University of Richmond
UR Scholarship Repository

1965

An account of measurements of the mean range of Polonium-210 alpha particles in liquid water

Clyde Eugene Moss Jr.

Follow this and additional works at: http://scholarship.richmond.edu/masters-theses

Part of the Physics Commons

Recommended Citation

Moss, Clyde Eugene Jr., "An account of measurements of the mean range of Polonium-210 alpha particles in liquid water" (1965). *Master's Theses*. Paper 788.

This Thesis is brought to you for free and open access by the Student Research at UR Scholarship Repository. It has been accepted for inclusion in Master's Theses by an authorized administrator of UR Scholarship Repository. For more information, please contact scholarshiprepository@richmond.edu.

AN ACCOUNT OF MEASUREMENTS OF THE MEAN RANGE OF POLONIUM-210 ALPHA PARTICLES IN LIQUID WATER

by

CLYDE EUGENE MOSS, JR.

A thesis submitted to the Faculty of the University of Richmond in partial fulfillment of the requirements for the degree of Master of Science.

Department of Physics
Richmond, Virginia
1965

LIBRARY
UNIVERSITY OF RICHMOND
VIRGINIA

ABSTRACT

A comprehensive account of seven previous measurements of the mean range of Polonium-210 alpha particles in liquid water is presented. Suggestions for obtaining more accurate range measurements are made. The importance of the mean range value to radiobiology is mentioned.

III. TABLE OF CONTENTS

Chapter Page

I. TITLE........ 1

II. ABSTRACT........ 2

III. TABLE OF CONTENTS........ 3

IV. LIST OF FIGURES........ 4

V. INTRODUCTION........ 5

VI. RANGE ANALYSIS........ 12

A. INTRODUCTORY REMARKS

1. History........ 12
2. Bethe's Stopping Power Equation........ 15
3. Bragg's Additive Law for Stopping Power.. 17
4. Integral and Differential Stopping Powers 20

B. WORK OF PRINCIPAL INVESTIGATORS

1. Michl........ 22
2. Appleyard........ 30
3. DeCarvalho........ 32
4. Wilkins........ 32
5. DeCarvalho and Yagoda........ 33
6. Aniansson........ 38
7. Palmer and Simons........ 45

VII. DISCUSSION........ 56

VIII. ACKNOWLEDGEMENTS........ 63

IX. BIBLIOGRAPHY........ 64

IV. LIST OF FIGURES

Figure Page

1. Relationships of different ranges to the number-distance curve.. 6
2. Energy level diagram for the decay of Po^{210} to Pb^{206}..11
3. Relationship between wire size and blackened zones on film..24
4. Schematic drawing of experimental set-up employed by Michl..24
5. A parabola symmetric with respect to the x-axis....26
6. Construction of the "leading-line" LL...............26
7. Experimental relationship between wire diameter and blackened zones using liquid water.............28
8. Schematic drawing of experimental set-up used in Appleyard's work...31
9. A photomicrograph of alpha particle tracks from colloidal particles of radium and its daughter elements..35
10. Aniansson's experimental set-up.....................40
11. Counting rates versus distance curves for typical liquid..43
12. Experimental set-up used by Palmer and Simons......49
13. A typical T versus L plot............................50
14. Range-energy relation for alpha particles in liquid water found by Palmer and Simons.............53

V. Introduction

In recent years there has been a growing interest in Polonium^{210} (Po^{210}) since it is found in many substances[1-5]. Po^{210}, discovered in 1898 by the Curies, is a naturally occurring radioactive substance which emits monoenergetic alpha particles. It is only natural that a number of investigators have determined the range of this isotope in a medium as basic to cellular structure as liquid water.

Alpha particles are doubly-ionized helium atoms which are emitted by certain radioactive nuclei. As alpha particles go through matter they interact with the bound electrons of the matter losing energy mainly by excitation and ionization of the atoms. These particles undergo many interactions and gradually lose energy as they traverse the medium until they cannot produce further excitation and ionization, and then they come to rest. The average distance the particles travel is known as their mean range.

If the number of alpha particles arriving at a certain distance from the source is plotted as a function of the distance, the curve in Fig. 1 is obtained. If the particles had exactly the same range, then the curve would drop vertically at the distance equal to the range. However,

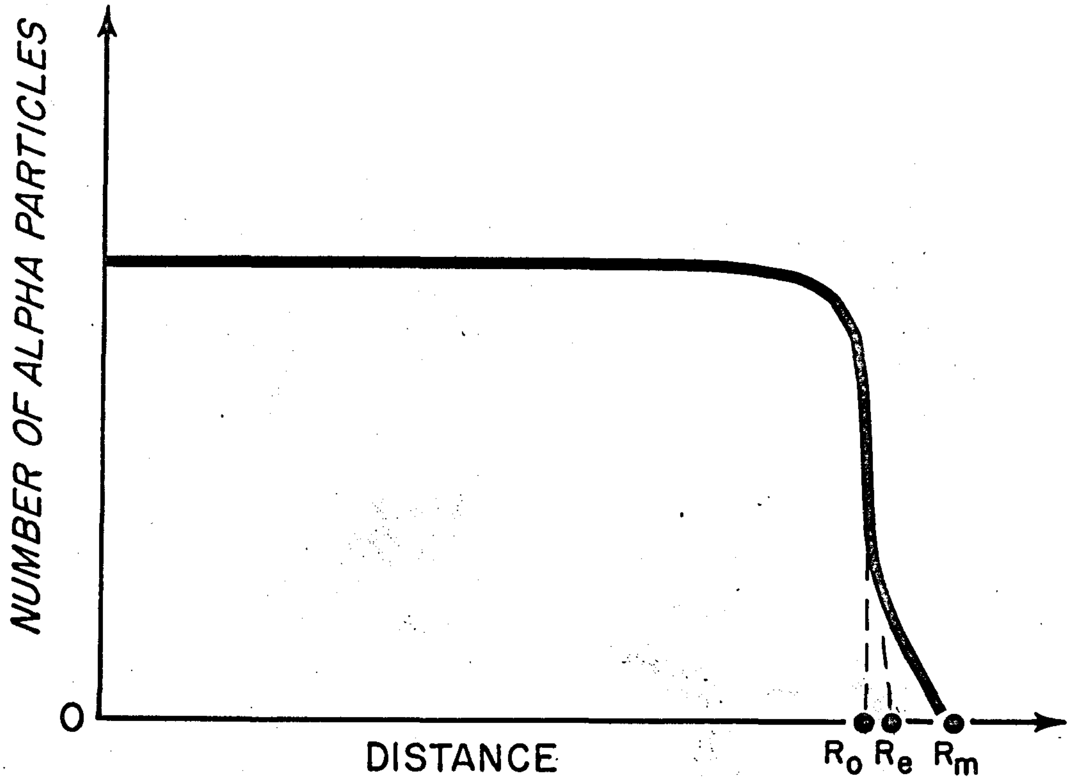

Figure 1 Relationships of different ranges to the number-distance curve.

the distance travelled by individual alpha particles varies because of their random interactions and collisions with the atoms of the medium. This variation, called straggling, prevents the curve in Fig. 1 from dropping vertically. Therefore, a mean range R_o is introduced and is defined as that distance at which one-half of the alpha particles have been stopped. This range is found by dropping a vertical line from the point of inflection and noting where it crosses the distance axis. The extrapolated range and the maximum range are also often mentioned in the literature. The extrapolated range is obtained by drawing the tangent to the curve at its inflection point and noting where the tangent line crosses the distance axis, while the maximum range is the greatest distance from the source that any alpha particle can travel.

Accurate assessments of the ranges of alpha particles in different media are necessary in the formulation of better stopping power theory. Stopping power is defined as the amount of energy lost by a charged particle per unit path length as it passes through a medium. Also, it is important to specify just where the end of the range occurs since the stopping power, or linear energy transfer (LET), is quite high near the end of the range. This means that radiological action depends on the distance that the alpha particle has travelled in a medium, and this dependence is, then, another reason why accurate range values are needed. In connection with energy loss, H. G. DeCarvalho[6] writes:

> "Since living biological tissues are composed to a large extent of water, then the range of alpha particles in this medium is of fundamental importance in the quantitative interpretation of the biological action of densely ionizing radiations, particularly with reference to the number of primary ionizations produced per micron of path, and in the determination of the rate of energy dissipation."

