown in Fig. 43, where dosage sensitivity is plotted against absorber thickness expressed in units of half thicknesses. In evaluating the significance of these curves, one must keep in mind that such data on a film ideal for the dosimetry of $\beta$ rays would consist of a single horizontal line for all isotopes at all absorber thicknesses. The actual curves deviate from the ideal in the variation of sensitivity both with the isotope and with the absorber thickness.

It is seen that the dosage sensitivity for the different isotopes decreases with $\beta$ ray energy. At the
higher energies, the dependence on energy is not nearly so great as at the lower energies. This behavior is just what we would expect on the basis of the monokinetic results already presented. We have seen there that two effects combine to make diffuse incidence dosage sensitivity lower at low energies than at high. First, absorption of the \( \beta \) rays in the emulsion is more important at low energies. Second, a higher energy dissipation per grain exposed exists at low energies than at high. We also saw in connection with the monokinetic data that above 1 Mev dosage sensitivity is substantially independent of energy.

One sees, also, in Fig. 43, that the sensitivity at first increases with increasing absorption, but later begins to decrease. Both of these conditions can be understood in terms of measurements and ideas already presented. To consider first the initial increase in sensitivity: We have seen that the placing of an absorber over a thin source changes the angular distribution from what has been called an isotropic distribution to a \( \cos \theta \) distribution. The significant feature of this change, for the present purposes, is that in the first distribution the high angles of incidence are emphasized more than in the second. We saw in the monokinetic angle of incidence measurements that at the lower energies, film is less sensitive for angular distributions emphasizing high angles of incidence. Consequently, the change in angular distribution caused by the absorber is in the direction of increasing sensitivity, as is observed. Some significant changes may also be produced in the shape of the energy spectrum by the initial absorption, especially at low energies where the film sensitivity is most dependent on energy. However, there is little evidence on this point even in Brownell's measurements. At greater absorber thicknesses, Brownell's data show that a decrease in average energy becomes significant. This probably accounts for the slight decrease in sensitivity at larger absorber thicknesses.
Figure 43
Eastman No Screen
Dosage Sensitivity vs Absorber Thickness
Figure #44
Cine Positive #5302
Dosage Sensitivity vs Absorber Thicknesses
b. Cine Positive #5302

The results on Cine Positive are shown in Fig. 44. The same features may be seen in these curves as in the No Screen data, except that the deviations from ideal are somewhat less pronounced. This fact is quite likely to be accounted for by the lower degree of β ray absorption in the thinner emulsion.

c. Eastman Type A, NTB, SWR

These three films were exposed to the 4 sources through only the thinnest absorber in each case. Because of rapid changes in angular distribution with absorber thickness, in this range of absorber thickness, these 4 values of sensitivity are not uniquely characteristic of the 4 isotopes. A better sensitivity value for intercomparison would be the peak sensitivity which occurs at a somewhat greater absorber thickness. The ratio of peak sensitivity to the sensitivity at minimum absorber thickness is undoubtedly a function almost exclusively of emulsion thickness, especially if the discrepancy has been correctly ascribed to differences in angular distribution. Since the emulsion thicknesses of Type A and NTB lie between those of No Screen and Cine Positive, it is possible to estimate the peak sensitivities of the former 2 films from the more complete data on the latter two. For this purpose, I have used a conversion factor which, for each isotope, is the average ratio, in Cine Positive and No Screen, of the peak sensitivity to the sensitivity at minimum absorber thickness. When, for example, the sensitivity of Type A for $\text{Y}^{91}$ at minimum absorber thickness is multiplied by the $\text{Y}^{91}$ conversion factor, the result should be a reasonably close approximation to the peak $\text{Y}^{91}$ sensitivity of Type A. Table 12 gives the experimental data on Type A, NTB, and SWR, with the estimated peak sensitivities of the first 2 films for each of the 4 isotopes.
Table 12
Dosage Sensitivity of Type A, NTB, SWR
(All sensitivity units are $(100 \text{ ergs}/\text{cm})^{-1}$)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Type A Sens.</th>
<th>Type A Peak sens.</th>
<th>NTB Sens.</th>
<th>NTB Peak sens.</th>
<th>SWR Sens.</th>
<th>SWR Peak sens.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>with min. absorb.</td>
<td></td>
<td>with min. absorb.</td>
<td></td>
<td>with min. absorb.</td>
<td></td>
</tr>
<tr>
<td>$^{35}S$</td>
<td>0.116</td>
<td>0.148</td>
<td>0.060</td>
<td>0.077</td>
<td>0.024</td>
<td></td>
</tr>
<tr>
<td>$^{185}W$</td>
<td>0.25</td>
<td>0.32</td>
<td>0.088</td>
<td>0.114</td>
<td>0.043</td>
<td></td>
</tr>
<tr>
<td>$^{204}Tl$</td>
<td>0.26</td>
<td>0.33</td>
<td>0.112</td>
<td>0.117</td>
<td>0.048</td>
<td></td>
</tr>
<tr>
<td>$^{91}Y$</td>
<td>0.39</td>
<td>0.41</td>
<td>0.111</td>
<td>0.117</td>
<td>0.070</td>
<td></td>
</tr>
</tbody>
</table>

From the data of Table 12, as from all other measurements comparing the different films, the energy response of NTB is seen to be more nearly ideal for dosimetry measurements than is that of any other film.

d. General conclusions from continuous spectrum exposures

The results obtained in these thin source measurements show, particularly at the high energies, that the continuous $\beta$ ray exposing efficiency does not change very rapidly with absorption of the $\beta$ rays. Consequently, fairly reliable $\beta$ ray dosimetry measurements can be made with film, even if the dosage sensitivity is a rather strong function of energy, provided only that the film is calibrated with a thick standard source containing the same isotope as is under experimental investigation. This is true for any spatial distribution of activity in the source, except the special case of a thin plane source very close to the film, with little or no absorber interposed. Even point sources of activity may be rather reliably measured under conditions where resolution is not too low, as is now to be
shown. Consider a point of activity within a thick tissue section contributing a dosage, at the tissue surface, which is to be measured with film. One might say, for present rough purposes, that if the active point were farther than 20µ from the tissue surface, film resolving power would be adequate, whereas if it were less than 20µ from the surface, resolution limitations would begin to decrease the accuracy of quantitative dosage measurements. (For example, if the point were right on the surface, the dosage rate there would be infinite, whereas film density would of course be limited by less than perfect resolution resulting from finite emulsion thickness.) Let us take now a point source of a high energy isotope, say Y⁹¹, at a distance of 20µ from the surface. One sees from the scattering data of Table 1 that the high energy β rays would not be scattered much in 2 mg/cm² of tissue, so that immediately below the active point the β rays would be incident mostly at small angles of incidence, rather than diffusely like the β rays from the thick Y⁹¹ calibration source which would be establishing absolute sensitivity in this experiment. However, one sees from the monokinetic dosage sensitivity curves of this chapter that at high energies (perhaps greater than about 0.3 Mev) the film dosage sensitivity is approximately the same for both perpendicular and diffuse incidence. At lower energies, where film sensitivity is strongly dependent on the angle of incidence, Table 1 shows that scattering is important in even 2 mg/cm² of tissue, so that beneath the point source the β rays are tending to approach diffuse incidence in their angular distribution. Hence, for both the low and high energy β particles from this point source, the film response would not be very different from the diffuse incidence situation, and calibration with diffusely incident β rays from the thick standard source would be justified.
E. Comparison of Monokinetic and Continuous Spectrum Data

The monokinetic and continuous spectrum data give two completely independent measurements on the dosage sensitivities of these films. If the angular distribution and the energy spectrum of the β rays from one of the thin sources were known, it should be possible to calculate, from the monokinetic and angle of incidence measurements, the dosage sensitivities of the various films for exposures to that source. I have made the comparison between the two different methods of sensitivity measurement for No Screen and Cine Positive in the following way. The energies of P32 and Y91 are sufficiently close, and sufficiently high, that the dosage sensitivity for these two spectra should be substantially identical. Brownell has given data on the angular distribution and spectral shape of P32 under various conditions of absorption and backscatter. I have taken the following conditions for computations: thick lucite backscatterer, 90 mg/cm² lucite absorber, spectrum measured for β's emitted at 30° with perpendicular to absorber. The angular distribution is essentially cosθ at this absorber thickness, which would give diffuse incidence on the film if this source were exposed to film. The spectral shape is slightly different at different angles of emission, so that 30° was chosen as being a more or less average value. Using this spectrum and the rate of energy loss data of Fig. 4, I found the relative surface dosage which was contributed by the β rays at each energy. Then, from the monokinetic diffuse incidence dosage sensitivity curves, I computed the film sensitivity which would exist for such a source. This computed value of the dosage sensitivity is to be compared with the measured sensitivity for the Y91 thin source under 80 mg/cm² absorption, which would represent, within a few percent, Brownell's P32 source under the conditions used in the calculation.
For No Screen, the computed sensitivity was 7% high; for Cine Positive, it was 33% high. The somewhat large discrepancy in the case of Cine Positive is no doubt due at least in part to inaccuracies in the angle of incidence measurements. As previously mentioned, these measurements are the least accurate in the whole thesis, and seem to exaggerate the high energy diffuse incidence sensitivity. In any case, a very large number of different measurements went into the final comparison. On the one hand were the extrapolation chamber and the film measurements on the continuous spectrum sources. On the other hand were Brownell's $P_{32}$ measurements, my spectrograph spectrum determination, and my spectrograph film measurements requiring an integration over angle of incidence and energy. In view of the experimental difficulties, I am satisfied that the discrepancies between the 2 sets of results are not so large as to suggest basic fallacies in either.

F. Reciprocity Law

As previously stated, the weight of previous experimental evidence in support of the reciprocity law in electron exposures was sufficiently great that this law was assumed in my own experiments. However, I have made a test of the law in the case of the one film Cine Positive #5302. This film was selected as being the most likely of the 5 examined to deviate from the law, for the reason that its initial D vs. E curve shape indicated a larger average number (1.2) of hits per grain than any other film. A single hit event could not possibly give reciprocity law failure, whereas the multi-hit light exposure does, as discussed in Chap. 4. The test was made using the Sr$^{90} - Y^{90}$ radiator described in Appendix 4. The ratio of activities of the sources in the radiator had been accurately determined with the proportional counter, and in particular, that between the first and fifth had been found to be 24.2. Two
exposures were therefore made on a Cine Positive film strip, in the time ratio of 24.2, and the film was processed as usual. If the reciprocity law were valid and the test accurate, the fifth and first sources, respectively, in the two different exposures of the radiator, would have produced the same blackening. Actually, of course, identical densities were not obtained, and for this test the discrepancy was to be interpreted in terms of a reciprocity law failure. Making use of the known exposure intensity and exposure time ratios, and of the measured densities, it was possible to find the Schwarzchild constant $p$ in the expression $E_{\text{eff}} = It^p$ (see Chap. 4.). The results obtained in 2 repetitions of this test, together with the estimated error in the measurements, are shown in Table 13.

Table 13

<table>
<thead>
<tr>
<th>Test number</th>
<th>$p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.002 ± 0.006</td>
</tr>
<tr>
<td>2</td>
<td>0.998 ± 0.006</td>
</tr>
</tbody>
</table>

The reciprocity law therefore appears to be very nearly valid with Cine Positive in the intensity range of roughly $1.2 \times 10^6$ to $3 \times 10^7 \, \beta\text{s/cm}^2\text{min}$.

G. Accuracy of Small Area Density Measurements

Some of the photographic properties limiting resolution have been discussed in Chaps. 4 and 5. In order to investigate the limitation imposed by graininess under practical conditions, a set of experiments has recently been carried out in this laboratory by Mr. C. J. Maletskos and the laboratory staff, with my collaboration. The results of this investigation are of interest to the problems
of this thesis, and are briefly reported in this section.

The goal of these experiments was to determine the magnitude of fluctuations in repeated small area density measurements over a uniformly exposed region of film, and to find how these fluctuations depended on the average density. Four uniform exposures, increasing in magnitude by factors of 2, were made on double emulsion Eastman No Screen, using the very homogeneous Y\textsuperscript{91} thin source previously described. The uniformity of exposure was further insured by leaving a 1/8" gap between source and film. Approximately 300 to 400 density measurements were made in the center of each exposure, using a densitometer aperture area of $2.3 \times 10^{-4}$ cm\textsuperscript{2}. A graph was then plotted of frequency of occurrence vs. density reading, giving an approximately Gaussian distribution for the 300 to 400 density measurements. From this graph, the standard deviation of the density was estimated, and converted, via the H and D curve, to the associated standard deviation in exposure. This error in associated exposure, rather than in density, is the quantity of interest when one is making actual quantitative autoradiographic measurements. The results are shown in the second column of Table 14, where the standard deviation in exposure, $\sigma_{E,\text{meas.}}$, is expressed as a percent of the actual average exposure.

It is possible to compare this measured standard deviation with the value calculated as in Chap. 5 on the basis of the statistical character of the exposure. Using the experimental results reported earlier, the average number of electrons/cm\textsuperscript{2} in each of the 4 exposures was estimated, and from this the standard deviation in the number per $2.3 \times 10^{-4}$ cm\textsuperscript{2} was calculated. These calculations are shown in the fourth column of Table 14.

The measured and calculated values of $\sigma_{E}$ are in very good agreement, in terms of the accuracy sought when
Table 14

Standard Deviation of Small Area Exposures

<table>
<thead>
<tr>
<th>Av. net D</th>
<th>$\sigma_{E, \text{meas.}}$</th>
<th>Av. # elecs/cm²</th>
<th>$\sigma_{E, \text{calc.}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.40</td>
<td>3.6 %</td>
<td>$3.0 \times 10^6$</td>
<td>3.8 %</td>
</tr>
<tr>
<td>0.80</td>
<td>2.1</td>
<td>6.0</td>
<td>2.7</td>
</tr>
<tr>
<td>1.56</td>
<td>1.5</td>
<td>12.0</td>
<td>1.9</td>
</tr>
<tr>
<td>2.93</td>
<td>1.1</td>
<td>24.0</td>
<td>1.3</td>
</tr>
</tbody>
</table>
the experiment was designed. One may conclude, therefore, that the fluctuations obtained in these small aperture density measurements are at least in large part a result of the statistical character of the exposure, and are therefore unavoidable.

H. Summary of Conclusions

The detailed conclusions drawn from the experimental investigations into the fundamental processes of photographic β ray dosimetry have been given throughout this chapter. In this section only the major conclusions are summarized in a general way.

1. For most practical conditions, the H and D curve shape is independent of electron energy.

2. Dosage sensitivity for perpendicularly incident electrons is a strong function of energy at low energies. It starts at 0 for 0 energy, rises to a peak at roughly 0.1 Mev, and thereafter decreases to a plateau of about 1/3 to 2/3 maximum sensitivity for energies greater than approximately 0.5 Mev. All these numerical values depend on the particular film.

3. Dosage sensitivity is a strong function of the angle of incidence of the electrons at low energies and high angles of incidence. For the special case of diffuse incidence, which is approximated in exposures to thick sources in contact with the emulsion, dosage sensitivity increases monotonically with energy, reaching a flat maximum at about 1 Mev. The initial rise between 0 and 0.1 Mev is rapid, reaching perhaps 2/3 the maximum. This is followed by a gradual increase up to the flat maximum. Again, the numerical values depend on the particular film.

4. Sensitivity increases monotonically with development time. The increase is probably somewhat less rapid the lower the electron energy producing the exposure.

5. For continuous spectrum β rays, absorption of
the β rays by sheets of matter has relatively little effect on their exposing efficiency, particularly at high energies. This is a very useful condition in the accurate calibration of β ray exposures for dosimetry measurements, and is probably the most important practical conclusion of this thesis.

6. Of the 5 films tested, the response of NTB stripping film to diffusely incident electrons was found to be the least dependent on electron energy. Therefore NTB is the best emulsion for purposes of dosimetry of diffusely incident electrons, provided its rather low sensitivity, and perhaps poor reproducibility, are not handicaps.

7. Overall reproducibility of β ray film exposures is good within a few percent when calibration with standard sources is carried out.

I. Suggestions for Future Investigations of Fundamental Emulsion Properties

I believe that the chief extension of the research work of this thesis should be into actual dosimetry applications, rather than into more detailed analysis of fundamental emulsion properties. A few suggestions pertinent to applications are given in the next chapter. However, there is at least one problem of a fundamental nature in which further investigation would be very valuable.

The most useful addition to my research, along fundamental lines, would be an extension, to thicker absorbers, of my measurements on the effect of absorber thickness on β ray exposing efficiency. Data collected thus far reach only to approximately a single half thickness of absorber, whereas in practical problems one may wish to study β ray dosage at considerably greater distances from the source. (An example might be the investigation of depth
dose from a β ray applicator of the type described in the next chapter.) It is quite probable that for most films a plot of dosage sensitivity vs. absorber thickness (such as Fig. 43) would remain substantially flat to much greater absorber thicknesses than I have studied. If this were found to be the case, it would increase confidence in the calibration of unknown exposures made through any thickness of absorber, using a single standard source (e.g., a homogeneous thick source) whose β rays have passed through only a moderate thickness of absorber.
dose from a β ray applicator of the type described in the next chapter.) It is quite probable that for most films a plot of dosage sensitivity vs. absorber thickness (such as Fig. 43) would remain substantially flat to much greater absorber thicknesses than I have studied. If this were found to be the case, it would increase confidence in the calibration of unknown exposures made through any thickness of absorber, using a single standard source (e.g., a homogeneous thick source) whose β rays have passed through only a moderate thickness of absorber.
CHAPTER 8: APPLICATIONS

A. Beta Ray Applicators

One of the practical applications to which the foregoing results have been put is the measurement, with photographic film, of the surface dosage rate of β-ray applicators. These devices are strong sources of energetic β-rays with which to carry out clinical radiation therapy on surface lesions. It seems probable that more precise techniques are available for this purpose, in particular the extrapolation ionization chamber. However, film has 2 chief arguments in its favor. First, its high resolution permits one to study in detail the quantitative variation of dosage over the surface of the applicator. The nonuniform distribution of activity over the surface of currently available commercial applicators makes this a significant question. Second, the convenience of film, once calibration sources are obtained, will probably permit its use in clinical problems of dosimetry either where extrapolation chambers are not available or where the geometrical conditions of interest cannot be reproduced with an extrapolation chamber.

Investigations were made on 2 applicators. The first, loaned by the Canadian Radium and Uranium Corp., contained RaD and RaE; the second, loaned by Tracerlab, Inc., contained Sr$^{90}$-Y$^{90}$.