When one thinks of the weak penetrating power of alpha particles, capable of being stopped by a sheet of paper or by the dead tissue of a person's skin, one may tend to rule them out as a biological hazard. It is true that the weak alpha particles do not form an external hazard as do gamma rays, but they can cause serious damage when released from a radioactive substance that has been taken inside the human body. It is of interest, biologically speaking, to know how far certain radioactive particles will penetrate in liquids, especially liquid water, since tissue contains a high proportion of water. The diameter of a typical cell is about 40 microns, which is close to the range of Po^{210} alpha particles in liquid water. It can be shown that in liquid water the range values for alpha particles are within 1% of the ranges in tissue cells[7].

In following up these biological aspects R. F. Zirkle[8] has pointed out that the study of cell irradiation by biologists is hampered by three factors: the mean penetration into any cell is very difficult to estimate because most cells contain liquid water, and accurate range measurements in this substance are not easily done; the individual particles in a monoenergetic beam straggle; and the LET varies greatly near the ends of the particle tracks.

One of the reasons that Po^{210} is used in experiments concerned with the determination of the range of alpha particles is because it is practically 5.3 MeV monoenergetic. This is a trait greatly desired in range work since it eliminates the problem of determining which energy level gives what range. This reason, coupled with the facts that Po^{210} has a fairly long half life and is readily available, makes it a natural choice for experimental work.

$Polonium^{210}$ is the immediate disintegration product of $Bismuth^{210}$. The Polonium isotope emits alpha particles virtually all of which have 5.3054 MeV[9] of energy. It forms its daughter, Pb^{206}, at such a rate that the lead content of a freshly prepared Po^{210} sample increases about .05% a day. Pb^{206} is a stable isotope of lead and is the ultimate product of the Uranium-Radium family. Po^{210} has an atomic number of 84, and its alpha particles emerge from the nucleus with an initial velocity of 1.6×10^9 cm per second. Its disintegration constant is $4.95 \times 10^{-\cancel{8}3}$ days^{-1}, which yields a value of 140 days for the half life. Figure 2 gives the decay diagram for Po^{210}.

The purpose of this account is to review critically the literature which deals with the mean range of Po^{210} alpha particles in liquid water. A comprehensive review of this kind has never been published. In doing this study emphasis was placed on the uncertainties arising in the individual measurements.

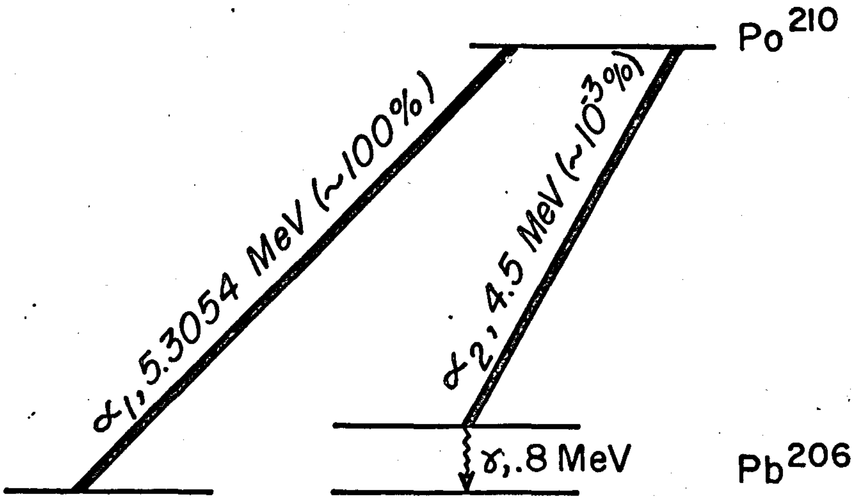

Figure 2 Energy level diagram for the decay of Po^{210} to Pb^{206}.

VI. Range Analysis

A. Introductory Remarks

1. History

The experimental measurements of the mean range of Po^{210} alpha particles in liquid water are few in number and varied in agreement, as Table 1 illustrates. There have been only seven reported values for this range in a period of fifty years. It is noted that the value for the mean range has tended to increase with time.

The first investigator of this range was W. Michl[10], who used wires coated with Po^{210} placed in contact with photographic plates surrounded by liquid water. He developed a method which led to a value in liquid water of 32 microns. Michl was the first to show that alpha particles have a definite range in liquids.

Thirty-five years later R. K. Appleyard[11-12] conducted experiments on the stopping power of liquid water using Po^{210} from which he calculated a value for the mean range. These studies were made with the aid of a thin-window Geiger counter which was used to measure the effective thickness of a Po^{210} source dispersed in water and hydrochloric acid.

In 1950 DeCarvalho[13], using emulsion techniques, found the range to be 39 microns. In this experiment

Table 1. The reported mean range in liquid water for Po^{210} alpha particles.

Author	Date	Mean Range (microns)
Michl	1914	32
Appleyard	1949	33.2
DeCarvalho	1950	39
Wilkins	1951	38
DeCarvalho & Yagoda	1952	38.1
Aniansson	1954	39.88
Palmer & Simons	1959	46

specially prepared radiocolloidal aggregates of polonium sulfate serving as micropoint sources were suspended in water which was in direct contact with the emulsion.

J. J. Wilkins in 1951 wrote an unpublished report for the British government (AERE G/R-664) on the moisture factor in nuclear emulsions in which he concluded that Appleyard's value of 33.2 microns was in error. His revised calculations showed a mean range of 38 microns for Po^{210} alpha particles in liquid water.

DeCarvalho and Yagoda[6] in 1952 wrote a very interesting paper on the ranges of alpha particles in water, ice, and heavy water. Their method was similar to DeCarvalho's 1950 effort in which radiocolloids and emulsions were used.

The next report came from G. Aniansson[14], who developed an entirely new approach for measurements of alpha particle range in liquids. His method consisted of placing a liquid between a flat piece of calcium tungstate crystal and a flat steel piston surface on which the source was deposited. A scintillation detector was placed in contact with the crystal, and a range value of 39.88 microns was determined for liquid water. Aniansson published a later paper[15] which strengthens his claims for the accuracy estimated for his first determination.

The most recent measurement of the mean range of

5.3 MeV alpha particles in liquid water was performed by Palmer and Simons[16] in 1959. Their work, based upon emulsion techniques, gave the highest value yet reported for this range, 46 microns.

There are two other methods for determining the mean range value of Po^{210} alpha particles in liquid water besides direct measurement. One of these methods involves evaluating Bethe's stopping power equation, and the other yields a range value from calculations based on Bragg's additive law for stopping powers.

2. Bethe's Stopping Power Equation

In 1930 Bethe, through a quantum mechanical treatment, derived an equation to account for energy losses in collision processes. His original expression has been modified over the years, and the equation generally given for the stopping power, $-\frac{dE}{dx}$, is

$$\left(\frac{-dE}{dx}\right) = \frac{4 \pi e^4 z^2 N}{mv^2}\left[Z\left\{\ln\left(\frac{2mv^2}{I}\right) - \ln\left(1 - \frac{v^2}{c^2}\right) - \frac{v^2}{c^2}\right\} - C_k\right] \quad (1)$$

where v is the velocity of the moving particle, m the mass of the electron, e the electronic charge, z the number of unit charges on the moving particle, I the average excitation potential of the absorber atoms, Z the effective nuclear charge of the medium traversed, N the number of atoms per cubic centimeter of the medium, and C_k a correction term that depends on absorber material and speed although it is unimportant at high speeds[17].

However, in the case of Po^{210} alpha particles passing through matter the rate of energy loss is given by the expression

$$\left(-\frac{dE}{dx}\right) = \frac{4\pi e^4 z^2}{mv^2} N \left[Z \ln\left(\frac{2mv^2}{I}\right) - C_k \right] \qquad (2)$$

since the relativistic effect is negligible at the velocity of Po^{210} alpha particles.

The stopping number B is used in defining relative stopping power. The defining equation for B is

$$B = Z \ln\left(\frac{2mv^2}{I}\right) \qquad (3)$$

In principle the expression for $-\frac{dE}{dx}$, the mean rate of energy loss, can be used to evaluate the mean range of a beam of alpha particles emitted with energy E_o by means of

$$R_o = \int_0^{E_o} \frac{dE}{-\frac{dE}{dx}} \qquad (4)$$

However, mere integration of equation (4) does not yield a precise value for the mean range. Despite the correction terms in equation (2) it is still not valid at low energies because of the capture and loss of bound electrons. Hence, in order to obtain an accurate mean range value it is necessary to use experimental range data for the low energy portion of the integration. Then

$$R_o = R_{oE_1} + \int_{E_1}^{E_o} \frac{dE}{\frac{-dE}{dx}} \qquad (5)$$

where E_1 is the lowest energy for which equation (1) is valid, and R_{oE_1} is the known mean range of alpha particles with energy E_1.

Calculations using Bethe's stopping power equation have still another disadvantage when liquid media are involved, for the stopping power equation has not been shown to hold for molecular media[18].

3. Bragg's Additive Law for Stopping Powers

There is still another way that might be used to obtain a value for the range, and it comes from Bragg's additive law for stopping powers. It was suggested by Bragg[19] that atoms of a medium act independently of each other as well as independently of binding forces in the process of stopping a charged particle. Hence, the energy lost by the charged particle in the medium can be considered as being the sum of the losses to the different species of atoms in the medium considered separately. This statement is known as Bragg's law, and for water the law means that the stopping power of a molecule of water is nearly equal to the sum of the stopping powers of the component oxygen and hydrogen atoms.

The key word is "nearly", since deviations could arise[20] from chemical binding, intermolecular interactions, and polarization. These three factors seem to be the greatest contributors to deviations from ideality.

The validity of this law has been tested by Aniansson[15], who obtained good comparative results for the stopping power of twenty-two hydrocarbons for Po^{210} alpha particles. Deviations of about 1% have been reported[21] for 340 MeV protons traversing several hydrocarbon liquids. It has further been reported by Gray[22] that departure from the additive law for stopping powers did not exceed 3% for thirty-eight range measurements in fifteen gaseous compounds of hydrogen, carbon, nitrogen, and oxygen except in three cases. Gray seems to believe that for alpha particle energies above 0.3 MeV deviations from Bragg's law should not exceed 2%; however, his conclusion is for gaseous compounds rather than liquids.

A strong interest in Bragg's law has been to learn whether the same matter in different phases really has the same stopping power per molecule regardless of phase. Allison and Warshaw[23] appear to support the idea of equivalence for stopping power per molecule of solid and gaseous phases within experimental uncertainties. However, there has been much disagreement among experiments as to whether water has the same stopping power value for all phases. Earlier experiments, those of Michl and Appleyard, seem to have indicated that the stopping power per molecule of the liquid phase was higher than that of the vapour phase; however, later data by DeCarvalho and Yagoda[6], Ellis[24], and Aniansson[14], definitely indicate these phases are close to being equivalent in terms of stopping power. In fact the last three authors obtained stopping power values which suggest that Bragg's law is valid for liquid water with an uncertainty of less than ±5%. It now appears that above 150 kev (This law is not valid at lower energies.) the stopping power of liquid water can be given by Bragg's additive law with an uncertainty not exceeding 3%.

The mean range of a substance and its molecular stopping power are related by the equation

$$\frac{r_o}{r} = \frac{d\ A_o\ S_{av.}}{d_o\ M} \qquad (6)$$

where r_o and d_o are the mean range in air and the density of air and r and d are the mean range in the test substance and its density[25]. A_o is the average atomic weight of air, M is the molecular weight of the test substance and S_{av} is the molecular stopping power, which in turn is defined as

$$S_{av.} = \sum_{i} N_i S_i \qquad (7)$$

Here the N_i's refer to the number of atoms of each type in one molecule, and the S_i's are the relative atomic stopping powers of the elements forming the test substance. A given S_i is defined as the ratio of the stopping number B of the given atomic species to that of air.

At the present time departures from Bragg's law are so small, perhaps because of inaccuracies in stopping power data, that the measurements seem to imply that the binding of an atom in a molecule has no effect on its stopping power.

4. Integral and Differential Stopping Powers

In discussing the stopping power of liquid water it is necessary to distinguish between differential and integral stopping powers. In order to differentiate between these terms the differential stopping power is denoted as s, while the integral stopping power is designated as S.

The differential stopping power of one molecule of water relative to the average atom of air is

$$s = \frac{s_w}{s_a} \qquad (8)$$

where s_w is the stopping power of a water molecule at a particular particle energy, and s_a is that of the atom of air at the same energy. To determine s the thicknesses of air and water required to reduce the mean energy of the alpha particles from some initial energy E_o to a final value E_1 not much smaller than E_o are measured. These thicknesses are expressed as the number of atoms per unit area in the case of air and as the number of molecules per unit area in the case of water. The ratio of these two quantities gives the mean differential stopping power of water. The defining equation is

$$s = \frac{\int_{E_1}^{E_o} \frac{dE}{s_a}}{\int_{E_1}^{E_o} \frac{dE}{s_w}} \tag{9}$$

The integral stopping power for alpha particles of initial energy E_o is the ratio of the range in air, expressed as the number of atoms per unit area, to the range in water, expressed as the number of molecules per unit area, or

$$S = \frac{\int_{o}^{E_o} \frac{dE}{s_a}}{\int_{o}^{E_o} \frac{dE}{s_w}} \tag{10}$$

It should be clear that usually differences will exist in the values of these two quantities.

There have been very few s measurements in water since the range can be measured more easily and expressed in terms of S. It is S that is needed to apply Bragg's law.

Since the methods that have been used to find the mean range in liquid water of Po^{210} alpha particles have been covered briefly, it is now possible to discuss critically the reported measurements of this range.

B. Work of Principal Investigators

1. Michl

The first experimenter to report a measurement of the mean range of Po^{210} alpha particles in liquid water was Wilhelm Michl in 1914. He undertook this work in order to demonstrate conclusively that alpha particles in liquids have a well defined range value, which fact, prior to 1914, had not been proved. Since at that time no electrical method was available to measure accurately the postulated distances alpha particles would travel in liquids, Michl decided to use photographic emulsions.

His experimental set-up consisted of platinum wires, each covered with a thin layer of polonium, resting on photographic film which was immersed in the test liquid. The mean range was determined from the contour of blackening that appeared on the plate as a result of the action of alpha radiation.

His method, at least for liquid water, depended on measuring two variables and plotting them on graph paper. These two variables were the radius of the platinum wire used and the width of the blackened zone caused by alpha irradiation of the film. Appropriate extrapolation produced the range value.

In doing this experiment Michl used different size platinum wires, in fact eight wires, whose cross-sectional dimensions were determined very carefully. On the outside of each wire was deposited a very thin layer of Po^{210}. He designated the widths of the blackened zones by the letter y and the radii of the wires as x. Plotting corresponding values of x and y led to a curve like that shown in Fig. 3.

Figure 4 shows why a range value can be obtained from this plot. Suppose a wire of diameter 2x were laid on a photographic film in a container of some test liquid as shown in Fig. 4. The variable y is found by using a microscope, and R, the mean range in the liquid, can be calculated.