1. RaD, RaE Applicator

The RaE applicator had the following approximate construction: The container of the activity was a shallow monel metal cup approximately 6 mm in diameter. In this, roughly 15 mc of RaD (0.025 Mev) and 15 mc of RaE (1.17 Mev) were deposited, as a mixture of approximately 3 mg PbSO$_4$ and 2 mg silver, on a silver foil backing (thickness unspeci-
fied). A 0.05 mm (13.5 mg/cm²) Al foil served as the window through which the RaE (but not RaD) β rays were emitted. The problem was to measure the radiation dosage rate at the surface of the aluminum window.

The experimental approach will be clear enough from the discussion of the preceding chapters. A RaD, RaE calibration source was prepared, fairly closely duplicating the pertinent features of the applicator, and its surface dosage rate was determined in the extrapolation chamber. (The details of this calibration source are described in Appendix 9.) This was exposed to film along with the applicator and the Sr⁹⁰-Y⁹⁰ radiator. The calibration sources established the absolute sensitivity of the film to RaE β rays, while the radiator gave the H and D curve so as to permit comparison of exposures producing different densities. Cine Positive #5302 was chosen as the film because of its relatively good sensitivity vs. β energy characteristics, its low absolute sensitivity, and its rather reliable reproducibility. As a result of the very high activity of the applicator, short exposures of about 3 seconds duration were required. These were accurately accomplished by means of a simple jig to hold the applicator and a micro switch-stop clock arrangement to time the duration of contact between film and applicator. Upon investigation of the switching process, it was found that the clock started and stopped with delays of less than 0.05 second. The positioning of the film could also be very rapidly accomplished. This could be demonstrated by the fact that as the film slid over the applicator surface to its position of exposure, no detectable blackening was produced outside the region receiving the 3 second exposure. An exposure equivalent to 1% of 3 seconds would have been readily detectable. Six identical exposures were made of both the calibration source and the radiator, and 3 of the Sr⁹⁰-Y⁹⁰ radiator. A sample autoradiograph is shown in Fig. 45. This sample has been
deliberately underexposed to give a density suitable for visual inspection. The nonuniformity of the activity over the surface of the applicator is readily apparent.

![Image of applicator surface]

**Fig. 45**

The data were analyzed in the usual manner, by comparison of the resultant photographic densities, so as to determine in absolute units the dosage rate at representative points on the surface of the applicator. These representative points were (1) the most active spot, (2) the geometrical center, and (3) - (6) 4 points displaced 0.13 cm from the geometrical center along 2 mutually perpendicular lines through the center. The densitometer was used with an aperture of 1/16" diameter, although an aperture as small as 0.004" could have been used if higher resolution had been desired. To illustrate the degree of reproducibility of the measurements on the applicator itself, all the data on the 6 exposures are listed below in Table 15.

The absolute error attributable to the averages is in all probability less than 10%, and the relative error less than 5%. The most likely source of consistent error is the existence of differences between film sensitivity for the applicator β rays and for the calibration source β rays, respectively, made possible by differences in construction between the 2 sources. However, this error should be only a few percent, because of the relative independence of film sensitivity from absorber thickness, etc., at the energy of RaE. The calibration source surface dosage rate
was probably measured with good accuracy; the timing was sufficiently accurate; and random errors are seen to be rather small.

Table 15
Dosage Rates at Surface of RaE Applicator
(All units are 100 ergs/gm min)

<table>
<thead>
<tr>
<th>Test</th>
<th>(1)</th>
<th>(2)</th>
<th>(3)</th>
<th>(4)</th>
<th>(5)</th>
<th>(6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.29x10^3</td>
<td>2.04x10^3</td>
<td>2.18x10^3</td>
<td>2.04x10^3</td>
<td>1.32x10^3</td>
<td>2.06x10^3</td>
</tr>
<tr>
<td>2</td>
<td>2.31</td>
<td>2.09</td>
<td>2.16</td>
<td>2.04</td>
<td>1.48</td>
<td>2.04</td>
</tr>
<tr>
<td>3</td>
<td>2.45</td>
<td>2.13</td>
<td>2.36</td>
<td>2.04</td>
<td>1.36</td>
<td>2.23</td>
</tr>
<tr>
<td>4</td>
<td>2.54</td>
<td>2.18</td>
<td>2.32</td>
<td>2.16</td>
<td>1.39</td>
<td>2.18</td>
</tr>
<tr>
<td>5</td>
<td>2.54</td>
<td>2.18</td>
<td>----</td>
<td>----</td>
<td>----</td>
<td>----</td>
</tr>
<tr>
<td>6</td>
<td>2.52</td>
<td>2.07</td>
<td>----</td>
<td>----</td>
<td>----</td>
<td>----</td>
</tr>
<tr>
<td>Av.</td>
<td>2.45</td>
<td>2.11</td>
<td>2.25</td>
<td>2.07</td>
<td>1.39</td>
<td>2.13</td>
</tr>
</tbody>
</table>

2. Sr\(^{90}\) - Y\(^{90}\) applicator

The Sr\(^{90}\) - Y\(^{90}\) applicator was calibrated in the same manner, with the exception that a different calibration source was used. This applicator consisted of approximately 15 mc of Sr\(^{90}\) and 15 mc of Y\(^{90}\) in a stainless steel cup 0.5" in diameter. The activity was mounted on a ceramic backing and exposed through a 100 mg/cm\(^2\) aluminum window.

The calibration source in this measurement was not ideal. For lack of time to prepare a Sr\(^{90}\) - Y\(^{90}\) source, it was decided to make use of the Y\(^{91}\) source already available, with a small correction estimated to account for differences from the applicator \(\beta\) rays. The correction was the following. If one assumes exponential absorption of the 0.61 Mev Sr\(^{90}\) and 2.3 Mev Y\(^{90}\) \(\beta\) rays in the appli-
cator window, one can compute, on the basis of the formula
\[ \mu/\rho = 17 E^{-1.43} \text{ cm}^2/\text{gm}, \]
that approximately 8% of the surface dosage rate will be caused by Sr\(^{90}\) and 92% by Y\(^{90}\).

It was assumed, rather arbitrarily from Fig. 44, that the sensitivity of Cine Positive #5302 for Sr\(^{90}\) \(\beta\) rays absorbed through 4 half thicknesses would be only \(2/3\) that of Y\(^{90}\) absorbed through 1 half thickness. Therefore the sensitivity of the film for the applicator \(\beta\) rays would be \(1/3\) of 8% lower than for pure Y\(^{90}\) \(\beta\) rays. The sensitivity of the film for Y\(^{90}\) \(\beta\) rays was estimated by means of the information for other isotopes from Fig. 44. If one plots, from Fig. 44, the log of maximum dosage sensitivity for each of the 4 isotopes vs. \(1/E_{\text{max}}\), the points fall on a reasonably straight line. The extrapolation from Y\(^{91}\) sensitivity to Y\(^{90}\) sensitivity is very short and amounts to only a 3% increase. The absolute errors associated with these two oppositely directed corrections of 3% would seem to be sufficiently small to permit a fairly accurate calibration of the Sr\(^{90}\) - Y\(^{90}\) applicator by means of the Y\(^{91}\) \(\beta\) rays.

The dosage rates at representative points on the surface of this applicator, analogously located with those in the RaD, RaE applicator, are shown in Table 16. Only average values are given.

<table>
<thead>
<tr>
<th>Site</th>
<th>(1)</th>
<th>(2)</th>
<th>(3)</th>
<th>(4)</th>
<th>(5)</th>
<th>(6)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dosage rate</td>
<td>(2.01 \times 10^3)</td>
<td>(1.75 \times 10^3)</td>
<td>(1.78 \times 10^3)</td>
<td>(1.66 \times 10^3)</td>
<td>(2.03 \times 10^3)</td>
<td>(1.90 \times 10^3)</td>
</tr>
</tbody>
</table>

The apparent anomaly of site (1), the most active spot, having lower dosage rate than site (5) is caused by
the availability of only 4 separate measurements at site (5), whereas 6 are averaged for site (1).

It seems justifiable to attribute an absolute error of less than 15% to these figures, and a relative error of less than 5%. The absolute error is quite likely greater than that for the RaD, RaE data because the calibration source here used was not as reliable as for the other applicator.

The results from the Sr\textsuperscript{90} - Y\textsuperscript{90} applicator have been discussed in some detail for the reason that they furnish the only check, against completely independent measurements, of the experimental work reported in this thesis. This same applicator was analyzed with an extrapolation chamber by the manufacturers, and was claimed by them to have a surface dosage rate at the center of $2.16 \times 10^3$ units of 100 ergs/gm min. This is to be compared with my value of $1.77 \times 10^3$. They estimate their absolute error to be $\pm 10\%$, with the probability that their value is too high rather than too low. Much of this error they attributed to a certain anomalous behavior of the extrapolation chamber which was never satisfactorily explained. (The problem was a divergent extrapolation slope in their extrapolation of current per unit volume to zero volume. This problem is considered in detail in Appendix 10.) If the manufacturer's calibration is correct, mine is in error by 19%. However, in view of the experimental errors attributed to the respective measurements, the results cannot be said to be in disagreement.
B. Dosage from $^{45}\text{Ca}$ in Dog Bones

The second application which has been made of photographic film in $\beta$ ray dosimetry is the measurement of dosage rates associated with deposits of $^{45}\text{Ca}$ in dog bones. This is the problem, mentioned in the introduction, which originally suggested this thesis. Chronologically, this application came before the technique had been carefully investigated; the accuracy of the results obtained may only now be evaluated.

The specific experiment to which the biological problem reduced itself was this: For a given concentration of $^{45}\text{Ca}$ in the blood of a dog, what will be the maximum local radiation dosage rate resulting in the bone, and what will be the effect thereon of calcium mobilizing salts and hormones? The experiment was carried out jointly with Dr. Brown M. Dobyns of Massachusetts General Hospital, who was responsible for many of the biological aspects not here reported in detail.

1. Experimental procedure

Measurements were made successively on 2 dogs. The first was a 4 to 5 year old, 5.7 kg, mongrel, while the second was a 4 month old, 7 kg mongrel. The pertinent biological procedure followed with each of these dogs was the following:

Dog I was given, by stomach tube, a dose of 890 mg of low specific activity Ca$^{45}$ in a CaCl$_2$ solution. Immediately thereafter, and throughout the day, blood samples were taken for activity and calcium concentration measurements. On the 11th day a rib and a tooth were resected for dosage studies. On the 14th day, 890 mg of inactive calcium in CaCl$_2$ solution were administered, by stomach tube, in an attempt to replace some of the active Ca$^{45}$ previously
laid down in the bones. On the 18th day a second rib was resected for dosimetry measurements. On the 20th day 700 mg of NH₄Cl were administered in an attempt to remove the active Ca⁴⁵ through changing the blood pH. On the 25th day a third rib was removed. On the 31st and 32nd days, a total of 6 cc of parathyroid hormone were injected, again as an attempt to mobilize the Ca⁴⁵ in the bones. On the 38th day the dog was sacrificed, and most of its carcass became available for dosimetry measurements.

Dog II was subjected to a similar set of experiments. It was given, by stomach tube, a dose of about 20 mg (220 μc) of Ca⁴⁵ in a CaCl₂ solution. The blood activity was followed for a week. On the 21st day 2 ribs were resected for dosage measurements. On the 28th day a 3rd rib was removed. On the 30th, 31st, and 34th days a total of 4 cc of parathyroid hormone were injected as before. On the 39th day the animal was sacrificed.

The experimental procedure employed in the autoradiographic measurement of local radiation dosage rates in these bones was described in some detail in a paper by Dudley and Dobyns (D-4). In brief, the method was the following: The bones were sawed in half, cleaned of fat and marrow by soaking in ether, and ground down to a smooth flat surface on sandpaper. (This method of preparation, although crude, was considered adequate in view of the fact that only regions of maximum dosage were of interest in the present study. Grinding might redistribute activity slightly, but would be incapable of increasing the maximum local concentration of activity.) The bones were then clamped to sheets of double emulsion Eastman No Screen X-ray Film for the production of an autoradiograph. This film was chosen solely because its high sensitivity was desirable at the low activity levels involved. Single emulsion film would have been preferable for these dosimetry purposes, but only a negligible fraction of the Ca⁴⁵ β rays penetrate through the 32
mg/cm² of front emulsion and film base to the rear emulsion.

A set of calibration sources was prepared in the form of plaster of Paris discs containing a known fraction, per gram, of the Ca⁴⁵ activity fed to the animals. These sources, which quite closely duplicated bone in atomic constitution, were exposed on the same sheet of film as the active bones. They provided both the relative D vs. E curve shape and the absolute sensitivity calibration of the film. The exposures were developed (although without sufficient attention to agitation) and fixed approximately as recommended by the manufacturer.

The autoradiographs showed a highly nonuniform distribution of Ca⁴⁵ throughout the bone, much of the activity being located in the epiphyses or in small spots (presumably blood vessel sites) along the shaft of the bone. Comparison of densities between autoradiograph and calibration exposures, in the standard manner, permitted an evaluation of the local radiation dosage rates. Density measurements were made with a Hilger microphotometer, using a square aperture of area 0.003 mm².

2. Results

The results which were obtained in the above animal studies and which are pertinent to this thesis are the following:

a. Reproducibility of autoradiographic measurements

Since the autoradiograph spots to be densitometered were chosen because of their high densities, it is probable that film defects (such as background grain clumping in the small area of 0.003 mm²) would selectively enter the measurements, indicating higher local radiation dosage rates than actually existed. Therefore an attempt was made to estimate the magnitude of this error, and to check on the
overall reproducibility of the method. For this purpose duplicate autoradiographs of a rib were prepared and 20 of the blackest spots on the first autoradiograph and the 20 corresponding spots on the second autoradiograph were densitometered. "Exposures" due to film defects could selectively enter into the first set of measurements but not the second. When the 2 sets of data were compared, it was found that the first set indicated, on the average, a 10% greater local exposure than the second set. This, then, is the approximate error introduced by the selective contribution of film defects to local "exposure." Regarding reproducibility as such, it was found that for the 20 sets of 2 corresponding measurements, the average discrepancy between the 2 readings was 10%. This discrepancy seems quite moderate for this type of high resolution, low activity measurement.

b. Blood activity curve

The variation of blood activity with time was quite similar in the 2 dogs, although beyond the peak the activity dropped somewhat more rapidly in Dog II. The data for Dog II are shown in Fig. 46.

c. Maximum local dosage rates in bones

When the data of Fig. 46 are combined with data on the (chemical) calcium content of the blood, one obtains the specific activity of the blood as a function of time, and in particular, one finds the maximum specific activity. Finally, when this information is combined with the dosimetry measurements on the bones, one is able to make an estimate of the extent to which the active Ca$^{45}$ was diluted by inert calcium when passing into the bones. This is the primary information which was sought in the experiment.
Figure 46

DOG II BLOOD ACTIVITY
% Co$^{45}$ DOSE /cc PLASMA vs TIME AFTER DOSE
The most direct way of expressing the relationship between the measured bone dosage rate and the measured blood specific activity is in terms of the ratio: dosage rate in bone divided by dosage rate which would exist within a solid block of calcium having the maximum blood specific activity. This quantity is hereafter referred to as the fractional dosage factor. When the dilution of Ca$^{45}$ is expressed in this way, it deals with dosage, the property of interest, and does not require conversion of the measured bone dosage rate into the bone specific activity, a conversion which would not be completely reliable because of uncertainties in the local bone composition. Since bone is roughly 20 to 30% calcium by weight, a fractional dosage factor of about 0.20 to 0.30 would exist even if the bone calcium specific activity were identical with the maximum blood specific activity.

The fractional dosage factors observed in various bones of the 2 dogs are shown in Table 17. The decrease in activity with time by radioactive decay has been eliminated from the data. It is a very interesting, although perhaps partly coincidental, fact, that the fractional dosage factors were closely similar in the 2 dogs, despite the differences in the ages of the animals and the quantities of administered calcium. It can be seen in these data that a relatively large amount of dilution takes place for the Ca$^{45}$ passing from blood to bone under the conditions of these experiments. In human administration this would of course represent a much desired safety factor for the use of Ca$^{45}$. To be precise, if humans were identical with these 2 dogs in their metabolism of calcium, a peak blood specific activity of roughly $10^{-3}$ μc/mg Ca could be permitted without exceeding the currently accepted tolerance of 0.3 reps per week at any point in the skeleton. On the other hand, these experiments have shown that dosage rates at local sites in a bone considerably exceed the average dosage rate throughout the bone. By digestion of these
same bones after autoradiography, followed by GM counter measurements of the average bone activity, it was found that in Dog I the maximum local dosage rate exceeded the average throughout the same bone by a factor of about 10 to 20, while in Dog II the same figure was approximately 5 to 10.

Table 17
Fractional Dosage of Ca$^{45}$ in Bones of Dogs I and II

<table>
<thead>
<tr>
<th>Bone</th>
<th>Time of resection (days after administration of Ca$^{45}$)</th>
<th>Fractional dosage factor</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>DOG I</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rib 1</td>
<td>11</td>
<td>7.7 x 10^{-3}</td>
</tr>
<tr>
<td>2</td>
<td>18</td>
<td>6.5</td>
</tr>
<tr>
<td>3</td>
<td>25</td>
<td>7.6</td>
</tr>
<tr>
<td>4</td>
<td>38</td>
<td>4.9</td>
</tr>
<tr>
<td>5</td>
<td>38</td>
<td>5.2</td>
</tr>
<tr>
<td>Tooth</td>
<td>11</td>
<td>0.7</td>
</tr>
<tr>
<td>Humerus</td>
<td>38</td>
<td>5.4</td>
</tr>
<tr>
<td>Upper jaw</td>
<td>38</td>
<td>9.4</td>
</tr>
<tr>
<td>Lower jaw</td>
<td>38</td>
<td>9.4</td>
</tr>
<tr>
<td><strong>DOG II</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rib 1</td>
<td>21</td>
<td>6.6 x 10^{-3}</td>
</tr>
<tr>
<td>3</td>
<td>28</td>
<td>8.2</td>
</tr>
<tr>
<td>4</td>
<td>40</td>
<td>6.3</td>
</tr>
<tr>
<td>5</td>
<td>40</td>
<td>5.9</td>
</tr>
<tr>
<td>Humerus</td>
<td>40</td>
<td>7.2</td>
</tr>
<tr>
<td>Tibia</td>
<td>40</td>
<td>8.3</td>
</tr>
<tr>
<td>Femur</td>
<td>40</td>
<td>10.0</td>
</tr>
</tbody>
</table>
d. Effect of decalcifying salts and hormones

On Dog I, autoradiographic measurements were made to determine whether or not the decalcifying salts and hormones were effective in reducing the maximum local dosage rates occurring within the bones. Comparison of the single maximum local dosage rate from rib to rib was not satisfactory simply for statistical reasons. Consequently, on each autoradiograph all the blackened spots were densitometered. From these measurements an average dosage rate was computed for the most active 8% of the spots (approximately 15). The fractional dosage factor associated with each of these "average maximum dosage rates" is listed in Table 18. Again, the effect of radioactive decay has been eliminated from the data. It can be seen that the dosage rates did decrease somewhat from rib to rib. However, this decrease was not very pronounced, and quite likely does not much exceed the normal rate of decrease which would have been observed had no treatment been given.

<table>
<thead>
<tr>
<th>Bone</th>
<th>Time of resection</th>
<th>Decalcifying treatment immediately preceding resection</th>
<th>Average maximum fractional dosage factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rib 1</td>
<td>11 days</td>
<td>None</td>
<td>6.4 x 10^{-3}</td>
</tr>
<tr>
<td>2</td>
<td>18</td>
<td>890 mg Ca in CaCl₂</td>
<td>5.8</td>
</tr>
<tr>
<td>3</td>
<td>25</td>
<td>700 mg NH₄Cl</td>
<td>5.6</td>
</tr>
<tr>
<td>4</td>
<td>38</td>
<td>6 cc parathyroid hormone</td>
<td>4.4</td>
</tr>
<tr>
<td>5</td>
<td>38</td>
<td>&quot;</td>
<td>4.6</td>
</tr>
</tbody>
</table>

e. Validity of measurements

The most significant inadequacy of these measurements is that they do not detect the radiation dosage, at
the point in question, which would have been contributed by the half of the bone which has been sawed away. This inadequacy is not peculiar to the autoradiographic technique of dosage measurement; it would exist for any other type of instrumentation in an analogous problem. It is a fundamental difficulty which could be circumvented only by slicing the bone in half, without loss of sawdust, and then autoradiographing both halves. Whether or not this is conceivably possible, I did not attempt to do it. In the absence of information regarding this dosage contribution from the sawed off bone, I have assumed in the above measurements that it is equal to the contribution from the half of the bone actually exposed to film.

Compared with the uncertainty just discussed, other errors are probably not large. Among the most important is the reproducibility and graininess problem already mentioned. A finer grained film would have been preferable had lower sensitivity been tolerable (as it may have been with the bones of Dog II). The imperfect dosage response of the film itself would introduce some error; however, one may estimate from the curves of Fig. 43 that this would not exceed 10 to 20%, inasmuch as the film was properly calibrated with a thick source of Ca$^{45}$ in phantom bone.

C. Suggestions for Future Applications

1. Depth dosage from β ray applicators

We expect in this laboratory to make one further application of these film dosimetry techniques in the immediate future. This will be a measurement of the dosage from the β ray applicators already described at depths up to several half thicknesses in a tissue-like medium. The procedure will be similar to that used in the determination of the surface dosage rates of the applicators, except for
the use of bakelite or cellulose acetate absorbers over the applicator surface and the substitution of NTB for Cine Positive film. This substitution is desirable in view of the more nearly ideal dosage response of NTB as a function of \( \beta \) ray energy, a matter of importance when the degree of absorption imposed on the applicator \( \beta \) rays is drastically different from that imposed on the calibration source \( \beta \) rays.

2. \( ^{131}I \) radiation dosage in thyroid

I have no doubt that there are a great many biological problems similar to our dog experiments in which the use of these film dosimetry techniques would be very useful. Perhaps the most obvious application is the measurement of \( ^{131}I \) \( \beta \) radiation dosage in the thyroid. A tremendous amount of effort has in the last 5 years been expended on studying, in both humans and animals, many phases of thyroid metabolism and the effects thereon of internal \( ^{131}I \) radiation. However, the dosimetry measurements have thus far been rather crude. For example, Seller and co-workers (S-5 1/2) have investigated the changes induced by \( ^{131}I \) radiation in the iodine metabolism of rats. Their dosimetry was accomplished by determining the total quantity of \( ^{131}I \) in the gland and the total mass of the gland, followed by a mathematical computation based on the assumptions of uniform distribution throughout a spherically shaped gland. Gorbman (G-2 1/2), in studying similar problems, comments on Seller's dosimetry technique: "Even this scheme errs somewhat in assuming, for purposes of mathematical convenience, that the thyroid is spherical - a liberty that makes the morphologist wince."

Out of consideration for the morphologist (and for the histologist who may be caused to suffer by the assumption of uniform distribution), I suggest the following procedure. The thyroid could be embedded according to the usual technique of preparation for autoradiography, and
sliced in half along the plane whose radiation dosage rate was in question. Both halves would then be laid on film for the production of autoradiographs. In this way, the dosage contributed by both halves at the point in question could be measured. (Thick sections, rather than thin microtomed sections, are necessary in order that all the β radiation which would in vivo reach the point of interest be permitted in this measurement to reach the film.) The film would be calibrated with exposures from thick standard sources having a uniform distribution and known concentration of I^{131} in an approximately tissue-like material, a situation which does permit a simple and accurate calculation of the surface dosage rate. (The thickness of the standard should be roughly that of the thyroid, in order to minimize differences between the small amounts of γ ray dosage present in each case.) Comparison between the photographic densities associated with the thyroid and calibration exposures, respectively, would then determine the local I^{131} dosage rates in the thyroid.

3. Standard sources

The problems, for doctors and biologists, of quantitative β ray dosimetry would be very much simplified if calibration sources were readily obtainable. It might be that the National Bureau of Standards or certain commercial enterprises could prepare a kit of such sources. This kit might consist of 4 radiators similar to my Sr^{90} - Y^{90} radiators described in Appendix 4, containing long lived β emitters of graded energies in a tissue-like material. For example, these isotopes might be considered:

(1) S^{35} 87d, 0.17 Mev
   or C^{14} 5000 y, 0.16 Mev

(2) W^{185} 70 d, 0.43 Mev
(3) $\text{Tl}^{204} \quad 3\gamma, 0.78 \text{ MeV}$

(4) $\text{Sr}^{90} - \text{Y}^{90}$ or $\text{Ru}^{106} - \text{Rh}^{106}$

$20\gamma, 0.6$ and $2.3 \text{ MeV, resp.}$

$1\gamma, 0.04$ and $3.5 \ (82\%)$,$2.3\ (18\%) \text{ MeV, resp.}$, plus $18\% \gamma$ rays

Group (4) contains essentially high energy isotopes, despite the multiple spectra. $80\% \left(\frac{2.3}{2.3 + .6}\right)$ of the surface dosage of the $\text{Sr}^{90} - \text{Y}^{90}$ source would be from the $2.3 \text{ MeV}$ spectrum and $99\%$ of the dosage from the $\text{Ru}^{106} - \text{Rh}^{106}$ would come from the 2 high energy spectra, for which film dosage sensitivity would be almost identical.

If the absolute surface dosage rates of these radiators were known, they would provide not only the H and D curve (essentially the same for all) but also a measurement of absolute film sensitivity. The 4 different energies would give enough information so that fairly accurate sensitivity interpolations could be made for isotopes of other energies. The ready availability of these calibration sources would remove $80\%$ of the difficulties of film dosimetry, and would permit rather accurate quantitative work to be carried out even in the less well equipped laboratories.
APPENDIX 1: SURFACE DOSAGE FROM BACKSCATTERED ELECTRONS

In this appendix is treated the radiation dosage contributed, in a very thin surface layer of a backscattering material, by the backscattered electrons. This dosage resulting from backscatter is considered as a fraction $f_d$ of the dosage contributed by the incident electrons, referred to as the incident dosage. It will be useful to study here 2 special cases of this problem. The first is an evaluation of $f_d$ for electrons incident on lucite diffusely; the second, for electrons incident on lucite perpendicularly. The methods used in this appendix for evaluating $f_d$ make free use of concepts and measurement techniques described in Chaps. 2 and 7.

The problem in principle can be solved in at least 2 different ways. The first method would consist of a direct measurement of $f_d$, using a technique such as the following: A beam of electrons would be passed through a very shallow, very thin walled, parallel plate ionization chamber. This would give the incident dosage. The backscattering material would then be brought up into contact with the rear chamber electrode, giving a measurement of incident plus backscattered dosage. The second method would consist of determining the angular and energy distribution of the backscattered electrons relative to the incident, and calculating $f_d$ by means of these data from the fundamental principles of dosage as a function of path length and specific ionization.

As far as I am aware, no direct measurements by the first method have been completed, although Bortner (B-19) is now performing this type of experiment with continuous spectrum $\beta$ rays at Oak Ridge. However, Baily (B-1) has made a somewhat related study and his data are useful in the present analysis, as shown shortly. The second approach
to the measurement of \( f_d \) can be made at least approximately from Brownell's data (8-23), which give the energy spectrum of \( P_{32} \) \( \beta \) rays when backscattered from a variety of materials.

**A. \( f_d \) for Diffusely Incident Electrons**

The problem of diffusely incident electrons is considered first. The most reliable method of analysis, using data currently available, seems to be a combination of the 2 methods discussed above. Baily's measurements were the following: His apparatus consisted of a parallel plate extrapolation ionization chamber (cf. Appendix 10). The lower electrode was water, in which a \( \beta \) emitting isotope could be dissolved. The upper electrode was replaceable, and Baily substituted various materials in this position ranging in Z from Be to Pb or U. In this way he was able to measure the ionization (which as discussed in Chap. 2 is closely related to dosage) in the very thin air gap of the chamber as a function of the Z of the backscattering electrode. Baily's results are shown in Table 19 for four isotopes: \( P_{32} \) (1.80 Mev), \( Sr_{89} \) (1.5 Mev), \( Ti_{204} \) (0.78 Mev), and \( S_{35} \) (0.17 Mev). In Fig. 47 are plotted the points for \( S_{35} \) and the average for the other 3 isotopes. I have normalized Baily's data, which were initially relative anyway, in the following way: the ionization observed with active electrode of water and backscattering electrode of atomic number \( Z + 1 \), \( (I_{W,Z+1}) \), is divided by the ionization which would be observed if both electrodes were water, \( (I_{W,W}) \). Where the material contains more than a single element, I have computed an average \( Z \), called \( \bar{Z} \), by the formula

\[
\bar{Z} = \frac{\sum p_i Z_i^2}{\sum p_i Z_i}
\]

where \( p_i \) is the fraction of atoms which have \( Z = Z_i \), and the summation is over all elements in the molecule.
Figure 47
IONIZATION IN EXTRAPOLATION CHAMBER
DEPENDENCE ON Z OF ELECTRODES

$I_{w,Z+1}/I_{w,w}$ vs $Z + 1$

- $P^{32}, Sr^{89}, Ti^{204}$
- $S^{35}$

Figure 48
DOSSAGE FROM BACKSCATTERED ELECTRONS
$f_d(Z)$ vs $Z$

- From Brownell
- From Baily

$f_d(W) = 0.40$
- 0.30
- 0.20

1 2 4 6 8 10 20 40 60 80 100

1 2 4 6 8 10 20 40 60 80 100
Table 19
Baily's Dosage Backscatter Data

<table>
<thead>
<tr>
<th>Backscattering material</th>
<th>Z</th>
<th>$P^{32}$</th>
<th>Ionization in Relative Units</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>$^{89}$Sr</td>
</tr>
<tr>
<td>Be</td>
<td>4</td>
<td>0.916</td>
<td>0.913</td>
</tr>
<tr>
<td>Paraffin</td>
<td>4.75</td>
<td>-----</td>
<td>-----</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>4.75</td>
<td>0.945</td>
<td>0.943</td>
</tr>
<tr>
<td>Polystyrene</td>
<td>5.28</td>
<td>0.932</td>
<td>0.921</td>
</tr>
<tr>
<td>Carbon</td>
<td>6</td>
<td>0.974</td>
<td>0.978</td>
</tr>
<tr>
<td>(Water)</td>
<td>(6.6)</td>
<td>(1.000)</td>
<td>(1.000)</td>
</tr>
<tr>
<td>Al</td>
<td>13</td>
<td>1.13</td>
<td>1.13</td>
</tr>
<tr>
<td>Fe</td>
<td>26</td>
<td>-----</td>
<td>-----</td>
</tr>
<tr>
<td>Brass</td>
<td>29.5</td>
<td>1.33</td>
<td>1.30</td>
</tr>
<tr>
<td>Sn</td>
<td>50</td>
<td>-----</td>
<td>1.42</td>
</tr>
<tr>
<td>Pb</td>
<td>82</td>
<td>1.56</td>
<td>1.50</td>
</tr>
<tr>
<td>U</td>
<td>92</td>
<td>-----</td>
<td>-----</td>
</tr>
</tbody>
</table>

The outstanding feature of these results is that the ionization in the thin air gap varies almost identically with $Z$ at energies between $P^{32}$ and $Tl^{204}$, and not too differently from these at $S^{35}$ energies. If these data included values for no backscatterer present ($\overline{Z} = 0$), they would give immediately a unique determination of $f_d$ for the whole range of $\overline{Z}$ and for the different energies. However, the actual extrapolation to $\overline{Z} = 0$ cannot be carried out reliably directly from Baily's data. In the absence of this or similar information, I am forced to make an assumption here which will have considerable influence on my experimental results on film sensitivity as presented in Chap. 7.
The assumption which I make is that the data for all 4 of these isotopes would extrapolate to the same value at \( Z = 0 \). This leads immediately to the conclusion that \( f_d \) for low Z materials is substantially independent of energy below about 2 Mev, and that an evaluation of \( f_d \) for one energy is applicable to all energies. (This probably appears at this point to be a reasonable assumption. However, it will be clearer further on that it is based on a rather long extrapolation, and is not as sound as could be desired.)

A second line of reasoning which makes it at least plausible that \( f_d \) be essentially independent of energy is this: \( f \), the backscattered fraction on a particle number rather than a dosage basis, is rather independent of energy (especially in low Z materials). The angular distribution of the backscattered electrons is approximately \( \cos \theta \), independent of energy. Then, if the energy of the backscattered electrons is close to that of the incident electrons, their specific ionization would be the same and \( f_d \) as well as \( f \) would be independent of energy. Unfortunately, this last link in the chain of reasoning is rather weak, for Brownell's results show the energy of electrons backscattered from lucite to be considerably decreased. However, if specific ionization were increased proportionately for electrons of all energies when backscattered, the independence of \( f \) from energy would still imply the independence of \( f_d \) from energy.

These 2 lines of reasoning, using Baily's measurements on \( f_d \) and other experimental data on \( f \), point toward the same conclusion that \( f_d \) is substantially independent of energy. This is henceforth assumed to be the case. The next step is the evaluation of \( f_d \).

The magnitude of \( f_d \) for \( P^{32} \beta \) rays (and therefore for all energies) may be estimated from Brownell's data.
used in conjunction with Baily’s. Brownell showed the number and energy of $^3$P $^3$ $\beta$ rays backscattered from various Z materials, when incident in an isotropic angular distribution from a thin source in contact with the backscatterer. What we would like to have is this same information for diffusely incident electrons. However, no such experimental data exist. In what follows I have therefore made 2 further assumptions: (1) the number and energy of electrons backscattered in the direction perpendicular to the source surface is identical in isotropic and diffuse incidence, and (2) in diffuse incidence the dosage backscattered at angle $\theta$ with the normal is the same fraction of the dosage incident at angle $\theta$, independent of $\theta$. The first assumption probably overestimates slightly the value of $f_d$ for diffuse incidence, since backscattering is actually less important in diffuse than in isotropic incidence. However, the discrepancy is probably less important in the perpendicular direction than in any other. The second assumption is reasonable in view of the fact that in the case of diffuse incidence the angular distribution of the backscattered electrons is very similar to that of the incident electrons; that the $\cos \theta$ distribution is the stable distribution for backscattered electrons has been shown in Chap. 2.

Using these assumptions, it is now possible to evaluate $f_d$ from Brownell’s measurements. I have done this by making use of his data for perpendicular backscattering in the following way: I have multiplied the number of electrons backscattered at a particular energy by the relative rate of energy loss at that energy, as taken from Fig. 4. When this is integrated over energy, it gives the total relative dosage backscattered perpendicular to the backscatterer. The total relative dosage incident perpendicular to the backscatterer may be computed in the same way, making use of the fact that the spectrum incident perpendicular
to the backscatterer is the same as that measured by Brownell as being emitted perpendicular to the source, with no backscatterer present. $f_d$ is the ratio of the backscattered to incident dosage, and was found to have the values in Table 20 for backscatterers of various $Z$.

Table 20

$F_d$ from Brownell's Data

<table>
<thead>
<tr>
<th>Backscattering material</th>
<th>$\overline{Z}$</th>
<th>$f_d$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lucite</td>
<td>5.85</td>
<td>0.19</td>
</tr>
<tr>
<td>Al</td>
<td>13</td>
<td>0.42</td>
</tr>
<tr>
<td>Cu</td>
<td>29</td>
<td>0.60</td>
</tr>
<tr>
<td>Sn</td>
<td>50</td>
<td>0.66</td>
</tr>
<tr>
<td>Pt</td>
<td>78</td>
<td>0.74</td>
</tr>
</tbody>
</table>

The value for lucite, which is the quantity desired, was thought to be the least reliable because much of the backscattered dosage was contributed by low energy electrons for whose abundance Brownell's results were not very accurate. Therefore, a comparison of these $f_d$ data with Baily's was made in such a way that $f_d$ for the materials other than lucite helped determine the value of $f_d$ for lucite. This is possible since Baily's results establish rather accurately the relative dependence of $f_d$ on $Z$, although not the absolute value of $f_d$ for any particular $Z$. The analysis was carried out as follows: Baily measured the ionization $I_{W,Z}$ for an active electrode of water ($W$) and a backscattering electrode of material having atomic number $Z$. $I_{W,Z}$ is the sum of the ionization produced by the $\beta$ rays on their first passage through the gap, together with that produced in repeated backscattering between the 2 electrodes. If the angular distribution of the $\beta$ rays is the same following each backscattering, and if $f_d$ is
independent of energy, then:

\[ I_{w,z} = K(1 + f_d(z) + f_d(z)f_d(w) + f_d^2(z)f_d(w) + \ldots) \]

Since \( K \) is unknown because the measurements are relative, it may be eliminated by taking the ratio of \( I_{w,z} \) with \( I_{w,w} \):

\[ \frac{I_{w,Z}}{I_{w,W}} = \frac{1 + f_d(z) + f_d(z)f_d(w) + f_d^2(z)f_d(w) + \ldots}{1 + f_d(w) + f_d^2(w) + f_d^3(w) + \ldots} \]

\( \frac{I_{w,Z}}{I_{w,W}} \) is just the quantity listed in Table 19.