It is seen from Fig. 4 that

$$R = \sqrt{x^2 + y^2} - x \tag{11}$$

which can be written as

$$y^2 = R^2 + 2Rx \tag{12}$$

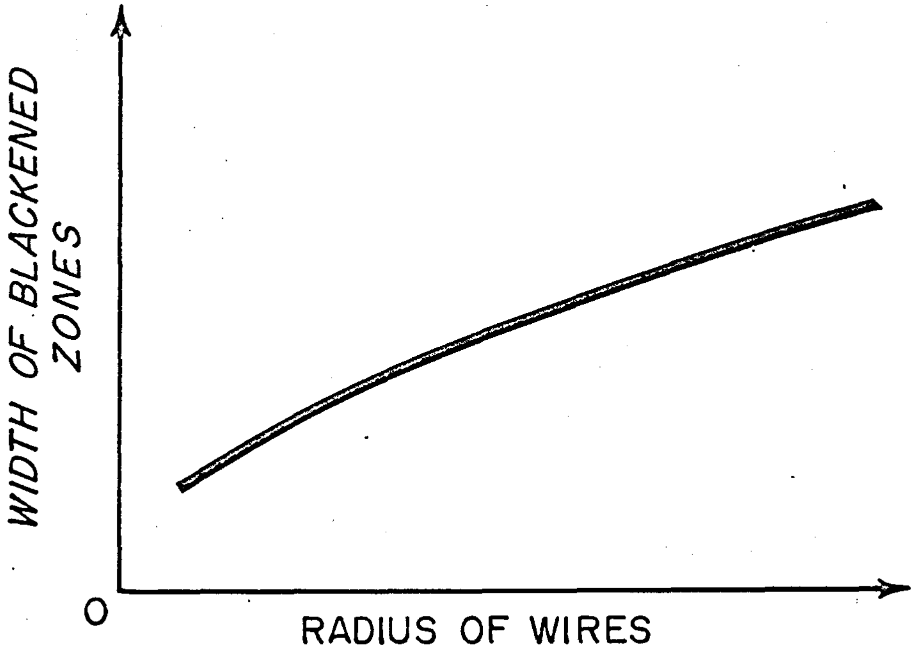

Figure 3 Relationship between wire size and blackened zones on film.

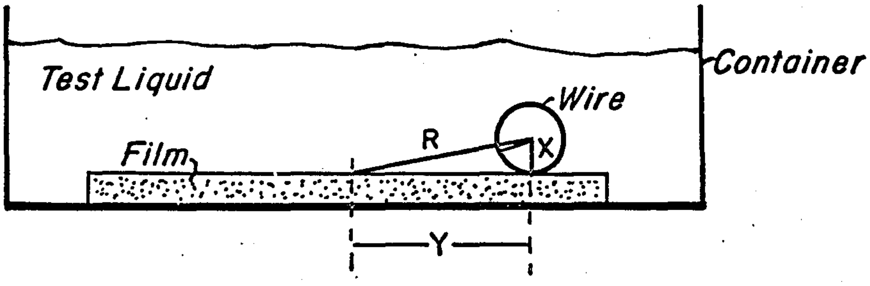

Figure 4 Schematic drawing of experimental set-up employed by Michl.

which reduces to

$$x = \frac{-R}{2} \tag{13}$$

when $y = 0$.

However, it should be obvious that the above mentioned results are true only for the ideal case when the wire does not sink into the film, when the liquid does not affect the gelatin (i.e. does not produce a milky cloud), and when the wire does not cut or scratch the film and hence affect the sensitivity. With each liquid tested it was possible by graphical methods to ascertain whether these factors were significant and to what extent they affected the range measurement. These things could be done by using what Michl called a "leading line."

In order to understand better the "leading line" concept, consider a parabola as shown in Fig. 5. The distances from the origin to P_1, P_2, and P_3 are called d_1, d_2, and d_3, respectively. If these distances are laid off in a direction parallel to the x-axis starting from the points P_1, P_2, and P_3 then a straight line can be drawn through the end points as shown in Fig. 6. This straight line is what Michl called the "leading line." It is, of course, the directrix of the parabola.

Since equation (~~8~~ 12) is parabolic in form, one would expect in all ideal cases to obtain a straight "leading line"

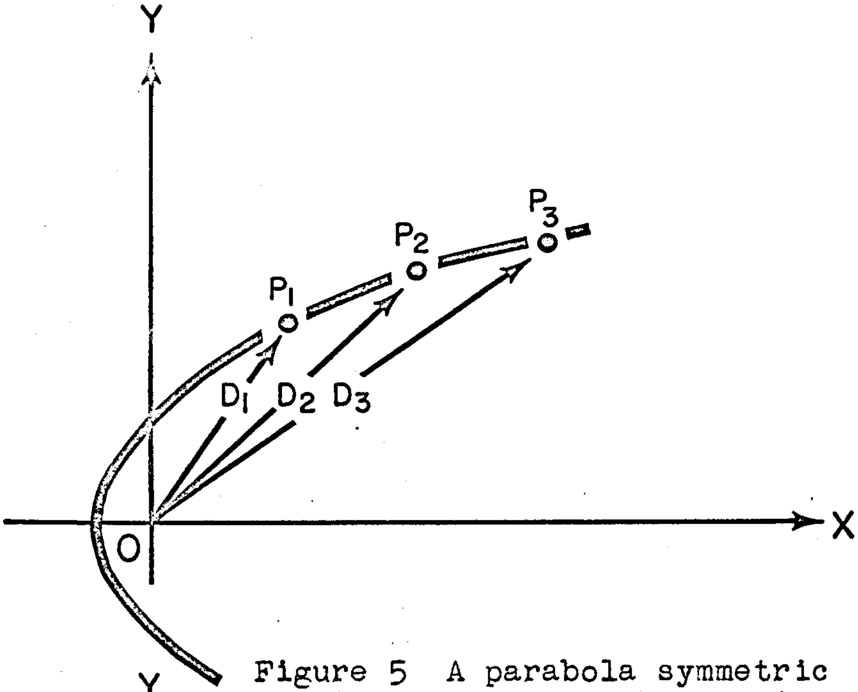

Figure 5 A parabola symmetric with respect to the x-axis.

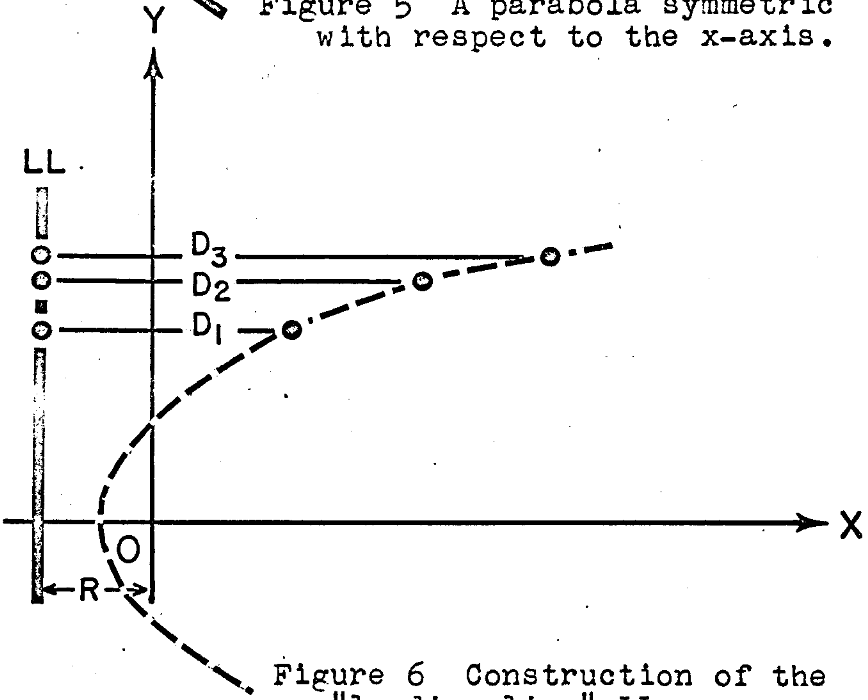

Figure 6 Construction of the "leading line" LL.

as shown in Fig. 6. If such a line is not present, then deviations from the ideal case must have occurred. Any such deviations must be accounted for in the measurements of the y variable, since the x values, the cross-sectional dimensions, do not change.

Not only does the "leading line" aid in detecting the presence of deviations in the experiment, but its extrapolation to the x-axis gives the mean range of the alpha particles in the liquid. The reason for this fact is implicit in equation (11).

Since water was the liquid that gave Michl the greatest trouble, it is not surprising that the "leading line" was not straight. Of course the increased weight of the larger wires caused depressions in the gelatin which made the y measurements too large. This meant that the mean range value in liquid water was based on the assumption that at smaller wire diameters the deviations were negligible. Figure 7 is a copy of the actual curves obtained by Michl for liquid water.

Michl considered many possible causes for variations in the range value such as the developing procedure, the length of time the film was exposed to alpha radiation, swelling of the gelatin, and solubility of Po^{210} in liquid water. He was not able to eliminate entirely errors arising from these causes but was able to minimize them. He did notice, however, that the greatest source of error in his range results, outside of the sensitivity of the film, was his inability to determine

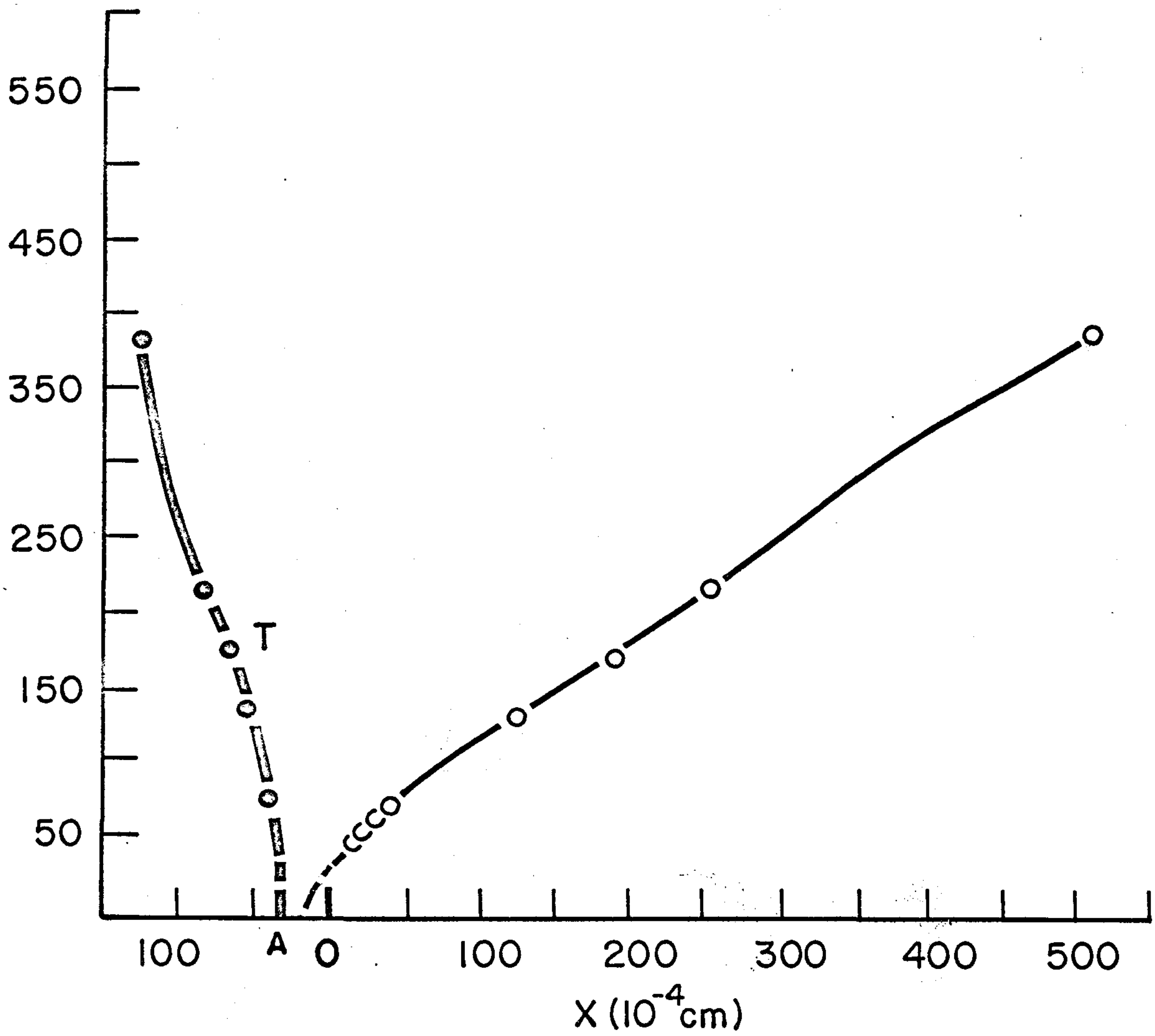

Figure 7 Experimental relationship between wire diameters and blackened zones using liquid water as the test liquid. The curve T is Michl's "leading-line." OA = 32 microns.

accurately the end points of the alpha tracks, a difficulty which still exists today.

Another source of error in Michl's work could have existed in his alpha source. It is an established fact that early work with Po^{210} was complicated by the extremely small quantities available and sometimes by incomplete separation of radioactive impurities. In fact it has only been within the last fifteen years that the chemistry of Po^{210} has been placed on a sound basis[26]. This could mean that Bi^{210} with an alpha group at 4.94 MeV could have been present in the solution from which Michl made his source. The smaller energy of the alpha particles from Bi^{210} could account in part for the fact that Michl's range values are smaller than those that have been obtained recently.

Michl's value of 32 microns for liquid water is about 17% less than the value calculated by Bragg's law. The value he determined for the integral stopping power was 1.83. Michl was the first to show definitely that alpha particles have a well defined range in liquids; he devised a photographic method for determining the mean range of alpha particles in liquids; and his mean range and stopping power values for liquids agree approximately with accepted theory. However, it is generally assumed that the films available in Michl's time were not of adequate response especially when swollen with liquid water for accurate evaluation of the alpha track near the end of its range. Therefore, his mean range value should probably be considered as an approximation only of the true value.

2. Appleyard

It was not until Appleyard made his measurements on the stopping power of liquid water that the range of Po^{210} alpha particles was determined again. This work by Appleyard did not include a range measurement, but it is mentioned here, because he calculated the mean range using his stopping power measurement. His experiment was the first designed to measure the stopping power for alpha particles in liquid water.

The unique method devised by Appleyard was based upon evaluating the effective thickness of an aqueous solution of Po^{210} in hydrochloric acid using a specially prepared Geiger counter as his detector. The variation in effective thickness of the source with the distance in air from the surface of the liquid to the counter was used to measure the stopping power of the liquid. Appleyard was able to determine a differential stopping power value of 1.71 for alpha particles between 4 and 5 MeV. Using this stopping power value, he determined the excitation potential I which appears in Bethe's stopping power equation. Using this value of I and the energy of the alpha particles from Po^{210}, Appleyard calculated the mean range value - a distance he found to be 33.2 microns.