Using this last equation, it is possible to find \( f_d(z) \) which will lead to Baily's values when any particular value is assumed for \( f_d(w) \). Brownell's values for \( f_d \) may then be compared with \( f_d(z) \), so that finally \( f_d \) for high \( Z \) backscatters (these high \( Z \) measurements being more accurate than the lucite measurements), using a reliable extrapolation based on Baily's measurements. In Fig. 48 are plotted, as a function of \( Z \), Brownell's \( f_d \) values, together with three curves for \( f_d(z) \) from Baily's data based on assumed values for \( f_d(w) = 0.20, 0.30, \) and 0.40, respectively. It can be seen that Brownell's points fit rather well (except lucite) with Baily's more accurate relative values for \( f_d(z) \) if \( f_d(w) = 0.30 \). The net result, then, of this rather devious argument, is this: Brownell's absolute but rather inaccurate values for \( f_d \) vary with \( Z \) as they should according to Baily's rather accurate but relative values for \( f_d \) as a function of \( Z \). This increases confidence in the accuracy of Brownell's \( f_d \) values, and permits an extrapolation of his more accurate high \( Z \) \( f_d \) values to \( f_d(\text{lucite}) \approx 0.28 \).

It can now be seen why one has some reservation in concluding from Baily's data that \( f_d \) is relatively in-
dependent of energy. There exists a function \( f_d(Z) \) such that Baily's data can be reconstructed for any specified absolute value of \( f_d(\text{lucite}) \). The best that one can say is that the "principle of minimum astonishment" leads one to believe that \( f_d(Z) \) is independent of energy (to the extent that Baily's results are independent of energy) rather than to believe that there is a different function for \( f_d(Z) \) at each energy which fortuitously reconstructs Baily's results.

The whole derivation of \( f_d \) is indeed filled with many unproved assumptions, but it seems to be the best that can be done. The percentage error in \( f_d \) is probably rather large, but where it is later used in evaluating my experimental data, it is only a correction term. I believe that the error introduced into the total dosage, incident and backscattered, as a result of the error in \( f_d \), is not excessive relative to the accuracy there required.

B. \( f_d \) for Perpendicularly Incident Electrons

In this section \( f_d \) is estimated for perpendicularly incident electrons. The analysis makes use of the second approach discussed in the introduction of the appendix, together with the results of the analysis, in the preceding section, of \( f_d \) for diffusely incident electrons.

It has been shown in Table 3 in Chap. 2 for normally incident electrons on carbon, that \( f_d \) the fraction of the number of incident particles which is backscattered, is equal very nearly to 0.06 at all energies up to at least 0.37 Mev. The continuous spectrum \( \beta \) ray measurements discussed in the same chapter, although not for normal incidence, show that up to energies at least as high as \( P^3_{2} \) the backscattering is substantially independent of energy. Certainly no large error is involved in extrapolating Ek-lund's measurements to higher energies, and stating that,
for normally incident electrons on C, \( f = 0.06 \) over the whole energy range at which I have made measurements requiring this correction (0.03 to 1.8 Mev). For lucite the value may be considered the same.

It has also been shown in Chap. 2 that normally incident electrons are backscattered with an essentially \( \cos \theta \) angular distribution. It can be shown by a simple geometrical analysis that because of the differences in angular distribution, the average path length of the \( \cos \theta \) backscattered electrons is just twice as great, in a very thin surface layer, as is the path length of the normally incident electrons. Therefore, if the energy (and consequently specific ionization) of the backscattered particles were the same as the incident particles, \( f_d \) for normally incident electrons on lucite would be equal to \( 2 \times 0.06 = 0.12 \).

It was shown in Table 4, Chap. 2, that for \( \beta \) rays from a thin source (isotropic angular distribution) \( f = 0.15 \). For diffusely incident electrons (\( \cos \theta \) angular distribution \( f \) would lie between its values for perpendicular and for isotropic angular distributions, respectively, or in the neighborhood of 0.12. Since the angular distribution of the backscattered electrons would be closely \( \cos \theta \), as discussed in Chap. 2, \( f_d \) would equal approximately 0.12 also if the energy were not decreased in the backscattering process.

We have therefore arrived at the conclusion that, were there no change in energy during backscattering, \( f_d \) would be very nearly the same (0.12) for both perpendicular and diffusely incident electrons. We have seen in the preceding section that \( f_d \) in diffuse incidence actually is equal to approximately 0.28, a value higher than 0.12 because of the decreased energy (increased specific ionization) of the backscattered electrons. The best we can do
in evaluating \( f_d \) for normal incidence is to assume that the average energy decrease of the electrons during back-scattering is nearly the same as it is in the case of diffuse incidence, leading to \( f_d = 0.28 \) at all energies up to 1.8 Mev for perpendicular as well as for diffuse incidence. The error in this assumption is not likely to be very great in terms of the accuracy later required.

For incidence under angular conditions other than perpendicular or diffuse, one is justified in expecting \( f_d \) to have closely the same value as in these 2 widely different special cases. This would be true at least under all conditions not approaching the third limiting case of grazing incidence, a case which I have not attempted to analyze. The underlying reason for this independence of \( f_d \) from the angular distribution of the incident electrons is that two opposing effects balance each other: as the angle of incidence increases, the fraction \( f \) of electrons backscattered increases, but their average path length, in a thin surface layer, decreases relative to the path length, in that layer, of the incident electrons.


In this appendix a theoretical derivation is carried out for the dosage in the neighborhood of a plane source of β rays, using a type of analysis taken from Evans (E-6). The basic assumption is made that the β rays travel in straight paths without scattering, but that they undergo exponential absorption.

The experimental conditions for which it will be useful to carry out calculations are those illustrated in Fig. 49.

A plane source is covered by one absorber of thickness a, density $\rho_1$, and linear absorption coefficient $\mu_1$, and by a second absorber of thickness s, density $\rho_2$, and linear absorption coefficient $\mu_2$. The application of Chap. 2 is a special case of this situation, only a single absorber being involved. In the application to which I will put this derivation in Appendix 10, the second absorber is air.

Consider an elementary ring on the source surface of radius $y (= r \sin \theta)$ and width $dy (= \frac{r \cos \theta}{\cos \theta})$. The number of β's/sec, $dN$, emitted from this ring is given by:

$$
\frac{dN}{d\theta} = k_1 2\pi y \, dy = k_1 2\pi r \sin \theta \frac{r \cos \theta}{\cos \theta}.
$$
An elementary volume of material \(2\), having area \(\Delta A\) and thickness \(\Delta s\), is located at \(s\). The fraction of the \(\beta\)'s from any point on the elementary ring of source which passes through \(\Delta A\) is given by:

\[
\frac{\Delta A \cos \theta}{r^2} e^{-\left(\mu_1 \sec \theta + \mu_2 s \sec \theta\right)}
\]

If the number of \(\beta\)'s from the elementary ring passing through the elementary volume is called \(dN_v\), we have from the two preceding equations:

\[
dN_v = k_1 2\pi r \sin \theta \frac{r \, d\theta}{\cos \theta} \frac{\Delta A \cos \theta}{4\pi r^2} e^{-\left(\mu_1 \sec \theta + \mu_2 s \sec \theta\right)}
\]

or

\[
dN_v = k_2 \Delta A \sin \theta \, d\theta \, e^{-\left(\mu_1 \sec \theta + \mu_2 s \sec \theta\right)}
\]

Each \(\beta\) particle has a path length in the elementary volume given by \(\Delta s \sec \theta\). If \(\bar{E}\) is the average energy of the \(\beta\) rays, the average energy loss per \(\beta\) particle in the elementary volume is \(\bar{E} \mu_2 \Delta s \sec \theta\). The energy loss by \(dN_v\) \(\beta\) rays is therefore \(dN_v \bar{E} \mu_2 \Delta s \sec \theta\), and the energy loss per unit mass, called \(dR\), is given by:

\[
dR = k_3 \tan \theta \, d\theta \, e^{-\left(\mu_1 a \sec \theta + \mu_2 s \sec \theta\right)}
\]

upon substitution for \(dN_v\). (\(\bar{E}\) has now been lumped into \(k_3\).)

Now let \(\sec \theta = z\). Then \(dz = \sec \theta \tan \theta \, d\theta = z \tan \theta \, d\theta\) and

\[
dR = k_3 e^{-\left(\mu_1 a + \mu_2 s\right)} \frac{dz}{z}
\]

If \(b\) is set equal to \(\mu_1 a + \mu_2 s\), and \(dR\) is integrated to include radiation from all points on the source, one has
\[ R = \int_0^R dR = k_3 \int_1^{\sec \theta_0} e^{-bz} \frac{dz}{z} = k_3 \int_1^{Z} e^{-bz} \frac{dz}{z}, \]

where \( Z = \sec \theta_0 \). This expression can be split into 2 integrals:

\[ R = k_3 \int_1^{\infty} e^{-bz} \frac{dz}{z} - k_3 \int_1^{Z} e^{-bz} \frac{dz}{z}. \]

The second integral can be transformed by a change of variables into

\[ k_3 \int_1^{\infty} e^{-Zbz} \frac{dz}{z}, \]

so that

\[ R = k_3 \left[ \int_1^{\infty} e^{-bz} \frac{dz}{z} - \int_1^{\infty} e^{-Zbz} \frac{dz}{z} \right]. \]

These integrals are known as logarithmic or exponential integrals, and their values may be found in tables. The first term is the exponential integral of \( b \), and the second of \( Zb = b \sec \theta_0 \), represented respectively as \( -E_1(-b) \) and \( -E_1(-b \sec \theta_0) \). Therefore the dosage along the axis of the system is given finally by the expression

\[ R = k_3 \left[ -E_1(-b) + E_1(-b \sec \theta_0) \right]. \]

For sources of the dimensions used to obtain the data of Figs. 10 and 11, \( \theta_0 \) is very large and the second term is consequently negligible. Furthermore, in the same data only a single absorber is involved, making \( s = 0 \). The dosage at depth \( a \) in an absorber covering a plane source of large diameter is therefore given by

\[ R = k \left[ -E_1(-\mu a) \right]. \]
APPENDIX 3: MEASUREMENT OF EMULSION THICKNESS AND SILVER HALIDE CONTENT

As shown in several of the chapters, both the emulsions thickness (mg/cm²) and the amount of silver halide in the emulsion are significant quantities in the response of a particular film to β rays. The methods evolved to measure these properties of the films investigated in this research are described in this appendix.

A. Emulsion Thickness (mg/cm²)

The measurement of emulsion thickness in terms of mg/cm² turned out to be a rather simple problem. A sample strip of the film under investigation, of area about 20 cm², was merely weighed before and after the removal of the emulsion, and the mass of emulsion per unit area computed accordingly. Emulsion removal can be simply accomplished by soaking the film in dilute (approximately 1 N) HCl for 10 or 15 minutes. The cellulose acetate film backing is not attacked at all, and gelatin coatings over the emulsion (T coat), or under the emulsion (subbing) are sufficiently thin as to introduce very little error (perhaps 5% (S-10) ) into the measurement of the thickness of the emulsion proper.

B. Silver Halide Content of Emulsion

The finding of a method for determining the silver halide content of the emulsion proved to be a much more troublesome task. A method was desired which would be accurate, relatively convenient in routine work, and sufficiently sensitive as to permit the analysis of small areas of film (in order to make possible the study of variations in mg AgBr/cm² over the surface of a single film, if desired). The most promising methods appeared to be color-
imetric chemical tests, and this later proved correct.

Several tests relating to my problem have been described in the chemical literature. The method finally adopted grew out of a technique described by Sheppard and Ballard (S-6) for a related purpose. The final procedure was the following: A disc of film of 1 or 2 cm$^2$ area was cut from the film sheet with a cork borer or punch. Its area could be determined with good accuracy from a simple measurement of its diameter. The silver halide was then dissolved out by soaking the disc for 15 to 30 minutes in an accurately measured volume (several ml) of 30% hypo. To an aliquot (about half) of this silver halide solution were added 1 ml of 1% gelatin solution, 1 drop of concentrated NH$_4$OH, and a few tenths ml of 10% Na$_2$S. Ag$_2$S is precipitated quantitatively as a very fine brownish yellow sol in the presence of the protective colloid. The amount of silver was measured colorimetrically by comparison with standard silver solutions made up from AgNO$_3$ and precipitated in the same method. As a colorimeter we adopted the color densitometer described in Chap. 7. Any densitometer would be satisfactory, but the use of a green filter gave increased sensitivity. The results from 3 entirely independent sets of standards showed good reproducibility and the possibility of achieving accuracy within about 5% in a typical case of 1 mg Ag. The reproducibility of measurements on solutions of Ag derived from film was equally good.

However, there appeared to be a possibility that other emulsion ingredients than AgBr, not present in the standard, might affect the reaction in such a way as to make comparison with the standard unjustified. Therefore an attempt was made to determine the Ag content of film by some absolute method purely as a check on the colorimetric method. (Absolute methods appeared to be too cumbersome for routine use.) A great deal of time was wasted on this step. Recovery of the silver for gravimetric analysis
was attempted by ignition of the emulsion and by electro-
plating from hypo or cyanide solutions. For a variety of
reasons none of these methods proved trustworthy. Finally
it was decided to make a second chemical analysis, based
on different reactions from the colorimetric, and to con-
sider the colorimetric method as reliable if the two meth-
ods led to the same answer.

The second chemical analysis was based on a ti-
tration method. The AgBr (several mg) was dissolved out
of the emulsion by a KCN solution of known concentration
and volume (in excess by a factor of 2 or 3). This was
back-titrated with a standard AgNO₃ solution in the presence
of about 0.5 mg NaI. The excess KCN ties up the AgNO₃ in
a soluble complex, and the endpoint is indicated by the
appearance of a slight milkiness caused by precipitation
of AgI when the KCN is used up. The end point was not too
distinct, but agreement with the colorimetric results was
obtained to about 5% for the single film (SWR) compared by
both methods. The colorimetric method was consequently
accepted as valid.
APPENDIX 4: Sr\textsuperscript{90} - Y\textsuperscript{90} CALIBRATION SOURCES

The Sr\textsuperscript{90} - Y\textsuperscript{90} calibration sources, or radiators as they are often called in this thesis, were used to provide reproducible exposures for development testing and for calibration of individual development batches. They were prepared, in duplicate, by mixing radioactive Sr\textsuperscript{90} - Y\textsuperscript{90} solutions into plaster of Paris. The most active Sr\textsuperscript{90} - Y\textsuperscript{90} solution contained about 300 µc/ml. This was diluted in steps of 1:2.25 to 6 different concentrations. From each dilution 0.80 ml solution was mixed with 1.35 gm of fine plaster of Paris, using a pestle and mortar. After fairly vigorous grinding, the paste was poured into the mold and allowed to set.

Each mold consisted simply of a small brass plate of dimensions approximately 3" x 1" x 1/4", through which six 1/4" holes were drilled. This was laid on a flat glass plate which served as the bottom of the mold. When the plaster of Paris had set, the mold with its active sources was taken off the glass plate. A second brass plate was screwed to the top of the mold as a handle and as protection for the fingers during handling of the radiators, while the smooth source surfaces which had dried in contact with the glass were laid directly on the film during exposures.

There was some doubt as to the accuracy of the relative activities of these sources, because dilution errors were cumulative and some chance existed for redistribution of the activity during setting of the plaster of Paris. Consequently, the final evaluation of the relative activities was accomplished by counting (with the proportional counter described in Appendix 8), wherein each source was exposed to the counter in a standard geometry. It was found that the successive source activity ratios were
1:2.25 within a few percent, indicating little or no redistribution of the activity during the setting process.

A word might be put in at this point regarding this plaster of Paris method as a general technique for source preparation, a problem which always presents more difficulties than one foresees. The method, of course, is useful only for thick sources. A major difficulty, as already suggested, is that only a fraction of the water used in preparation of the paste (namely 18% of the dry powder weight) remains in the set plaster of Paris, while the rest evaporates from the surface. During this redistribution of the water, a redistribution of the dissolved activity can take place. Whether or not this actually happens depends on the chemical nature of the activity. The effect appears to be small when the active ion has a chemical affinity toward one of the ions of the plaster of Paris molecule (CaSO$_4$·2H$_2$O). Fairly successful sources have to date been made with Ca$^{45}$Cl$_2$, Sr$^{90}$Cl$_2$, H$_2$F$^{32}$O$_4$, and H$_2$S$^{35}$O$_4$. On the other hand, Co$^{60}$Cl$_2$ was conspicuously unsuccessful, a great deal of it concentrating on the surface of the source. Another difficulty is that the granular nature of the powder does not permit really adequate uniformity of distribution for such low energy isotopes as S$^{35}$. Within these limits, however, this method appears to give a reasonably simple and reliable source having a known activity per gram.
APPENDIX 5: STANDARDIZED DEVELOPMENT PROCEDURE

The initial film development procedure tested was based on a method described in the literature (P-3). In this method the development tank consisted of a small Dewar flask fitted with a stopper. To the stopper a glass strip was attached which reached almost to the bottom of the Dewar. The film strip (approximately 1" x 3") was held on the glass plate by rubber bands. When the Dewar was about half filled with developer at 20°C, and the film strip and stopper were in place, the tank was held in a horizontal position and agitated rhythmically both by rocking it up and down to an angle of 45° with the horizontal and by rotating it about a horizontal axis. This procedure of course gives good agitation, but we found that the temperature rose during development by about 0.5 to 1.5°C. As this fluctuated in an unpredictable manner and seemed somewhat serious in view of the strong dependence of development rate on temperature, a modification of the method was evolved.

In this modification, which was the procedure used throughout almost all the experiments of this research, the developer was contained in a full 4 oz. bottle immersed to its neck in a bath of water at 20 ± 0.5°C. The film strips were again attached to small glass plates suspended from a rubber stopper. In this case the oversize stopper merely rested loosely on the lip of the bottle so that it could be rotated back and forth relative to the bottle, thus agitating the film in the developer. Agitation was standardized at about 30 to 60 cycles per minute, each cycle consisting of a rotation of the stopper through about 45° and back. This agitation is not as complete as in the first procedure, but no variations in development rate could be detected between the top and bottom of the bottle. With 2 glass plates attached to the stopper, and a film strip on each side of each plate, 4 strips could be developed simul-
taneously. When double coated films were used, requiring development on both sides, bent glass rods were substituted for the glass plates, and provided for the simultaneous development of 2 strips. The apparatus employed in this development procedure is shown in Fig. 50. In actual use, the bottles were set into a tank of water at 20°. Neither the tray nor the particular hand shown were standard features.

Tests indicated that exhaustion of developer was negligible in a single run, so that 4 strips could be developed in one bottleful of developer without fear that degree of development would depend on area of exposure.
APPENDIX 6: MAGNET AND ASSOCIATED EQUIPMENT

The electromagnet used in this research was borrowed in operating condition from Prof. S. C. Brown. Essentially no development work was required on it. The magnet core was chromium plated Armco steel. The pole pieces, which measured in cross section 10.8 x 14.9 cm, were mounted in such a way that the gap could be set at will. In this investigation the gap was constant at 5.1 cm. The field intensity distribution in the gap was systematically plotted out with the search coil described in the next paragraph. The uniformity was found to be sufficiently good over a central area such that at no point on the electron path within the spectrograph was the field intensity more than 2% lower than the maximum.