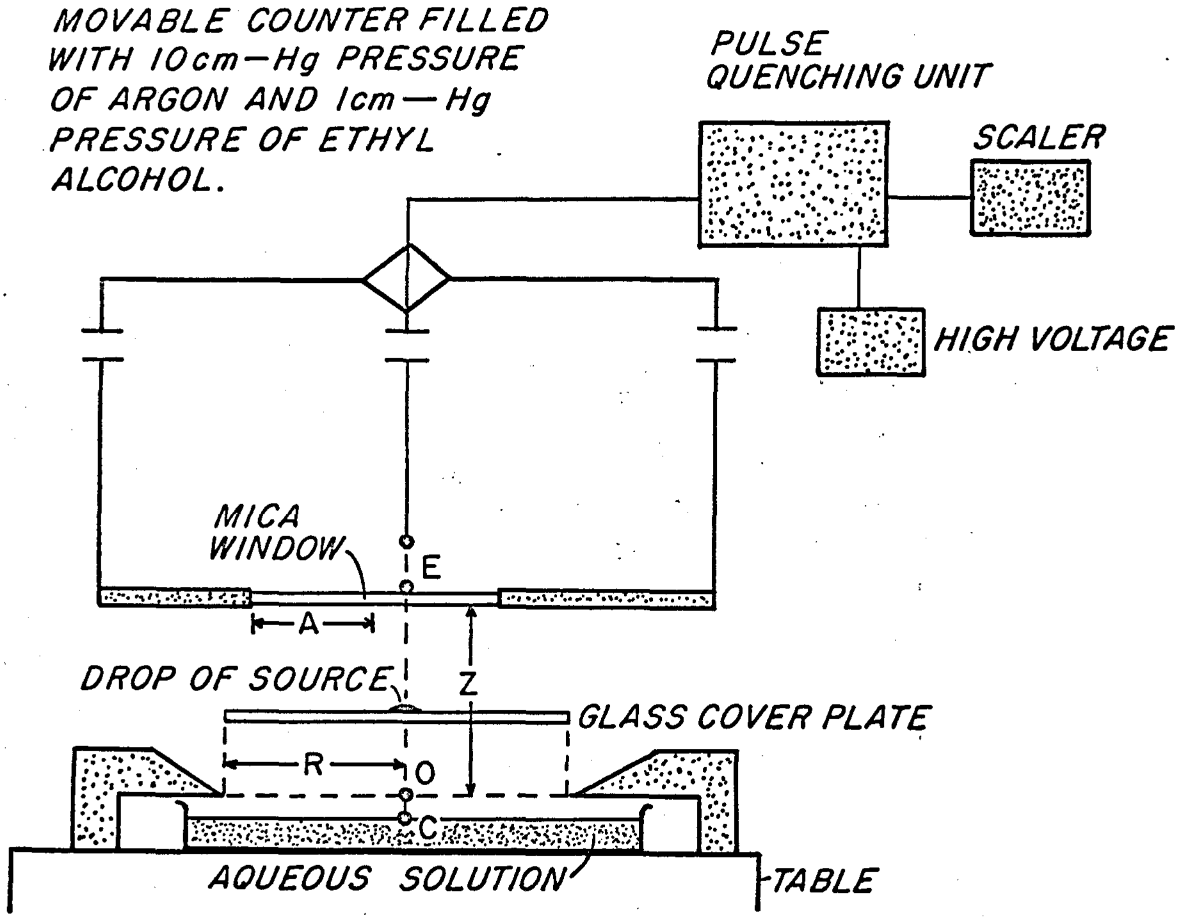

Figure 8 A schematic drawing of the experimental set-up used in Appleyard's stopping power measurements of liquid water.

3. DeCarvalho

The next investigator, H. G. DeCarvalho, reported 39 microns[13] as a preliminary estimate for the range of Po^{210} alpha particles in liquid water. This measurement was made using nuclear emulsions that were sensitive only to alpha particles which possessed energy greater than 0.2 MeV. DeCarvalho gives a complete description of his system in a later report written with Yagoda, which is discussed in detail later in this account.

4. Wilkins

An Englishman, J. J. Wilkins, was the fourth investigator to arrive at a value for the mean range of Po^{210} alpha particles in liquid water. His report, done for the Atomic Energy Research Establishment in England, carries the restriction "official use only," and, therefore, was not available to the author for examination. Wilkins carried out experiments dealing with the effect of moisture content on nuclear emulsions.

Other reports[13-14] indicate that Wilkins was able to find experimental values for the stopping power of water-loaded emulsions and to compute the stopping power of water using Bragg's law as a basis for calculation. Wilkins discovered some marked inconsistencies when he attempted to compare these stopping power values with Appleyard's result, and he concluded that Appleyard's values for the stopping power and mean range were in error. Wilkins calculated a value of 38 microns as the mean range of Po^{210} alpha particles in liquid water.

5. DeCarvalho and Yagoda

Emulsion techniques were used again in the experiments of H. G. DeCarvalho and H. Yagoda. Radiocolloidal particles of polonium chloride, which were compatible with the properties of the emulsion, were placed in contact with the surface of the emulsion which was immersed in distilled, air free water. The measurement of the alpha particle tracks emitted from the radiocolloids constituted a determination of the mean range in liquid water.

An ionizing particle travelling through a nuclear emulsion leaves a track containing a number of developable silver bromide grains. Whether or not a track is left depends on several factors such as specific ionization, development process, and background.

If the specific ionization is low, then the number of grains that are developable (grain density) will be proportional to the loss of energy, while for high specific ionization the grain density is constant because every grain in the path of the charged particle is developable. As far as development is concerned, it is not difficult to distort the gelatin during processing or to overdevelop the outer layer. This development process is very sensitive to external factors such as temperature, humidity, and oxidizing agents; furthermore, the phenomenom of fading of the latent image can occur[27]. Finally, the general background can "fog" the emulsion to such a degree that it may be difficult to distinguish a particular track. An example of tracks on an emulsion plate is shown in Fig. 9.

In the work of DeCarvalho and Yagoda there was a minimum of fading and distortion, because both the source and emulsion were kept in a refrigerator at a temperature of (5 ± 1)°C. Furthermore, the development process was stopped as soon as the surface abrasion marks became visible, and this certainly tended to minimize any overdeveloping.

Several drops of the source, polonium chloride in a slightly acid solution having an activity of 0.1 millicurie per milliliter, were placed on the emulsion plates which sat in a box filled to a depth of 5 millimeters with liquid water. Most of the colloidal particles stuck firmly to the surface of the gelatin and remained there during the development process.

As a radiocolloidal aggregate falls on the emulsion, alpha particles which are directed normal and nearly normal

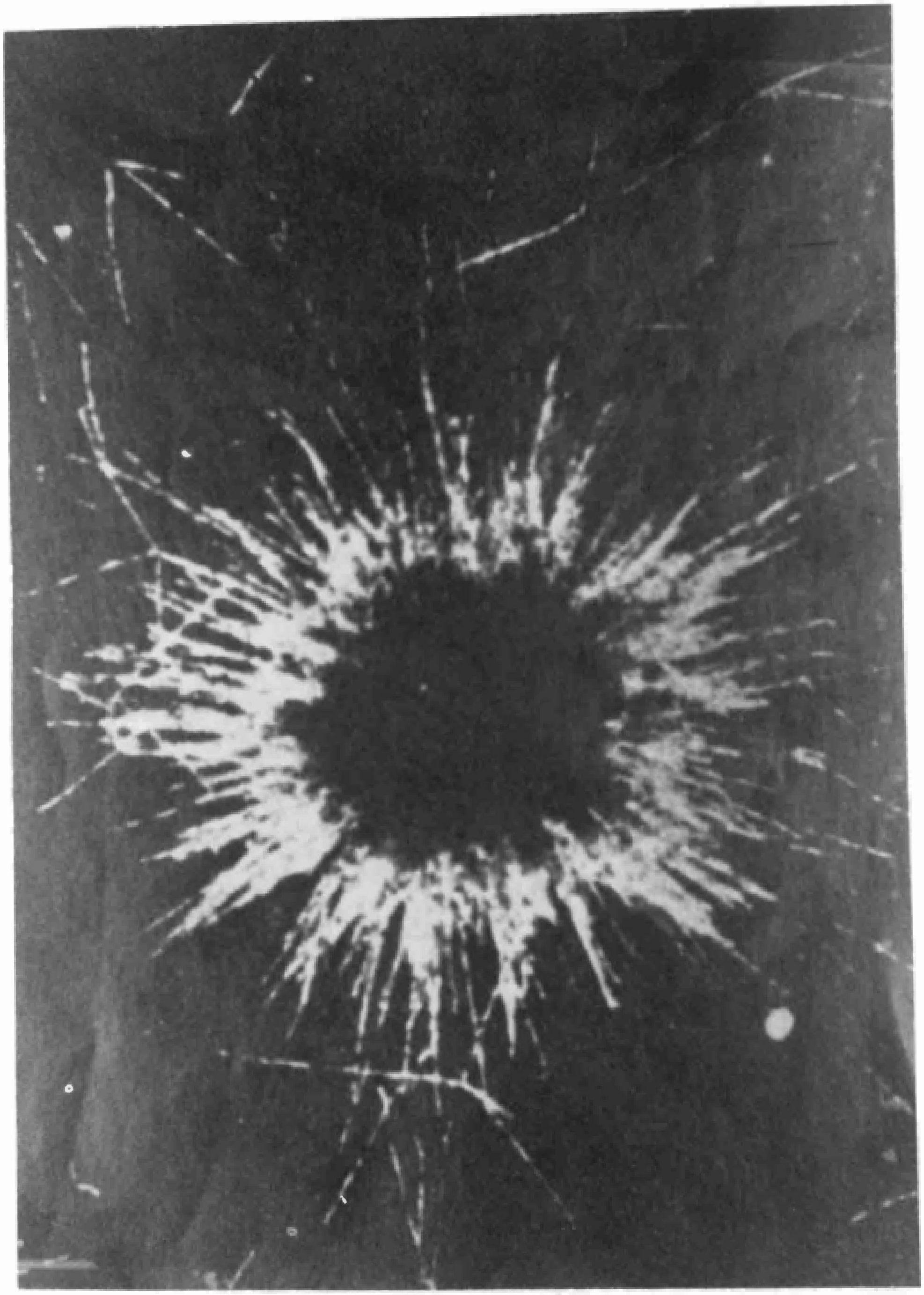

Figure 9 A photomicrograph of alpha particle tracks from collodial particles of radium and its daughter elements. Reprinted from The Tracks of Nuclear Particles by H. Yagoda, Scientific American Off-print 256, 1956.

to the plane of the emulsion will cause a black core of developed silver to appear whose area is a function of the activity of the radiocolloid, the energy of the particles, and the length of time the radiocolloid is exposed to the emulsion.

Since a radiocolloidal particle is about 0.1 micron in diameter, it can be considered a point source, and the center of the developed black core is the position of the source. The authors, using well-defined images that had at least ten recognizable end points, defined the average of these end points, measured from the last grain developed to the center of the radiocolloid, as the mean range of the alpha particles in liquid water. In order to insure consistent results these measurements were limited to radiocolloidal particles of average size which had their surrounding black cores circular in nature.

Both visual and photomicrographic methods were used to measure the actual ranges. In the visual method the center of the radiocolloid was judged by estimation of the center of the black core. This method was suitable as long as the blackening associated with the images was not severe. The visual method was naturally subject to human limitations; however, when a picture of the kind shown in Fig. 9 was taken of the image, the center was determined very precisely by drawing radial lines through the grain of the tracks. The center was taken as the average intersection of these lines.

To demonstrate the accuracy possible with nuclear emulsions, the range of Po^{210} alpha particles was determined in air. A coated steel needle was placed 0.3 millimeter from the emulsion and allowed to remain exposed to the film for 24 hours. The film was removed and developed and appropriate corrections were applied. The result was a range value of (3.82 ± .02) centimeters, which is in good agreement with the accepted value of (3.842 ± .0006) centimeters[28]. The end points of alpha particle tracks are much easier to determine for travel through air than travel through a liquid, and, therefore, the closeness of these two results does not imply a similar degree of accuracy for range values found for liquids. Since the emulsions were sensitive to alpha particles with energy greater than 0.2 MeV only, then all ranges determined were shorter than the true value and a slight correction had to be made to each final result.

DeCarvalho and Yagoda state that contamination to a small degree should not affect the range value, since impurities have a tendency to reduce the number of alpha tracks arriving at a certain point and not their maximum track lengths.

The results of fifty-eight measurements showed the mean range in liquid water to be (38.1 ± .5) microns and the integral stopping power to be (1.56 ± .02). The value for the range agrees well with Wilkins' result, and because of the reproducibility of DeCarvalho and Yagoda's results, these values are probably quite dependable.

6. Aniansson

In 1952 Aniansson developed a new method for determining the mean range of alpha particles in liquids. In his method the liquid was placed between two plane surfaces one of which was a scintillation crystal and the other of which held the source. This was the first time a scintillation counter was used for this measurement although Philipp[29] found the mean range of Po^{214} alpha particles by counting the visual flashes that were emitted when the alpha particles struck a zinc sulfide phosphor.

In a scintillation counter a charged particle hits the crystal (phosphor) where the particle can lose its residual energy by causing ionization and excitation of the atoms of the crystal. The energy lost is converted into photons. These photons strike the photo-cathode of the multiplier tube and cause emission of photo-electrons. The photo-electrons are attracted towards a series of dynodes each one having a higher potential than the previous dynode. Electrons are emitted by secondary emission at each of the dynodes. This electron multiplication process permits an appreciable number of electrons to reach the collector plate. At the plate a voltage pulse is produced the size of which is proportional to the energy lost by the alpha particle in the crystal. After amplification the pulse is counted by the scaler.

Aniansson's experimental set-up is illustrated in Fig. 10. A steel piston ground plane to within 0.1 micron is used for the source support and opposite it a piece of calcium tungstate crystal ground to the same tolerance is mounted. The distances in his experiments were measured with "mikrokators," devices available commercially in Sweden which Aniansson claims are calibrated to within 0.05 micron.

The assumption is made by Aniansson that the counting rate as a function of the distance between the surfaces can be expressed as

$$N = \int_{x}^{\infty} \frac{1}{2} N_0 \, S(r) \, (1 - \frac{x}{r}) \, dr \qquad (14)$$

where $S(r)$ is the straggling function, N the count rate at a particular distance between the surfaces equal to x, N_0 the total number of disintegrations per unit time taking place in the source, and r the variable in the straggling function.