The current for the magnet was supplied by a regulator consisting of four 6AS7 triodes in parallel acting as a current gate. The voltage drop across a constant resistance in the current circuit was compared with a reference voltage from a VR tube circuit, the difference being amplified and fed back negatively to the current gate. At currents greater than the rated value for the triodes, a bypass shunt was thrown in, while at very low currents a higher constant resistance in the current circuit was used to give improved regulation. Regulation was such that a 15% drop in line voltage produced less than 1/2% change in current.

The magnetic field strength was measured by a continuously operating search coil nicknamed the "gaussometer". This device consisted of a 3000 to 4000 turn coil of #36 insulated wire, wound on a plastic spindle of dimensions approximately 1" x 1/2". This was inserted in the end of a plastic rod projecting out 5" from the shaft of a 30 cps synchronous motor. The alternating voltage generated
in the coil as it was rotated in the magnetic field was picked off slip rings and measured by a Ballantine vacuum tube voltmeter. The design of this voltmeter is such that the meter reads a rectified voltage. Consequently, even harmonics of the signal, if small relative to the signal, add no net contribution to the rectified signal voltage and hence are undetected. In the presence of 60 cycle pickup this has a definite advantage. This voltmeter was checked periodically against standards in the EE Department, and was found to be stable within 1 or 2%. The gaussometer was initially calibrated in Prof. Bitter's laboratory with a magnetic field whose strength was known accurately by nuclear resonance experiments. It was found to generate 7.19 mv per gauss, within an accuracy of about 1%.

It was desired to provide for a continuous monitoring of the field, but with the spectrometer in place the gaussometer probe could not be positioned between the pole faces. Consequently the gaussometer was mounted on a stable platform just above the pole faces, and the ratio of field strength in the 2 positions determined. This ratio was found to be the same for all field strengths.

Once the spectrograph spectrum had been measured with the proportional counter, it became possible to check the overall gaussometer calibration at frequent intervals. The counting rate was simply checked at a field strength where the counting rate was very sensitive to the field strength. The reproducibility of the counter itself was tested by counting at a field intensity where the counting rate was a very insensitive function of the field. This system worked very efficiently, and was useful at a time near the end of the experiments when a short circuit apparently developed in the gaussometer coil, thereby changing its calibration.
APPENDIX 7: BETA RAY SPECTROGRAPH

A. Design of Instrument

The basic requirements to be filled by a spectrograph designed for the calibration of photographic film are high intensity and, as is always important, convenience. Resolution, usually of prime concern, is only a secondary consideration in this application. The effect of scattering, which must be completely eliminated in most applications, here depends on the location where it takes place. Back-scattering from the source mount is beneficial in that it increases intensity without decreasing resolution. Scattering occurring further on in the slit system is detrimental because it does decrease resolution.

A wide variety of β ray spectrographs can be found in the literature, each designed with certain specific purposes in mind. At least 2 basically different types are now in common use. The older type is the 180° spectrograph, in which the β particles travel through a circular path of approximately 180° between the source and exit window. A more recent variety is the lens type spectrograph in which the magnetic field focuses the electrons in the same type of geometry as a lens focuses light. The second has a higher transmission factor than the first, which would be a definite advantage in my problem, but is much more complicated to design and build. Because of the simplicity of the 180° spectrograph and the availability of a satisfactory electromagnet, this basic type was the obvious choice for my investigation. Its low transmission was surmounted by using a strong source.

The basic features of the instrument, as finally constructed, are shown in Fig. 51. The main body consisted of a single brass casting. The central chamber, lined with
Figure 51  BETA RAY SPECTROMETER
aquadagged lucite to reduce scattering, was made accessible by a removable bottom plate. Access to the exit window was made possible by a removable top plate. Film, held to the bottom surface of the film holder, was positioned over the exit window by means of a rod extending to the outside of the spectrograph through an end plate fitted with an "O" ring vacuum seal. This end plate could be unscrewed to permit complete withdrawal of the film holder for loading.

Three different film holders were used. The first was designed for exposures with perpendicularly incident β particles. It was a 4" x 1" x 1/4" bakelite slab with a rubber pad on the bottom to grip the film and slide it over the exit window. The second holder was designed for exposures at 3 different angles of incidence: 0°, 17.5°, and 35°. It was a lucite piece initially of the same dimensions as the first holder, but with the edges milled down to give exposures at the desired angles. Film strips about 3" x 1/4" were beeswaxed to the 2 milled surfaces and to the unmilled central section, being thus held at the desired angles throughout exposure. With this arrangement, the exposures at the 3 angles were simultaneous, so that despite errors in timing or in magnetic field setting, the 3 exposures would be identical. The third holder was identical with the second, except that it permitted exposures at the 3 angles of 0°, 52.5°, and 70°.

Several different exit windows were used during film exposures. At energies less than 0.35 Mev, brass discs with either a circular aperture of 1/8" diameter or a slot of 1/8" width were employed. At higher energies, windows similar except having 1/4" diameter or width holes were used because of the disturbing effect of scattering from the edges of smaller holes at these energies. Tests were made with apertures up to 1/2" diameter to prove that the windows actually used were free from scattering effects.
When counting was to be done, the film holder and top plate were removed and the proportional counter was mounted at the position of the exit window.

The source was mounted on a removable plug. An aluminum gate could be swung in front of the source to permit accurately timed starting and stopping of the exposures. It, also, was operated by means of a rod extending through an "O" ring vacuum seal. The source consisted of approximately 17 mc of Sr$^{90}$ and 17 mc of its daughter Y$^{90}$, mounted on a thin strip of Pt to give high backscatter and a consequent increase in intensity. This combination of isotopes was almost ideal for my purposes. The 20 year half life, high energy, high specific activity, and absence of γ rays all were desirable properties. Furthermore, the presence of 2 spectra gave a more uniform beam intensity over a wide range of energies than would a single spectrum.

The mounting of the source was in principle a rather simple undertaking, but the high radiation level and the great hazard of Sr$^{90}$ if ingested introduced many complications. After numerous trial runs, the following procedure was carried out. The activity came from Oak Ridge in an 11.3 ml solution of SrCl$_2$ containing about 1 mg/mc total solids, with about 0.25 mg/mc non-volatile matter. The solution was evaporated to dryness on a steam bath to get rid of the volatile solids. After redissolving in dilute HCl, a small amount of Y(NO$_3$)$_3$ was added as carrier, and the Y$^{90}$ was precipitated out as Y(OH)$_3$ by the addition of NaOH to the phenolphthalein end point. This step reduced the radiation intensity during the remaining manipulation. The Sr$^{90}$ was then precipitated as SrCO$_3$ by the addition of Na$_2$CO$_3$. (An accident occurred at this point losing about 1/5 the original activity.) This precipitate was taken up as a slurry in a fraction of a ml of
acetone, containing a few mg of dissolved parlodion to act as a binder, and transferred with a dropper to the Pt strip. After the evaporation of the acetone, 2 more layers of parlodion, dissolved in acetone, were painted over the source surface as further insurance against flaking. Each layer was approximately 1 to 2 mg/cm² thick. The source appeared to be fairly permanently mounted, but 3 months later approximately 10% fell off. At this time another layer of parlodion was painted on the surface and a thin gold foil was cemented over the source. The final source as used throughout most of the experiment consisted of about 30 to 35 mc of Sr⁹⁰ and Y⁹⁰, in a strip of dimensions about 1.50 x 0.30 cm. The source, roughly 25 mg/cm² thick itself, was covered by about 2 mg/cm² of gold and backed by a 200 mg/cm² Pt strip.

The resolution and intensity of the electron beam incident on the film are established by the spectrograph slit system and source dimensions. The designing of these features is in some ways similar to that of an optical lens system. So many compromises must be made among the various desired properties, and the general solution is so complicated, that in practice one makes an educated guess as to design and determines whether or not the properties of that particular design meet the requirements. A large number of 180° spectrometers can be found in the literature. The design of mine is based primarily on that of Lawson and Tyler (L-5). Since my plans were completed, 2 very useful papers have appeared in the literature which would have given considerably more education to my guesses (0-1, 0-2).

The chief compromises to be met in my design were (1) large slits for high intensity vs. small slits for high resolution, (2) large source area (minimizing self absorption) for high intensity vs. small source area for high resolution, (3) large source length parallel to the magnetic
field for high resolution at a given source area vs. small length to permit narrow magnet pole gap and therefore good uniformity of field in the gap, and (4) small dimensions for high intensity (electrons per cm$^2$ film) vs. large dimensions to permit convenient positioning of the film or counter at the exit window. The final choice of parameters was: mean radius of curvature = 5.0 cm, source width = 0.30 cm, source length = 1.5 cm, defining slit width = 1.75 cm.

B. Resolution Calculations

The calculation of the resolution is a rather tedious geometrical problem. Therefore, many of the steps in the calculation are omitted, although the method of approach and results are shown.

The essential features of the system are shown in Fig. 52, in which the spectrograph has in effect been turned upside down relative to its operating position as shown in Fig. 51. All quantities shown are measured in the plane of the paper (i.e., perpendicular to the magnetic field).
The symbols in Fig. 52 have the following meanings:

\( w \) = source width = 0.30 cm
\( y \) = distance from center of source to point of emission of \( \beta \) particle
\( x \) = distance from center of source to point of impact of \( \beta \) particle on film
\( \rho \) = radius of curvature of \( \beta \) particle
\( \rho_m \) = mean radius of curvature of \( \beta \) particle = 5.00 cm
\( a \) = defining slit width = 1.75 cm
\( \Theta \) = angle of emission of \( \beta \) particle in plane of paper

Certain quantities which are necessary also, but are in the third dimension not shown, are:

\( s \) = distance in plane of film measured perpendicular to \( x \) direction
\( \phi \) = angle between direction of emission of \( \beta \) particle and plane of paper
\( A \) = area on film surface

The procedure is to express the fraction of the \( \beta \) particles of radius \( \rho \) emitted from a particular point on the source and incident at a particular point on the film. This expression is then integrated over the source to take account of all \( \beta \) particles of radius \( \rho \), and then integrated over the exit window, or film area, of interest. Limits on these integrations are determined by the source, exit window, and defining slit dimensions.

It can be shown that for the source dimensions of this spectrograph a very negligible error is committed by considering the source length as 0. (The significance of this fact is that better resolution could be obtained for the same source area if it were possible to increase the source length and decrease its width.) Therefore in the following calculations the source consists of a line in
the plane of the paper as drawn in Fig. 52.

The fraction of all the $\beta$ particles having radius $\rho$, which are emitted in the source element $dy$ in a solid angle increment $d\theta d\phi$ ($\theta = \text{approximately } \pi/2, \phi \text{ small}$) is

$$\frac{dy}{4\pi \rho} \ d\phi \ d\theta.$$

These particles traverse a path in the magnetic field and are incident on an area of film $dx ds$. When substitutions are made for $\theta$ and $\phi$ in terms of the linear coordinates of the system, one can express this same quantity, which is now the fraction of all $\beta$'s of radius $\rho$ emitted from $dy$ and passing through $dx ds$, as

$$f_{\rho}(x,s,y) dx ds dy = f_{\rho}(x,y) dAdy = \frac{1}{4\pi^2 \rho^2} \cdot \frac{dAdy}{\pm \sqrt{4\rho^2 - (x - y)^2}}.$$

The variation with $s$ can be shown to be negligible, by the same argument which permits one to ignore the source length. Hence the two expressions on the right are independent of $s$. Certain relatively unimportant approximations are involved in the constant term. The ($\pm$) is indicative of the fact that $\cos \theta$ can be positive or negative, corresponding physically to the fact that there are 2 values of $\theta$ (when $\theta \neq \pi/2$) for which the particles are incident on the same element of film. This expression is integrated over the source, $y$, in 2 parts, given by the ($\pm$) term. One of the limits on $y$ for the first part is set by the upper edge of the defining slit, while one for the second part is set by the lower edge of the slit. The other limit for each is the minimum value of $y$, set by $\theta = \pi/2$ or by the left edge of the source. It can be shown that when the integration over $y$ is performed, one has:
\[ f_\rho(x) = \frac{1}{4\pi^2 \rho} \left\{ \sin^{-1}\left(\frac{y_1 - x}{2\rho}\right) + \sin^{-1}\left(\frac{y_2 - x}{2\rho}\right) - 2\sin^{-1}\left(\frac{y_3 - x}{2\rho}\right) \right\} \]

\[ f_\rho(x) \] is the fraction of all \( \beta \)'s of radius \( \rho \) which are incident per unit area at \( x \)

\[ y_1 = x - 2\sqrt{\rho^2 - (a + \rho - \rho)^2} \] or 0.15, whichever is smaller

\[ y_2 = x - 2\sqrt{\rho^2 - (a - \rho + \rho)^2} \] or 0.15, whichever is smaller

\[ y_3 = x - 2\rho \] or -0.15, whichever is larger

For a particular value of \( x \) it is possible to plot out \( f_\rho(x) \) as a function of \( \rho \). This has been done, and in Fig. 53 \( f_\rho(x) \) is shown for \( x = 9.85, 10.0, \) and 10.15. The distribution is closely the same for all 3 values of \( x \), except for a displacement in \( \rho \).

The film area densitometered was later taken as a circle of 0.16 cm diameter. This was effectively the exit window. If it is desired to know the total fraction of \( \beta \)'s of radius \( \rho \) which pass through this exit window, it can be determined in the following way. This quantity is equal to the window area multiplied by the fraction of \( \beta \)'s of radius \( \rho \) incident per cm\(^2\), averaged over the exit window area \( A_w \), i.e., it is equal to \( A_w \bar{f}_\rho(x) \). Because of the similarity between the shape of \( f_\rho(x) \) and \( f_\rho(x + \Delta x) \), except for the displacement in \( \rho \), one can consider with sufficient accuracy that \( f_\rho(10.00) = f_\rho - \frac{0.08}{2} (10.00) \).

Therefore \( \bar{f}_\rho(x) \) for a 0.16 cm diameter exit window is given by the ordinate of \( f_\rho(10.00) \) averaged over \( \rho \) between \( \rho - 0.04 \) and \( \rho + 0.04 \) (i.e., 2\( \rho \), not \( \rho \), has a spread of 0.16 cm). In Fig. 54 is shown as a function of \( \rho \) the fraction \( F(\rho) \) of the \( \beta \) particles of radius \( \rho \) which pass through an exit window of diameter 0.16 cm. It can be seen that half the \( \beta \) particles incident on this film area are within a \( \rho \) or
momentum spread of about 2%, or an energy spread of about 2 to 4%. The maximum energy spread is 6% at high energies and 12% at low energies.
APPENDIX 8: MEASUREMENT OF SPECTROGRAPH BETA RAY FLUX

A. Choice of measuring instrument

In order to know, in absolute units, the exposure to which film was subjected in the spectrograph, it was essential to measure in some way the β ray flux at the position of the film. A choice was available between at least 2 different types of measurement: it would conceivably be possible to measure either radiation dosage, by means of a thin ionization chamber substituted into the position of the film, or number of β particles/cm², by means of some type of counter. Whichever measurement was made directly, the other quantity could be determined indirectly from the known rate of loss of energy of β particles at all energies of interest. Of course, the directly measured quantity would be expected to be most accurately known.

The emphasis of this thesis was to be on the measurement of radiation dosage. In principle, therefore, a preference should have been given to the ionization chamber as the spectrum calibrating instrument. However, the data sought in this thesis were expected to be of general usefulness in all applications of photographic detection of β rays, including problems where the exposure quantity of primary interest would be number of β particles/cm² rather than radiation dosage. Consequently, the choice of instrument was based chiefly on convenience, rather than the nature of the β ray flux property actually measured.

Consideration was given to these instruments: ionization chamber, Faraday cage, scintillation counter, proportional counter. The final decision was in favor of the proportional counter. In an analysis of these instruments, the other detectors were excluded for the following
reasons: The ionization chamber I believed to be too insensitive to measure, reliably and conveniently, small currents of about $10^{-14}$ to $10^{-15}$ amps; this was perhaps poor judgement. Furthermore, I expected that any window covering a sufficiently large aperture would distort the measurements at the lowest energy of about 0.03 MeV. The Faraday cage was expected to be insufficiently sensitive, and probably somewhat unreliable in view of secondary electron emission problems. These disadvantages seemed to outweigh the advantage of no window absorption problems. The scintillation counter was expected to be less than 100% efficient at energies of less than 50 keV. The chief difficulty anticipated with the proportional counter was window absorption at low energies, although in practice this did not turn out to be serious. Despite the fact that the proportional counter proved very successful, I believe, in retrospect, that a more detailed analysis should have been made of the feasibility of an ionization chamber.

B. Design of Proportional Counters

The first proportional counter designed was a small end window instrument of approximately 1/2" internal diameter and 1.5" length. In order that the bead at the end of the center wire should not intercept any β particles before they reached the sensitive volume, thereby reducing efficiency below 100%, various looped and off-axis center wires were tested. Probably as a result of this asymmetrical geometry, a good plateau in the counting rate vs. voltage curve was never attained. It soon became apparent that a side window design would offer several advantages over the end window type, and the latter was abandoned in favor of the former.

The design of this side window proportional counter, which was used in all spectrum calibrations underlying the
film measurements of this thesis, is shown at about full scale in Fig. 55. The cathode was a polished brass cylinder with aquadag coated surface. The 3 mil tungsten center wire, attached at each end to a larger nickel wire, was inserted and mounted through small tubes in the glass seals. This arrangement made possible replacement of the center wire without readjustment of these seals. The window consisted of several layers of parlodion film. These were prepared in a manner similar to that described by Blizard (B-12), wherein a drop of an amyl acetate solution of parlodion was allowed to spread on a clean water surface, and after drying, was picked up as a 10 - 20 μg/cm² film on a wire loop. (The thickness was readily determined by means of the interference colors.) Several such films could be superimposed for greater strength. This window was held between two small "washers," whose holes formed the counter window defining aperture of accurately measured area. These washers were beeswaxed into the window mount, which was in turn beeswaxed to the body of the counter. The gasket around the rim of the window mount rested in a groove around the exit window of the spectrograph so as to form a vacuum tight seal and to position the counter window in the same location as the film during its exposure. The counter was connected to the same vacuum system as the spectrograph. By manipulation of appropriate stopcocks, both instruments could be evacuated simultaneously so as never to put full atmospheric pressure across the thin counter window. After readjustment of the stopcocks, the counter could be filled to the desired pressure with the counting gas, while the spectrograph vacuum was still maintained.

G. Electronic Apparatus

The electronic equipment associated with the counter consisted of a battery high voltage supply, a model 204B Atomic Instrument Co. linear amplifier together
Figure 55

PROPORTIONAL COUNTER
with a preamplifier, and a scaler.

The voltage supply contained five 300 volt and twenty 67.5 volt B batteries. By means of 2 switches and a rheostat, the voltage could be continuously varied between 0 and 2850 volts. 10 megohms were distributed throughout the circuit to reduce the high voltage danger and to prevent the drawing of heavy currents during inadvertent shorting of the batteries. A one μf condenser was placed across the output to filter out noise.

The linear amplifier amplified the counter pulse, discriminated the pulse height, and shaped the output pulse for driving the scaler. It was operated at maximum gain and 0.2 μs rise time, without any changes of any sort being required in the circuit. The preamplifier was fitted with a non-linear load impedance in the form of a 1N34 crystal, which has the property that its resistance decreases as the voltage across it increases. The introduction of this modification reduced the range of pulse heights, and thereby improved the counter plateau characteristics through decreasing overloading of the linear amplifier by the larger pulses.

The scaling unit consisted of an electronic scale of 4096 preceding a mechanical register. This mechanical register was arranged to stop a clock at the end of collection of a predetermined number of counts. It was found that the resolving time of the whole counting apparatus was limited by the pulse inverter and first scaling stage at the input of the scaler. Modifications in these elements permitted a reduction in the overall resolving time.

D. Operation of Counter

Several filling gases were tested in this counter. Among these were an argon (90%) and methane (10%) mixture,
PROPORTIONAL COUNTER PLATEAUS (Discriminator = 30v.)
\[
\frac{\beta' \text{s}}{\text{cm}^2 \text{ min.}} \quad \text{vs Anode Voltage}
\]
pure methane, ethyl ether, isobutane, and n-butane. Considerably better results were obtained with n-butane than with any of the others. Two of the apparent reasons for its good qualities are its good quenching properties (common to most polyatomic organic gases) and its high electron stopping power, which produces a large number of primary ionizations for each entering β particle.

In a large number of tests, the operating conditions of gas pressure, voltage, and discriminator setting were systematically varied. Final conditions were standardized at 20 cm pressure, 2450 volts counter voltage, and 30 volts discriminator setting. In Fig. 56 are shown a fraction of the plateau curves taken at 3 β particle energies under these conditions. All other combinations of operating conditions which might reasonably have been chosen would have given the same counting rate within a few percent. Synchroscope observations indicated that no multiples occurred, within the limit of error of 1 or 2%. This conclusion was of course supported by the excellent plateau characteristics. Reproducibility, also, was excellent: even with several different center wires, and different gas fillings each day, the absolute reproducibility of measurements made in the spectrometer, under these prescribed counting conditions, was better than 2% over a period of a year. The resolving time, as measured by the 2 source method, was 4μs. Resolving time corrections to counting rates never amounted to more than a few percent. On the basis of all these very good operating characteristics, I believe that one may place considerable confidence in the absolute β ray flux measurements made with this instrument. Its reliable behavior was one of the most satisfying aspects of the entire experimental work reported in this thesis.
E. Calibration of Counter

As a result of my adviser's scientific thoroughness and my own scientific naivete, I made an attempt to check the absoluteness of the counter operation by independent tests. These proved to be the least satisfying aspect of the entire experimental work reported in this thesis.

The first type of test attempted was to mount the counter at a known distance from a known source of β rays, and to compare the measured with the expected counting rates. Many measurements were made, with various modifications of source and geometry.

Sources were prepared from 2 different isotopes: \( \text{Y}^{91} \) (1.5 Mev) and \( \text{Y}^{90} \) (2.3 Mev). The isotopes were deposited on thin parlodion films (about 20 \( \mu \)g/cm\(^2\)) by evaporation from the film of a small drop of a high specific activity water solution of the isotope. The film was then made conducting either by the superimposition of a 0.15 mg/cm\(^2\) aluminum foil or the evaporation of a very thin layer of aluminum onto the film from a hot filament. The absolute activity of the sources was measured in a 4\(\pi\) solid angle (i.e., 100% geometrical efficiency) proportional counter generously loaned by Dr. Randall S. Caswell. This counter was considered accurate within a few percent, on the basis of Caswell's calibration with Co\(^{60}\) standards coincidence counted both at the N.B.S. and at M.I.T., and with Na\(^{24}\) standards coincidence counted at M.I.T. A complete description of the instrument may be found in Caswell's thesis. One modification of this source preparation technique was to 4\(\pi\) count the activity of a diluted aliquot of the final source, rather than to calibrate the source itself. This removed resolving time corrections of 10 to 12%, but ran the risk of pipetting errors which seemed more serious. As finally prepared, these calibrated sources seemed to meet the essential requirements of absolutely known activity
and spatially isotropic radiation pattern.

The mounting arrangement of the source and my side window proportional counter during calibration was the following: The counter was clamped against a hole in one end of a 4" diameter, 9" long, hollow brass cylinder. This cylinder was evacuated during measurements to prevent air absorption of the β rays. Its walls were lined with lucite to reduce scattering. The source could be mounted in one of two positions. The position most frequently used was situated about 7 cm (distance known accurately within 1 or 2%) from the counter window. A brass baffle was located half way between source and counter, to further reduce β ray scattering effects. The other position was at a distance of 18 cm from the counter window, and about 6 cm behind a second baffle of lucite. Rough calculations of the increment in counting rate expected to result from scattering at the baffle edges and walls indicated that these effects would be only a few percent at most, for the first position, and a negligible percentage in the second position. Comparative measurements between the 2 positions showed that scattering in the first position was in fact small for Y\textsuperscript{91}, but apparently amounted to as much as 15% in the case of Y\textsuperscript{90}. This was one of the several rather strange phenomena observed in this set of tests.

The final results of these tests were that my counter appeared to be rather consistently counting high by roughly 15% in the case of Y\textsuperscript{91}, and low by about the same amount in the smaller number of measurements with Y\textsuperscript{90}. There is not space here to reproduce all the arguments for and against possible explanations of these discrepancies. Suffice it to say that the one feeling with which I was left was simply that the data could not have been obtained. Due to limitations in time, this method of calibration was abandoned in favor of a second simpler method.
The second method which I used in testing the absoluteness of my counter was to compare counting rates obtained with it against counting rates obtained under identical geometry with other counters. The same mounting arrangement for the source and proportional counter was used as before, although it was in this case not necessary to prevent scattering from the walls and baffles. As reference counters with which to compare my proportional counter, I used 3 Tracerlab thin end window GM counters. The plateau of one was measured and found to be very flat, indicating reliable behavior. All three gave essentially identical results. When these counters were used, the window assembly of my proportional counter was left in position over the hole on the end of the brass cylinder. The GM counters were then positioned directly behind the window assembly, so that the beam defining aperture was still the window of the proportional counter. Hence the counting geometry remained identical under all conditions. The activity as measured by the GM counters was slightly dependent on their position. When directly centered over the beam defining aperture, their counting rates were 6 to 8% lower than when mounted about 0.5 cm off center. This effect was attributed to the interception of some of the collimated \( \beta \) particles by the center wire bead. The off-axis reading was considered the better value. The degree of window absorption by the GM counter was estimated from the effect of extra absorbers interposed in front of the window.

Tests were conducted using 2 isotopes, \( \text{Y}^{91} (1.5 \text{ MeV}) \) and \( \text{Tl}^{204} (0.78 \text{ meV}) \). The final results indicated that if the GM counters were 100% efficient the proportional counter was counting 7% too high for both isotopes. Even if this discrepancy points up an actual inaccuracy in my counter, this inaccuracy is not very serious. Furthermore, since the only conceivable cause of an excessively high counting rate is the existence of multiples, which could be rather
conclusively ruled out by synchroscope observations, at least a part of this indicated inaccuracy is probably not real. No correction for this apparent 7% error was made in the spectrograph β ray flux measurements.

F. Beta Ray Flux Determination

The determination, with the proportional counter, of the absolute number of β's/cm² which would be incident on the film at all energies between 0.03 and 1.8 Mev, was attended by a few small difficulties not yet discussed. At the lowest energies, window absorption had to be investigated. This was accomplished by 2 sets of measurements with window thicknesses of about 40 and 60 μg/cm², respectively; these gave identical results and indicated that absorption was low or non existent. At all energies it was necessary to know the effective window area. The geometrical area of 0.0785 cm² was accurately known. However, this was not the effective area for 2 reasons. First, the β particles were not actually all incident perpendicularly, but rather at all angles up to about 10° with the normal. Consequently, the finite thickness of the window defining "washers" cast "shadows" which slightly reduced the effective window area. Second, at high energies the range of the β particles in the brass washers was appreciable compared to the window dimensions, so that some were able to penetrate the washer edge and enter the counter when incident outside the geometrical window area. These effects were extensively investigated through the use of washers giving windows of several different sizes. At high energies the correction to the geometrical window area rose to about 8%, but was probably accurate within about 2 or 3%. The background counting rate depended slightly on energy, but was a very small fraction of the total counting rate and could be measured with adequate accuracy. Resolving time corrections were never more than a few percent, as
previously stated. The final corrected β ray flux, as a function of energy, is shown in absolute units in Fig. 57. Allowing for all sources of error, it is probably accurate within about 5 to 10% over the whole energy range.
Figure 57

BETA-RAY SPECTRUM IN SPECTOGRAPH
(July 1950)

ELECTRONS/cm² min. vs ELECTRON ENERGY
APPENDIX 9: THIN SOURCE PREPARATION

As has previously been emphasized, the preparation of standard sources of various sorts is a key and sometimes tricky problem in the quantitative detection of \( \beta \) rays with photographic film. During the course of this thesis, a considerable amount of effort went into the preparation of large area, thin, uniform sources. In the expectation that others may be interested in some of the failures and successes, I will describe here the important features of my work.

As stated in Chap. 7, I wished to prepare sources of several isotopes which would be quite uniform (within 5 or 10\%, if possible) over a central circle of 1" diameter, and uniform within a somewhat greater tolerance out to a diameter of about 2 3/4". These sources were preferably to be sufficiently thin that their self absorption and self focusing properties would have little effect on the \( \beta \) radiation from the source. However, it did not seem necessary to insist that the sources be a great deal thinner than the photographic emulsion to which they were to be exposed. Therefore a thickness of about 0.5 to 1 mg/cm\(^2\) seemed tolerable, even for the lowest energy isotopes, although the thinner the better.

A. Previous Developments

Many other workers have been faced with source preparation problems, and have solved them sufficiently well to meet their needs. Most developments, however, have not provided for both the large area and the thinness which I wished to achieve, or else were applicable only to special elements. Among previous developments considered were the following:

(1) Evaporation of a compound containing the iso-
tope, or the element itself, from a hot filament onto the source mount (in my case a 3" diameter lucite disc). This method is used primarily when the ultimate in source thinness is required. It is capable also of yielding reasonable uniformity if the source mount is placed sufficiently far from the hot filament. This method was not used for several reasons, the chief one being that several of the isotopes which I wished to use were in a chemical form difficult to evaporate, or were of such low specific activity that a large amount of solid would have had to be evaporated.

(2) Spraying the activity, in the form of a solution, onto the source mount. Loevinger (L-7) used this method in preparing $^{32}$P sources, using a penicillin nebulizer as the spray gun. This method, which did not come to my attention until after most of my sources were prepared, has rather general possibilities. However, uniformity is not too easy to achieve, and the activity is likely to get spread around the working area.

(3) Deposition of the activity by evaporating the water from a small pool of an active solution on the source mount. If the source mount is flat and level, the initial pool of solution will be of uniform depth over the mount, and hence initially the activity per unit area is uniform. If there were no redistribution of activity during the evaporation of the water from the solution, the final dried source, also, would be uniform. The method here described is quite useless, however, because drastic redistribution does occur during drying, with the formation of small clumps of activity. Even with gelatin or other protective colloids present to limit the growth of clumps (L-4), uniformity is still entirely inadequate.

(4) Precipitation of the activity as a colloid, followed by evaporation of the water from a pool of the colloidal suspension. The precipitation tends to prevent
redistribution of activity during evaporation of the water. A very fine precipitate is essential in order that the individual particles be less than $1 \text{ mg/cm}^2$ in diameter for self absorption reasons, and in order that the precipitate remain uniformly dispersed during deposition on the source mount. After attempts at a few modifications of this method, it was finally adopted as described.

B. Exploratory Experiments

In these exploratory experiments my aim was to find a method which would be applicable to as wide a range of elements and compounds as possible, in order that it be useful for many different isotopes. My first attempts led up several blind alleys, for not only was I unfamiliar with many of the chemical reactions, but also I was ignorant of the fact that, as one chemist later told me, work with colloids is not chemistry; it is magic.

The first method which I investigated was the precipitation of the activity in the presence of a colloid, such as carbon black or bentonite (a very fine colloidal mud). Most colloids consist of charged particles, usually negative (like the two just mentioned). In fact it is to the electrostatic repulsion between particles that these colloids owe their stability. The hope was that the charged colloidal particles would serve as nuclei for the growth of a large number of crystals, which would consequently be of small size. This would have given a very general method of thin source preparation. However, the technique does not work; in general the electrolytes involved in the reaction tend to flocculate the colloid, due to their neutralization of the particle charges. (An interesting example of this phenomenon in nature is the flocculation of colloidal mud in river water by the salts of sea water to form river deltas.)
The second method I attempted was the direct adsorption, without precipitation, of the activity onto a colloid. This method offers definite possibilities, if the specific activity of the isotope is sufficiently high. In this case, the total amount of electrolyte added to the colloid may be sufficiently small that flocculation is not produced, and a very fine dispersion of the activity is achieved which does not redistribute itself greatly when the water is evaporated from the colloid. The flocculating effect of an ion increases very rapidly with increasing valence of the ion. This method was used in the preparation of the W\textsuperscript{185} source, as described later, but not under favorable conditions. Better methods were found for the other isotopes, which were in anionic form ($S_{35}^{2-}$, $P_{30}^{2-}$) or too low specific activity ($Y_{91}$, $Tl_{204}$, RaD) for best results.

C. Final Method

The remainder of the isotopes were prepared by method (4) just as stated above. Although trial and error formed a basic part of the experimentation, certain principles were useful guides and are discussed briefly here.

In general, particle sizes of about 1 $\mu$ diameter are most satisfactory. Particles that are much larger settle too rapidly, while true colloids are often too unstable and flocculate or redissolve during the evaporation process when pH and other solution properties are changing. The magic lay in the production of particles having the right size, for the results obtained are to some extent affected by such subtle variables as quantity of excess of reagents, order of mixing reagents, etc. Usually the difficulty lies in obtaining particles sufficiently small. Fine precipitates are favored by conditions of low solubility, i.e., an inactive ion which reacts with the active ion to give a compound of low solubility, low temperature, excess of inactive reagent, small solution volumes. It is also advantageous to carry out the precipitation in the presence of
a small amount of a so called protective colloid, such as gelatin, gum arabic, albumin, casein, or agar. This protective colloid serves the double purpose of preventing flocculation and particle growth, and of acting as a binder for the precipitate after drying.

My procedure in developing the recipes given below was to select from the Handbook of Physics and Chemistry three or four of the reagents giving the least soluble compound with the active ion or radical. In inactive "dry runs" I then systematically tried each reagent in conjunction with each of the above protective colloids, and examined the promising precipitates under a microscope to determine the particle sizes. These precipitations were sometimes repeated under different temperature conditions, and the best recipe was selected from the results of these tests carried all the way through to the dried deposit of precipitate. The actual deposition of the precipitate on the lucite discs was carried out in the following way:

Each disc was 2 7/8" in diameter, 1/4" thick, with the thickness very uniform over the whole disc. The surface of the discs was cleaned in most cases with a wetting agent, and dried. A low rim was attached to the disc by beeswaxing a strip of Al foil around the edge. This source mount was then floated on a pool of mercury, giving a level upper surface because of the uniformity of the disc thickness. Immediately after precipitation of the active isotope, the suspension was stirred thoroughly to give homogeneity, and poured onto the disc to dry undisturbed in a CaCl₂ desiccator. After drying, the surfaces of these sources were made conducting by the evaporation onto them of a thin layer of Al. Conductivity was required in the extrapolation chamber measurements on the sources.

In certain cases slight modifications of the above procedure were made. The Tl source was dried under a heat lamp instead of in a desiccator. The surface of the disc
for the F\textsuperscript{32} source was lightly aquadagged, and remained conducting even after the active deposit was dried. Hence no aluminum evaporation was required. This modification might well have been used on all the sources.

Several of the sources finally prepared were not as uniform as was originally desired. To circumvent this difficulty without wasting further time on source preparation, the following averaging procedure was carried out. The extrapolation chamber as later discussed, calibrated the average surface dosage rate of the sources over a central area of about 1" diameter. All film exposures were therefore densitometered at 23 points systematically located over the central area, thus obtaining an average exposure corresponding to the average dosage rate as measured by the extrapolation chamber. For each individual source discussed below, a quantitative measure of the non-uniformity is given in terms of the standard deviation of the exposure within the central 1" diameter circle. Two standard deviations are given: \( \sigma_n \) is the standard deviation for individual points in the area, and \( \sigma' = \frac{0.2\sigma_n}{n} \) is the standard deviation associated with the average of 23 points. \( \sigma' \) is the statistical error to be attributed to the thin source film data of Chap. 7.

D. Preparation of Individual Sources

In this section, the method of preparation of individual sources, and the results obtained, are described. The activity of the sources in terms of \( \mu \)c is not specified because it was never determined accurately. In all cases, however, it was of the order of 1 \( \mu \)c/cm\textsuperscript{2}.

1. \textsuperscript{35}S

Test tube #1

\begin{align*}
0.2 \text{ ml} & \quad 2\% \text{ BaCl}_2 \\
1.0 & \quad \text{H}_2\text{O}
\end{align*}
Test tube #2
0.3 ml  1% casein in 0.03 N NaOH
0.2  1.2% Na$_2$SO$_4$
1.0  H$_2$O
0.1  active solution (weak H$_2$SO$_4$)

Test tube #1 poured into #2, 1.5 ml H$_2$O added, stirred, poured onto lucite disc. Average source thickness about 0.25 mg/cm$^2$. $\sigma_n = 4\%$, $\sigma = 1\%$.

2. $^{185}$W

Formula only very approximate
Test tube #1
2 ml  0.5% active Na$_2$WO$_4$
Test tube #2
2 ml  0.5% bentonite suspension

Test tube #1 poured into #2, stirred, poured onto lucite disc. Average source thickness very roughly 0.5 mg/cm$^2$. $\sigma_n = 10\%$, $\sigma = 2\%$.

3. $^{204}$Tl

Test tube #1
0.5 ml  1% gum arabic
0.4  8% KI
Test tube #2
0.5 ml  1% gum arabic
1.5  3.3% active TlNO$_3$

Test tube #1 poured into #2, stirred, poured onto lucite disc. Dried under heat lamp to reduce recrystallization into needles. Average source thickness about 2.2 mg/cm$^2$. $\sigma_n = 8\%$, $\sigma = 2\%$.
4. $^{91}Y$

Test tube #1
- 0.5 ml $H_2O$
- 0.5 0.5% albumin
- 0.5 1.8% oxalic acid
- 0.35 1% $NH_4OH$

Test tube #2
- 0.6 ml $H_2O$
- 0.5 0.5% albumin
- 0.1 10% active $Y(NO_3)_3$

Test tube #1 poured into #2 at 0°C, 2 ml $H_2O$ added, stirred, poured onto lucite disc. Average source thickness = 0.45 mg/cm$^2$. $\sigma_n = 2\%$, $\sigma = 0.5\%$.

5. $^{32}P$

Test tube #1
- 0.5 ml 0.1N NaOH
- 0.1 1% gum arabic
- 0.4 2% $BaCl_2$

Test tube #2
- 0.25 ml 1% $NaH_2PO_4\cdot H_2O$
- 0.1 1% gum arabic
- 0.5 active $H_3PO_4$ (very dilute solution)

Test tube #1 poured into #2, 1 ml $H_2O$ added, stirred, poured onto lucite disc which was very lightly aquadagged. Average source thickness = 0.3 mg/cm$^2$. $\sigma_n = 2\%$, $\sigma = 0.5\%$. This $^{32}P$ source was never actually used in photographic measurements.

6. RaD

Test tube #1
- 0.7 ml 1% gum arabic
- 0.5 10% $NaH_2PO_4\cdot H_2O$
Test tube #2

1 ml RaD (in 0.2 N HNO₃)
1 0.2 N HCl (probably not necessary)
1 15% Pb(C₂H₃O₂)₂·3H₂O
0.7 1% gum arabic

Test tube #1 poured into #2 while very hot, cooled, stirred, poured onto silvered brass disc as source mount. Thickness of source itself = 5 mg/cm², with additional 9.4 mg/cm² Al foil superimposed on source surface (to duplicate applicator, as described under applications). $\theta = 3\%, \sigma = 0.5\%.$
APPENDIX 10: EXTRAPOLATION IONIZATION CHAMBER

In this appendix are described the extrapolation ionization chamber and the measurements which I made with it in calibrating the surface dosage rates of the thin continuous spectrum β ray sources of Appendix 9. The design and operation of the chamber are discussed first. This is followed by an analysis of the theoretical principles underlying the instrument. Finally, an example is given of the use of the chamber in the calibration of one of the thin sources. For complete details on the apparatus and the associated principles, the reader is referred to Caswell's thesis (6-2).

A. Design and Operation of Extrapolation Chamber

The design of the parallel plate extrapolation ionization chamber itself is shown in the full scale diagram of Fig. 58, taken from Caswell. The chamber consisted essentially of 2 telescoping brass cylinders. The upper cylinder in the diagram contained the assembly supporting the source and absorbers, which served as the high voltage electrode of the ionization chamber; the lower cylinder contained the collecting electrode, together with associated guard ring and electrometer connections. The upper cylinder could be slid up and down inside the lower cylinder, by the micrometer lifting screws, so as to vary the electrode separation.

The source mount, which in my case was one of the 1/4" thick, 2 7/8" diameter lucite discs, was glued to the aluminum backing shown. The cellulose acetate absorbers, when used in conjunction with each source, were glued to the source assembly in such a way as to be held taut across and in contact with the source surface. The absorber surface, or source surface if no absorber was used, served as the electrode proper. The orientation of this electrode
Figure 58
EXTRAPOLATION CHAMBER
surface could be adjusted by means of 3 spring loaded positioning screws so as to lie in a plane parallel with the collecting electrode.

The 2.87 cm diameter collecting electrode was electrically isolated from the guard ring by means of a teflon insulator. Both collecting electrode and guard ring originally used were polystyrene, made conducting by a thin surface layer of aquadag. This choice gave an essentially "tissue wall" ionization chamber, a condition, as is shown later, which permits a simple conversion of ionization, the quantity measured, to tissue radiation dosage, as desired in the calibration of the source. However, stable measurements were difficult to obtain with this electrode. Consequently, all measurements were finally made with an aluminum collecting electrode and guard ring. The ionization thus measured was then converted to the ionization which would have been measured, had the electrode been tissue-like (lucite, for concreteness) with the aid of Baily's data (Fig. 47, Table 19). The conversion factor is 0.86, almost independent of the $\beta$ ray energy.

The collecting voltage was supplied by batteries. Most of my measurements were made at field strengths in the range 100 to 300 volts/cm, a range within which Caswell found the ionization current to be essentially independent of field strength.

Ionization currents were measured with a vibrating reed electrometer (P-1) operating in the rate-of-charge method. This electrometer is a null instrument, operating in such a manner that as the charge, and consequently voltage, build up on the input condenser, a measured inverse voltage is fed back by the instrument so as to constantly maintain the collecting electrode within a few millivolts of ground potential. The time required for a specified voltage to build up on the known capacitance is a measure of
the ionization current. In some of my measurements this time was too short to measure accurately. It was possible to circumvent this difficulty by switching calibrated batteries into the feedback circuit, thereby extending the voltage range which the instrument could cover. One peculiarity was encountered in this procedure which I was never able to explain. The switching of the batteries into the circuit had an effect equivalent to increasing the input capacitance by approximately 4%. Since this increase was constant from run to run, it was simply subtracted out.

The plate separation of the extrapolation chamber was determined in absolute magnitude by an electrical method. This method, described in detail by Caswell, was in principle merely a measurement of the interelectrode capacitance. It was accomplished by switching a known extra voltage onto the electrodes, and measuring the time required for the now known ionization current to deposit a charge on the plates sufficient to counterbalance that voltage. As a check on this plate separation determination, one could measure relative separations accurately by means of the micrometers on the plate positioning mechanism. These relative micrometer measurements served another useful purpose, in that they provided, when compared with the electrical measurements, a calibration of the electrometer input capacitance. The result of this calibration was found by Caswell to agree with that from a second independent calibration.

B. Theoretical Principles of Extrapolation Chamber

1. Gray cavity theorem

The purpose of the extrapolation chamber is to measure the radiation dosage rate, or energy dissipation rate per gram, at the source or absorber surface. The fundamental principle by means of which one converts measured ion-
ization to desired energy dissipation per unit mass is the so called Gray cavity theorem (G-4). Consider an infinitesimally small air filled cavity located inside a solid medium undergoing irradiation. The cavity theorem states that $J_m$ (the number of ion pairs formed per unit mass of air per second) is related to $E_m$ (the energy which would be dissipated per unit mass per second in the solid medium, were the cavity actually filled with this solid) through the equation:

$$E_m = J_m W_{air} \rho_m$$

In this equation, $W_{air}$ is the mean energy required in the formation of one ion pair in the air, and $\rho_m$ is the mass stopping power of the solid relative to the air. Both $W_{air}$ and $\rho_m$ are essentially independent of electron energy, at least in the energy range of interest to us here. $W_{air}$ is equal to $32.5 \pm 1.0$ ev/ion pair. $\rho_m$ may be computed from the expression:

$$\rho_m = \frac{(n_m)^{solid}}{(n_m)^{air}} f(Z)$$

where $n_m$ is the number of electrons per unit mass, and $f(Z)$ is a number close to unity for light materials but depending slightly on the mean atomic number $\bar{Z}$ of the material. $\bar{Z}$ is defined, as in Appendix 1, by the equation

$$\bar{Z} = \frac{\sum_i p_i Z_i^2}{\sum_i p_i Z_i}$$

where $p_i$ is the fraction of the atoms having atomic number $Z_i$, and the summation is over all the atoms in the molecule. Gray shows a graph for the dependence of $f(Z)$ on $\bar{Z}$, from which one finds that $f(\bar{Z}) = 1.015$ for lucite.

2. Extrapolation theory

In practice, of course, $J_m$ cannot be measured in
an infinitesimally small cavity. It is in surmounting this problem that the extrapolation chamber has its particular value. With this instrument one determines the ionization in cavities of several different volumes, and then extrapolates to $J_m$ at zero volume. To give confidence that the extrapolation is valid (particularly that the curve of ionization per unit volume vs. plate separation does not diverge at small separations), it is desirable to have the guidance of a theoretical extrapolation curve. The theoretical extrapolation curve is considered in this section.

I have thought it wise to investigate this problem in some detail, for the shape of this curve has been the source of considerable confusion both in our laboratory and in others.

It must be recognized at the outset that a pure theoretical treatment of the problem is very complicated. The reason is that in this problem, as will be seen more clearly later, both absorption and changes in angular distribution of the $\beta$ rays are taking place simultaneously, and the nature of each of these 2 effects directly influences the shape of the theoretical extrapolation curve. No simple theory of the interaction of $\beta$ rays with matter gives a sufficiently accurate picture of both of these effects to warrant a great deal of confidence in its predictions.

In Chap. 2 we briefly considered 3 separate theories designed to treat the distribution of ionization in the vicinity of $\beta$ ray sources. Brownell's theory is inapplicable to thin sources covered by rather thin absorbers, as we have already seen, because it is designed only for situations where equilibrium angular distribution exists. Furthermore, it deals only with infinitely broad sources and absorbers. Evans' treatment makes allowance for changes in angular distribution as caused by absorption, but not as caused by scattering. In view of the large amount of scattering which $\beta$ rays do undergo in passing through matter, this method of
analysis can be expected to have limited validity for the present problem. We shall see shortly that its predictions do not accurately reproduce the experimental extrapolation curve over the measured range. Loevinger's theory does not consider either absorption or scattering individually, but lumps together the net effect of the 2 in an experimentally determined function for ionization around a point source in a homogeneous medium. The theory does not apply, however, at the boundary between 2 such different media as gas and solid, for here one must have specific knowledge on both the angular distribution and the rate of absorption, rather than on the 2 combined. (Furthermore, we would not yet be really justified in using this theory, since, to my knowledge, the point source ionization function has to date been determined with accuracy only by the type of experiment we are trying to interpret.)

The limitations of these last 2 theories can be illustrated in Fig. 59.

![Diagram of a point source of β rays, located at S, being covered by an absorber. Evans' treatment says that no ionization will occur at P, because to get there by a rectilinear path the β rays would have to pass through a thickness of absorber greater than R_max. Loevinger's theory, in entirely omitting the question of scattering, gives no information as to whe-]
ther or not ionization occurs at P. Actually, ionization would be produced at P by the scattered β ray shown.

Despite the limitations of these completely theoretical analyses, I will first briefly treat the problem by Evans' method, which is the only one that can be applied at all. This is followed by my own semi-empirical analysis which more nearly represents the observed extrapolation curve over the measured range, and which therefore warrants more confidence in its predictions regarding extrapolation to zero gap.

a. Extrapolation curve by Evans' treatment

Consider a circular thin source, as seen edge on in Fig. 60, which has a radius = B and is covered by an absorber of thickness a. The surface of the absorber adjacent to the air volume is the high voltage electrode of the extrapolation chamber. The collecting electrode is located at distance s from the absorber. We wish now to find how the ionization per unit volume in the air gap varies as s is decreased, under the assumption that the β particles travel in rectilinear paths under exponential absorption. To simplify the problem further, we will also assume that we are interested only in the axial ionization, as though the collecting electrode were very small.

![Diagram of the setup](image.png)
It will be seen immediately that a start has already been made on this problem in Appendix 2. It was shown there that the dosage rate \( R \) at distance \( s \) from the absorber is given by the equation

\[
R = k \left[ -E_1(-b) + E_1(-b \sec \theta_0) \right]
\]

where \( k \) is a constant, \( E_1 \) is an exponential integral, and \( b = \mu_1 a + \mu_2 s \). \( \mu_1 \) and \( \mu_2 \) are the linear absorption coefficients associated with the absorber and air, respectively.

We should now integrate to find the average \( \bar{R} (= R_a) \) throughout the gap; this would be proportional to the ionization current per unit mass of air, and could be differentiated with respect to \( s \) to give the extrapolation curve slope. However, the degree to which this theory is valid can be estimated sufficiently accurately without going into all the complications of a rigorous solution. Therefore I will use an approximation for the slope that reduces to exactness when \( R \) varies linearly with \( s \). In this case,

\[
R_a = \frac{R_0 + R}{2},
\]

where \( R_0 \) is the ionization per unit mass at \( s = 0 \), and \( R \) is the ionization per unit mass at \( s \). The extrapolation slope \( = \frac{dR_a}{ds} = \frac{1}{2} \frac{dR}{ds} \). The fractional slope, which is the quantity I will deal with hereafter, is \( \frac{1}{2R_0} \frac{dR}{ds} \).

The differentiation of \( R \) can be carried out in a straightforward manner, with proper attention to the rules for differentiating the integrand of a definite integral. One finds:

\[
\frac{1}{2R_0} \frac{dR}{ds} = \frac{1}{2 \left[ -E_1(-\mu_1 a) \right]} \left\{ \left[ \frac{\mu_2}{(\mu_1 a + \mu_2 s)^2} - 1 \right] e^{-(\mu_1 a + \mu_2 s)} \right. \\
- \left. \frac{\mu_2}{(\mu_1 a + \mu_2 s)} e^{-(\mu_1 a + \mu_2 s)} \right\}
\]
To see the inadequacy of this prediction, we may compare it directly with the observed extrapolation curve of a representative example. I have picked at random the curve for $Y_{91}$ covered by 48 mg/cm$^2$. This curve is shown in Fig. 61; it is seen to have a slope which is essentially constant over the measured range with a magnitude of $-3.5\%$ per mm gap. Upon substituting into the theoretical equation, above, the value $B = 3.5$ cm, together with the other constants, one computes a theoretical slope as shown in Table 21.

<table>
<thead>
<tr>
<th>Gap</th>
<th>Slope</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 mm</td>
<td>$-0.28%/\text{mm}$</td>
</tr>
<tr>
<td>2</td>
<td>$-0.14$</td>
</tr>
<tr>
<td>1</td>
<td>$-0.13$</td>
</tr>
<tr>
<td>0</td>
<td>$-0.13$</td>
</tr>
</tbody>
</table>

It is obvious that the predictions are so different from the observed data over the measured range of the extrapolation curve that the theory is useless as a guide in extrapolating the observed data to zero gap.

The physical reason for the failure of the theory in this example is fairly clear; it is just that discussed with the aid of Fig. 5'?. In the formula giving the theoretical slope, the effect of finite source radius ($B$) enters only through the first term in the curly brackets. However, when numbers are substituted, $B/s$ in the exponent is so large that the whole term is negligible, and the effect of finite source size drops out of consideration. This is merely another way of saying the absorption of the oblique $\beta$ rays is so high that none reach the sensitive volume from
Figure 61
EXTRAPOLATION CURVE; $Y^{91}, 48 \text{mg/cm}^2$ ABSORBER
CURRENT/UNIT SEPARATION vs SEPARATION

\[ \frac{i}{d} \times 10^{12} \text{ (amps/cm)} \]

\[ d \text{ (cm)} \]

0 \quad 0.5 \quad 1.0 \quad 1.5 \quad 2.0 \quad 2.5 \quad 3.0 \quad 3.5
the edges of the source, so that a larger source would behave no differently. However, as described in Fig. 5, β rays actually do get scattered into the sensitive volume even from the edge of the source, so that the effect of finite source size is more important than this theory predicts.

Despite its inapplicability in the general case, this theory is useful in indicating what happens when there is no absorber over the source, for in this case the β rays would in reality have approximately rectilinear paths. The theory shows that the dosage rate at distance s from the source surface goes as \( R = -E_1(-\mu s) \), and this exponential integral diverges as s goes to zero.

b. Semi-empirical extrapolation theory

Since none of the simple β ray absorption theories provide a useful guide for the necessary extrapolation of the ionization/unit volume to zero volume, I have been forced to use a semi-empirical theory which is more adequate. The fundamental empirical facts underlying this theory are these: First, as has already been discussed in Chap. 2, β rays are emitted from a thin plane source with an isotropic angular distribution, but rather quickly take on a \( \cos \theta \) distribution after passing through absorbers over the source. Consequently, it seems entirely justified, in analyzing the extrapolation curve shape, to ignore the mechanism of interaction of the β rays with the absorber and simply to postulate an appropriate angular distribution at the absorber surface. For thick absorbers (perhaps greater than half of one half thickness) one may postulate a \( \cos \theta \) distribution. For thinner absorbers (perhaps in the range of a few percent of one half thickness, as in the case of the thinnest \(^9\)Y and \(^{204}\)Tl absorbers), one might take the first two terms \((\cos \theta - \frac{1}{3} \cos 3 \theta)\) of the
Fourier series expansion for an isotropic angular distribution. This approximation appears to emphasize the large angles to very roughly the same degree as the data Brownell obtained with absorption of $^{32}\beta$ rays in aluminum foil having a thickness equal to a few percent of one half thickness. We will see that both of these angular distribution functions give almost the same form of extrapolation curve, and therefore intermediate angular patterns present no problem. If desired, the theory could be extended so as to include successively more terms in the expansion for an isotropic distribution, but I will take it only through these first 2 terms.

The second fundamental empirical fact is that absorption of the $\beta$ rays in the air gap is negligible insofar as it affects the extrapolation curve. There is clear internal evidence for this fact: In the range of the extrapolation curves where experimental points were taken, I found almost the same slope for all 4 isotopes; since the absorption coefficients for $^{35}\text{S}$ and $^{91}\text{Y}$ are so different, absorption could not be an important consideration. (If it were possible to use conclusions from this type of measurement to interpret the measurement, I would simply point out that, as described in Chap. 2, Loevinger has already shown the dosage distribution function about a point source to be inverse square with initially little or no apparent absorption. Even those $\beta$ particles which traverse the air space parallel to the absorber surface can have a maximum path "length" of only a few mg/cm$^2$ in the gap. In this range Loevinger's inverse square law, with little absorption, is valid. The familiar exponential absorption of plane geometry presumably would, however, exist within the air space if its diameter were a great many mg/cm$^2$, as is the diameter of the solid absorbers used in actual measurements showing exponential absorption.)
The third empirical fact which is to be used is that $\beta$ rays are backscattered in sufficient numbers that an important fraction of the ionization in the extrapolation chamber is contributed by $\beta$ particles backscattered one or more times from the electrodes. The magnitude of this backscattering has been investigated in detail in Appendix 1. On the basis of the definition of $f_d$ there used, it can be seen that the ionization contributed in a thin layer of air adjacent to the collecting electrode, by $\beta$'s backscattered from that electrode, is equal to $f_d \times$ the ionization contributed by the incident $\beta$ rays. The same is true of the other electrode. The magnitude of $f_d$, as a function of $Z$, is to be taken from the middle curve of Fig. 48.

Starting with these three sets of rather well verified empirical facts, we wish to compute the extrapolation curve shape. The problem of determining the average ionization per unit volume in the air gap is now a purely geometrical one; the reason the ionization per unit volume increases with decreasing $s$ is simply that fewer $\beta$ particles "leak out" the sides of the interelectrode gap when the gap is small. The computation is carried out with reference to Fig. 62.

![Diagram](image-url)
The calculation begins very similarly to that of Appendix 2. We first compute the energy dissipation per unit mass in an incremental volume $\Delta A \Delta s$ of air, on the axis of the electrodes and at the collecting electrode surface. Consider an elementary ring on the absorber surface of radius $y (= r \sin \theta)$ and width $dy (= r \cos \theta \, d\theta)$. The number of $\beta$'s/sec emitted from this ring, $dN$, is given by:

$$dN = k_1 2 \pi y \, dy = k_1 2 \pi \frac{r \sin \theta \, r \cos \theta}{\cos \theta}$$

The fraction of the $\beta$'s from any point on this ring, emitted per unit solid angle at the angle $\theta$, is equal to $k_2 f(\theta)$, where $f(\theta)$ is the appropriate empirical angular distribution function (e.g., $\cos \theta$). The fraction from any point which pass through $\Delta A$ is equal to $k_2 f(\theta) \frac{\Delta A \cos \theta}{r^2}$. The total number $dN_v$ of $\beta$'s/sec emitted from the ring and passing through $\Delta A$ is given by:

$$dN_v = k_1 k_2 2 \pi r \sin \theta \frac{r \cos \theta}{\cos \theta} f(\theta) \frac{\Delta A \cos \theta}{r^2}$$

$$= k_2 \frac{\Delta A \sin \theta}{r^2} f(\theta) \, d\theta$$

If the absorption coefficient of air is given by $\mu_2$ (at this stage we must introduce absorption, or there would be no dosage), the energy dissipation by each $\beta$ particle in the elementary volume of air $\Delta A \Delta s$ is, on the average, $\overline{E} \Delta s \frac{\Delta A \cos \theta}{r^2}$, $\overline{E}$ being the average energy of the $\beta$ rays. The energy dissipation by $dN_v$ $\beta$ particles is $dN_v \overline{E} \frac{\Delta s}{\cos \theta}$. The energy dissipation per unit mass is

$$dR = \frac{dN_v \overline{E} \frac{\Delta s}{\cos \theta}}{\rho \Delta A \Delta s} = k_4 \frac{\sin \theta}{\cos \theta} f(\theta) \, d\theta$$

$\rho$ in this case being the air density. The total energy dissipation per unit mass contributed by the entire source is

$$R = \int_0^{\theta_0} k_4 \frac{\sin \theta}{\cos \theta} f(\theta) \, d\theta$$

(16)
We will first treat the thick absorber situation, where \( f(\theta) = \cos\theta \). In this case,

\[
R = \int_0^{\theta_0} k_4 \sin\theta \, d\theta = k_4 (1 - \cos \theta_0) = k_4 (1 - \frac{s}{B})
\]

when \( s < B \). This approximation is entirely justified because in practice \( s < 0.1 \, B \). Hence this extrapolation curve has a constant (fractional) slope \( \frac{1}{2R_0} \frac{dR}{ds} = -\frac{1}{2B} \).

With \( B \) again = 3.5 cm, the slope predicted by this expression = \(-14\% / \text{cm} = \sim 1.4\% / \text{mm} \). The experimental data, an example of which has been shown in Fig. 61, does have a constant slope over the measured range, but its magnitude is greater than 3\%/mm.

However, the theory developed thus far is incomplete for two reasons. First, it treats only the axial ionization, instead of all the ionization in front of the 2.9 cm diameter collecting electrode. Second, it ignores scattering back and forth between the electrodes, with loss of electrons out the sides of the gap at each passage through the gap. These effects, both of which tend to increase the extrapolation slope, are now to be evaluated.

To calculate the average \( R \) across the finite width of the collecting electrode would be a complicated geometrical problem. A simple approximation which probably has rather good accuracy is the following. Consider the point \( P \), in Fig. 62, which is a distance \( x \) off axis. The source (i.e., absorber surface) could be split up into a large number, \( M \), of sectors, radiating out from a point on the absorber directly opposite \( P \). This is illustrated in Fig. 63, in which the source is observed from \( P \). The ionization contributed at \( P \) by sector number 1 is simply

\[
\frac{\theta}{2\pi} \times \text{the ionization which would be contributed by a circular source of radius } B_1, \text{ or } k_4 \frac{\theta}{2\pi} (1 - \frac{s}{B_1}). \]
\[ \alpha = \frac{2\pi}{N} \]

**Fig. 63**
at least for the initial passage of the $\beta$ rays through the
gap; because of reflections, however, it is actually a
slight approximation to the real situation.) The total
ionization at $P$ would equal the sum of the ionization con-
tributed by each sector. If the angle $\phi$ of all the sectors
were identical ($= \frac{2\pi}{M}$), then

$$R_P = \frac{k_4}{M}(1 - \frac{s}{B_1}) + \frac{k_4}{M}(1 - \frac{s}{B_2}) + \ldots + \frac{k_4}{M}(1 - \frac{s}{B_M})$$

$$= k_4 \left[1 - \frac{s}{M} \left(\frac{1}{B_1} + \frac{1}{B_2} + \ldots + \frac{1}{B_M}\right)\right].$$

The important conclusion may be drawn from these equations
that the linear form of the extrapolation is still preserved;
only its slope is changed. At this point the chief sim-
plifying approximation is introduced; instead of summing
(or integrating) over a large $M$, we let $M = 2$, and take
the maximum and minimum $B$'s: $B + x$ and $B - x$. This gives:

$$R_P = k_4(1 - \frac{sB}{B^2 - x^2}).$$

To find the average $R$, $\bar{R}$, over the collecting electrode
area, $\pi x^2$, $R_P$ is integrated over this area:

$$\bar{R} = \frac{1}{\pi x^2} \int_0^X k_4(1 - \frac{sB}{B^2 - x^2}) 2\pi x \, dx$$

$$= k_4(1 - \frac{s}{B} \left[B^2 \ln \frac{B^2}{B^2 - x^2}\right])$$

Substituting in for $B = 3.5$ cm and $X = 1.44$ cm, one has:

$$\bar{R} = k_4(1 - 1.09 \frac{s}{B}) = k_4(1 - .31 s)$$

The finite collecting electrode size is now seen to have
little effect on the extrapolation curve. The slope as
predicted with this refinement of the theory is still con-
stant with s, but has a slightly larger magnitude of 1.6%/mm.

The second refinement which is required deals with
the multiple back scattering between the electrodes. The
loss of electrons out the sides of the gap is cumulative
over successive "reflections", rather than entering only
once. It has been shown in Chap. 2 that backscattered elec-
trons have an essentially cosθ angular distribution. There-
fore the "reflected" electrons behave in the same manner as
the initial electrons, and the same analysis as above ap-
plies for each reflection. (This statement is not complete-
ly correct, for the reason that the reflected electrons
have a slightly lower intensity at the edge of the elec-
trode than at the center, whereas the initial electrons
had the same intensity at all points of the absorber sur-
face. However, this effect will be rather unimportant for
2 reasons: First, it has already been shown that the in-
tensity does not drop off rapidly with distance from the
center. Second, most of the ionization is caused by re-
flections in the center, where the collecting electrode is
located, rather than out at the edges of the guard ring
and high voltage electrode (absorber).)

To sum up the ionization contributions of the
successive reflections, it is necessary to treat from the
outset the average $R$ throughout the depth of the gap (which
I call $R_a$ ) instead of $R$ at the collecting electrode sur-
face. We again assume that $R_a$ is equal to the average of
$R$ at the surface of the absorber (which is called $R_1$ ) and
$R$ at the surface of the collecting electrode ($R_2$ ). Hence

$$R_a = \frac{R_1 + R_2}{2}.$$  

If the ionization produced by the initial passage
of the $\beta$ rays through a thin air layer over the absorber has
the magnitude \( k_4 \), and if the \( f_d \) values associated with aluminum and cellulose acetate are \( f_{d,A} \) and \( f_{d,c} \), respectively, then successive reflections add up to give:

\[
\frac{\bar{R}}{k_4} = 1 + f_{d,A}(1 - .31s)^2 + f_{d,A}f_{d,c}(1 - .31s)^2 + f_{d,A}^2 f_{d,c}(1 - .31s) \frac{f_{d,A}^2 f_{d,c}(1 - .31s)^4}{(1 - .31s)^2} + f_{d,A}^2 f_{d,c}(1 - .31s)^6 + \ldots
\]

with other terms neglected. Similarly

\[
\frac{\bar{R}^2}{k_4} = (1 - .31s) + f_{d,A}(1 - .31s) + f_{d,A}f_{d,c}(1 - .31s)^3 + f_{d,A}^2 f_{d,c}(1 - .31s)^5 + f_{d,A}^3 f_{d,c}(1 - .31s)^5 + \ldots
\]

The extrapolation curve slope, expressed as a fraction per cm gap, is given by

\[
\frac{1}{R_o} \frac{dR_o}{ds} = \frac{1}{2R_o} \left( \frac{dR_1}{ds} + \frac{dR_2}{ds} \right)
\]

where \( R_o \) is \( R_a \) at \( s = 0 \). Although the derivative is not written out, it can be seen that it is to a small extent a function of \( s \); therefore the predicted extrapolation slope is not quite constant. When numbers are substituted into this formula, taking \( f_{d,A} = 0.41 \) and \( f_{d,c} = 0.28 \) from Fig. 48, one obtains the predicted slopes of Table 22, in terms of \%/mm gap at 0, 1, 2, and 3 mm. On the basis of this table one may say that for thick absorbers over plane sources, the predicted slope is -3.1 \%/mm, with a small dependence on the gap which for practical purposes is entirely negligible.
Table 22
Predicted Extrapolation Slope

<table>
<thead>
<tr>
<th>Gap</th>
<th>Slope</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 mm</td>
<td>-3.2%/mm</td>
</tr>
<tr>
<td>1</td>
<td>3.1</td>
</tr>
<tr>
<td>2</td>
<td>3.1</td>
</tr>
<tr>
<td>3</td>
<td>3.0</td>
</tr>
</tbody>
</table>

This prediction may be compared with the experimental data for the thickest absorber over each of my 4 thin sources. Considering all the experimental curves together, there was no consistent deviation from a constant slope in the measured range between about 1 and 4 mm. The magnitudes of the observed slopes are shown in Table 23.

Table 23
Observed Extrapolation Slopes (Thickest Absorbers)

<table>
<thead>
<tr>
<th>Source</th>
<th>Absorber thickness (in half thicknesses)</th>
<th>Observed slope</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\times^{21}$</td>
<td>1.3</td>
<td>-3.2%/mm</td>
</tr>
<tr>
<td>$T^{120}_{4}$</td>
<td>1.1</td>
<td>2.7</td>
</tr>
<tr>
<td>$W_{185}$</td>
<td>0.81</td>
<td>3.0</td>
</tr>
<tr>
<td>$S^{35}$</td>
<td>2.1</td>
<td>3.1</td>
</tr>
</tbody>
</table>

The observed and predicted extrapolation curves for thick absorbers may be considered to be in complete agreement both as to shape of curve (linear) and magnitude of slope.

To compute the extrapolation curve shape for thin absorbers over plane sources, we must go back to the initial formulation of this problem and insert into eq. (16) the appropriate function $f(\theta)$ for the angular distribution pattern of the $\beta$'s at the thin source surface. As previous-
ly stated, we may take \( f(\theta) = \cos \theta - \frac{1}{3} \cos 3\theta \) as approximately representing the thinnest absorbers used in this investigation (in conjunction with \( Y^9 \) and \( T^{104} \)). We have

\[
R = \int_0^{\theta_0} k_4 \frac{\sin \theta}{\cos \theta} (\cos \theta - \frac{1}{3} \cos 3\theta) \, d\theta
\]

\[
= k_5 (1 - 1.28 \cos \theta_0 + 0.285 \cos^3 \theta_0)
\]

Since \( \cos \theta_0 < 0.1 \), an entirely negligible error is committed in making the approximation \( \cos \theta_0 = \frac{\theta_0}{\theta_0} \) and ignoring the term in \( \cos^3 \theta_0 \). Hence we have

\[
R = k_5 (1 - 1.28 \frac{\theta_0}{\theta_0})
\]

This equation is identical in form with the analogous equation for \( f(\theta) = \cos \theta \), except for the constant coefficient 1.28. Hence the analysis for \( R \) is the same as before, giving

\[
\bar{R} = k_5 (1 - 1.09 \times 1.28 \frac{\theta_0}{\theta_0}) = k_5 (1 - .40 \theta_0).
\]

As was the case before, all "reflected" \( \beta \) ray angular distributions will be \( \cos \theta \); in only their first passage through the gap do the \( \beta \) rays have the \( \cos \theta - \frac{1}{3} \cos 3\theta \) distribution. However, as shown in Appendix 1, \( f_d \) has approximately the same value in both cases. Using these facts, we have

\[
\frac{\bar{R}_1}{k_5} = 1 + f_{d,A}(1 - .40 \theta_0)(1 - .31 \theta_0) + f_{d,A}f_{d,c}(1 - .40 \theta_0) x(1 - .31 \theta_0) + f_{d,A}f_{d,c}(1 - .40 \theta_0)(1 - .31 \theta_0)^3 + ...
\]

\[
\frac{\bar{R}_2}{k_5} = (1 - .40 \theta_0) + f_{d,A}(1 - .40 \theta_0) + f_{d,A}f_{d,c}(1 - .40 \theta_0) x(1 - .31 \theta_0)^2 + f_{d,A}f_{d,c}(1 - .40 \theta_0)(1 - .31 \theta_0)^2 + ...
\]
When the slope is computed by differentiation and substitution, as before, one finds the predicted extrapolation slope to equal \(-3.7\% / \text{mm}\) with approximately the same negligible dependence on \(s\) as before. The most significant feature of the derivation is that, for practical purposes, this extrapolation, also, is shown to be linear. The actual magnitude of the slope will depend on just what absorber thickness is involved, as \(f(\theta)\) is sensitive to this if the absorber is thin. The prediction for this \(\cos\theta - \frac{1}{3} \cos 3\theta\) angular distribution condition may be compared with my observed slopes for the thinnest absorbers over each source, as shown in Table 24. Again, there was no consistent deviation from constancy in the slopes over the \(s\) range measured.

<table>
<thead>
<tr>
<th>Source</th>
<th>Absorber thickness (in half thicknesses)</th>
<th>Slope</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{91})Y</td>
<td>0.41</td>
<td>4.4 % / \text{mm}</td>
</tr>
<tr>
<td>(^{204})Tl</td>
<td>0.077</td>
<td>4.4</td>
</tr>
<tr>
<td>(^{185})W</td>
<td>0.13</td>
<td>3.5</td>
</tr>
<tr>
<td>(^{35})S</td>
<td>0.41</td>
<td>3.2</td>
</tr>
</tbody>
</table>

By comparison with Table 23, one observes an increasing slope with decreasing absorber thickness, which is in agreement with the prediction. The magnitude of the slope associated with the thinner absorbers agrees with the prediction as well as can be expected in view of the uncertainty in \(f(\theta)\).

In the case of absorbers much thinner than those listed above, the slope would presumably be less nearly constant, and at 0 absorber thickness (and infinitesimal source thickness) would diverge as predicted by Evans' treatment previously considered. At what point the devia-
tion from linearity becomes important I do not know. Measurements which I made on the sources with no absorbers (but with finite thickness) still gave extrapolation slopes which were nearly constant over the range measured, but of larger magnitude. For example, the $^{91}\text{Y}$ source thickness was approximately 0.5 $\text{mg/cm}^2$, which should be roughly equivalent to an infinitesimally thin source covered by 0.25 $\text{mg/cm}^2$ absorber = 0.003 half thicknesses; its slope appeared to be a constant 10%. Although I have not attempted to work out a theory for this very thin absorber condition, I would expect a linear extrapolation of the linearly arranged experimental points to be reasonably accurate; this was the procedure followed in the measurements on bare sources just as on the sources with absorbers present.

In general, this semi-empirical theory has proved very successful in predicting experimental extrapolation curves over the measured range, so that its predictions in the extrapolation to zero gap may be confidently accepted. It is my hope that this semi-empirical theory will permanently lay to rest the ghost of a divergent extrapolation slope, for all but the very thinnest sources.

C. Example of Source Calibration

In this section a brief description is given of a typical calibration procedure, in order to tie together the preceding remarks on the instrumentation and theoretical principles. I will pick at random the case of the $^{91}\text{Y}$ thin source covered by a 48 $\text{mg/cm}^2$ cellulose acetate absorber.

The lucite disc on which the $^{91}\text{Y}$ was deposited, and the absorber, were glued in position as previously described. The outer surface of the absorber was then made conducting by a thin coat of aquadag, and electrically connected to the batteries. After tests, described by Caswell,
which showed the insulation between collecting electrode and ground to be good (about $10^{14}$ ohms) ionization measurements were begun.

At a small plate separation of $\sim 1$ mm, measurements were made of the time required for the ionization current to build up a voltage of 0.896 volts on the electrometer input capacitance of $12.0 \pm 0.1$ μf. These measurements were made in duplicate, with both positive and negative polarity on the high voltage electrode, so as to average out the primary β ray current always flowing into the collecting electrode. Next, the capacitance, and therefore separation, of the electrodes was measured by the electrical method previously described.

This whole procedure was repeated at 4 other values of s, increasing in steps of 0.0635 cm as accurately measured by the micrometers. The absolute separations as measured electrically increased almost identically in units of 0.0635 cm, indicating that the initial separation, measured electrically only, could be trusted. From these 5 sets of readings, the current per unit separation was calculated at 5 different separations, and normalized to standard temperature and pressure. These 5 experimental points on the extrapolation curve were then plotted vs. separation s (or d) as in Fig. 61, and extrapolated to s (or d) = 0. From the known collecting electrode area the ionization current per unit separation was converted to ionization current per unit volume and thence to ionization per unit mass of air. By the factor 0.86, from Baily's data, this value measured with an aluminum collecting electrode was then converted to the value which would have been obtained with a lucite collecting electrode, giving $J_m$ of the Gray cavity theorem. On substitution into that formula, $E_m$ was obtained. This is the quantity desired: the dosage rate at the surface of the absorber when in contact with lucite.
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