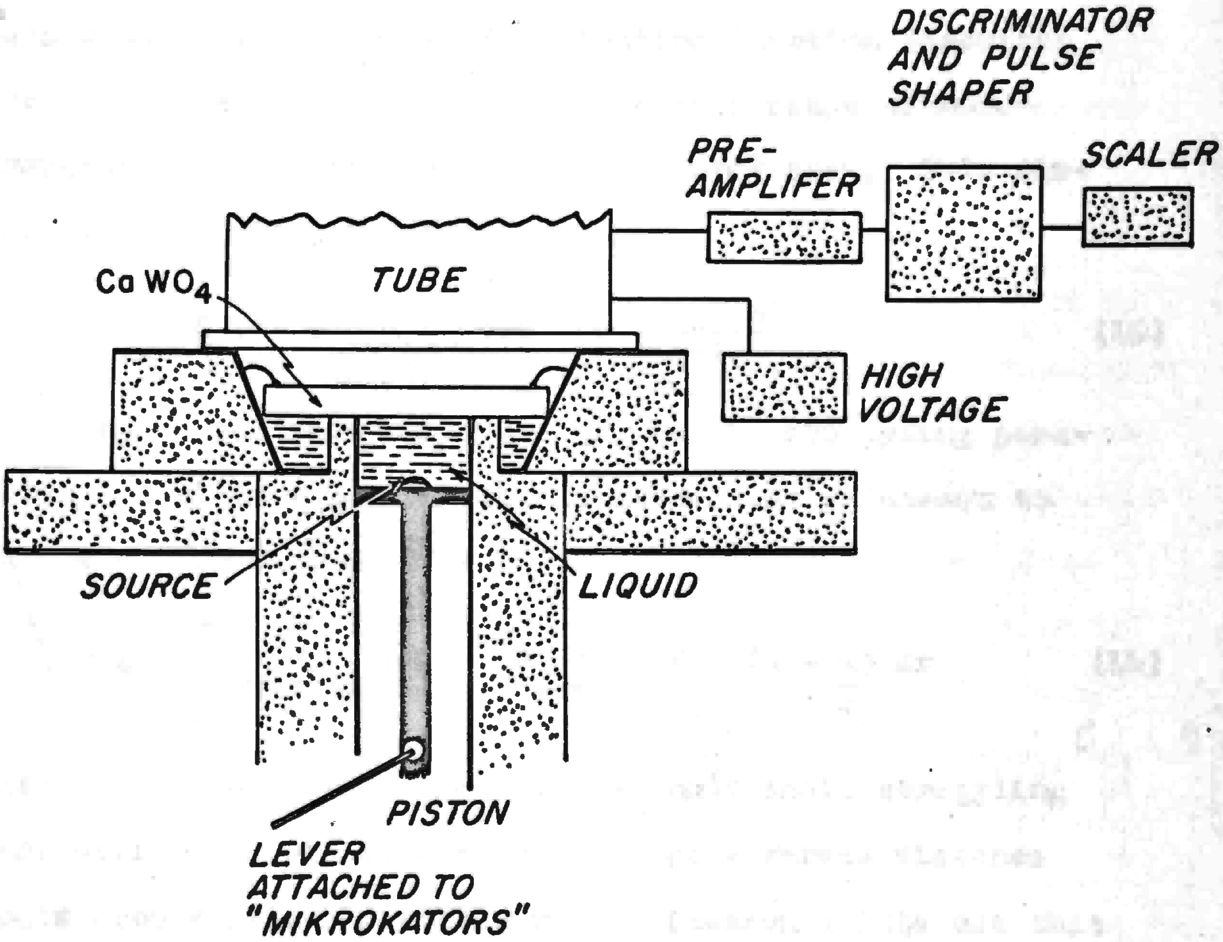

Figure 10 Aniansson's experimental set-up.

The fact that all of the alpha particles from a source fail to have the same range in any medium has long been known. The Gaussian distribution function describes the distribution in range about the mean range of mono-energetic alpha particles in a collimated beam. This distribution function is given by

$$G\ (r) = \frac{1}{\alpha\sqrt{\pi}} \exp \left[- \frac{(r-r_o)^2}{\alpha^2} \right] \qquad (15)$$

where r_o is the mean range and α is the straggling parameter. S(r) in equation (14) is assumed by Aniansson to take the form G(r) so that

$$N = \int_x^{\infty} \frac{1}{2} \frac{N_o}{\alpha\sqrt{\pi}} \exp \left[- \frac{(r-r_o)^2}{\alpha^2} \right] \left(1 - \frac{x}{r}\right) dr \qquad (16)$$

With a collimated beam and a vanishingly small straggling parameter the curve depicting count rate versus distance would drop vertically at $x = r_o$. Aniansson points out that with $\frac{\alpha}{r_o}$ vanishingly small equation (16), which takes into account the lack of collimation in his experiment, reduces to

$$N = \frac{1}{2} N_o \left(1 - \frac{x}{r_o} \right) \text{ for } x \leq r_o$$

and (17)

$$N = 0 \qquad \text{for } x \geq r_o$$

The straggling parameter is not much more than one percent of r_0; therefore, Aniansson claims that he observes equation (17) except for $(r_0 - \alpha) < x < (r_0 + \alpha)$. Within this small region the slope changes gradually, but these changes are of no consequence since he establishes the slope corresponding to equation (17) for $x << (r_0 - \alpha)$.

The plots of counting rate versus distance from the source will, then, give inclined straight lines that intersect horizontal straight lines which correspond to the background count rates at the discriminator levels used. A perpendicular line dropped from the intersection of an inclined line and its corresponding horizontal line to the x-axis would intersect the axis at a point which could be termed the mean range of the alpha particles which lost sufficient energy in the crystal to give pulses larger than the discriminator level used for this pair of lines. Aniansson calls such a mean range an apparent mean range. For example, if a perpendicular were drawn from point B in Fig. 11 to the x-axis, the intersection with the axis would indicate an apparent mean range of about 37.6 microns corresponding to a discriminator level of 10 volts.

Aniansson determined a series of points like B and K in Fig. 11 and was able to fit a straight line to them. He extrapolated this line to the zero discriminator level and

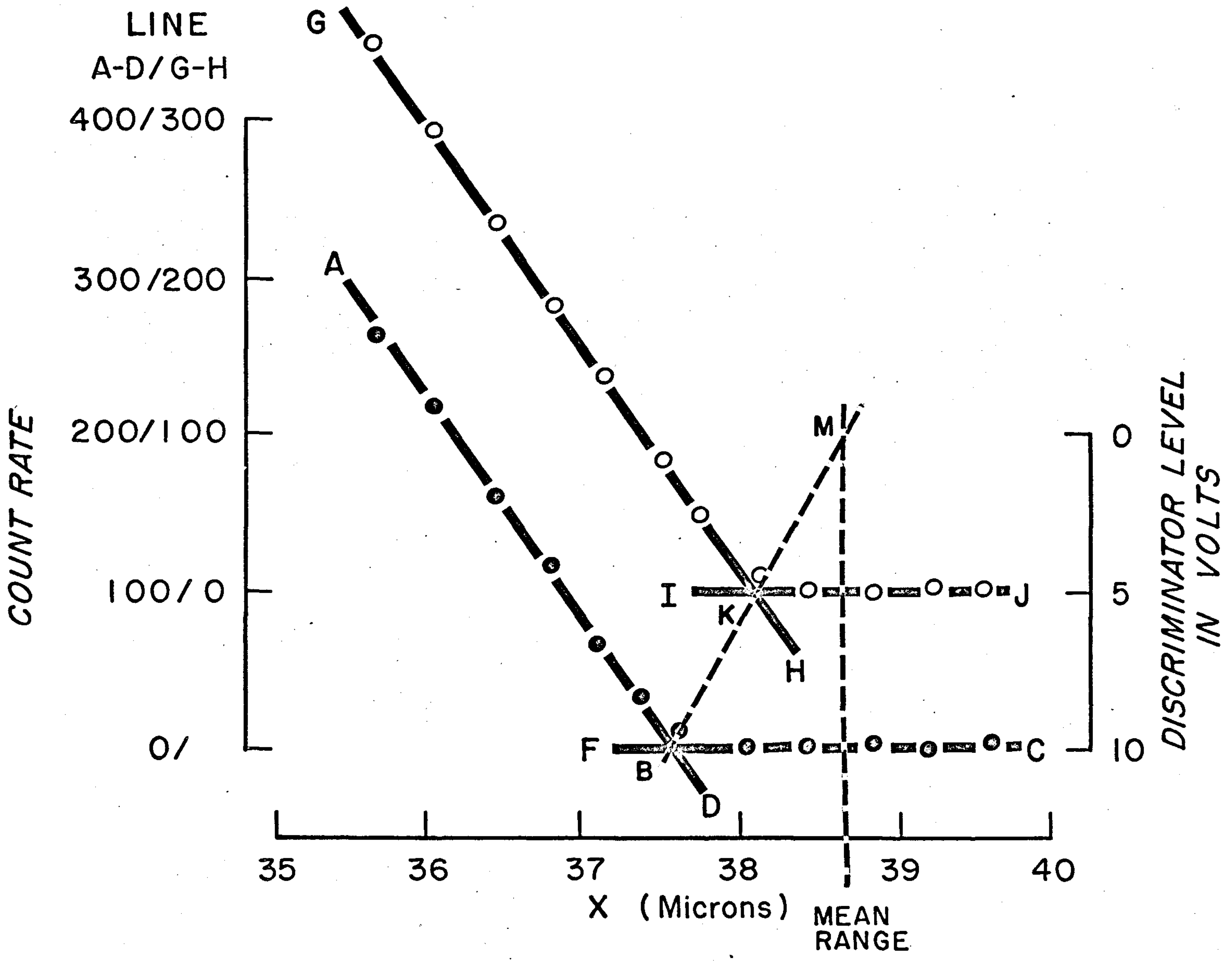

Figure 11 Counting rate versus distance curves for typical liquid.

obtained the point M. The intersection of the perpendicular line from M with the x-axis yielded for him the uncorrected mean range. Aniansson could not get data for the zero volt level, of course, because of the enormous interference from electronic noise at that level.

One source of error could lie in the method of deposition of the source[30]. Aniansson used an evaporation technique to place his source on the support. In order to check for the possibility that the source was deposited unevenly he took an autoradiograph. This autoradiograph showed that his source was evenly deposited and occupied a circular area 0.1 millimeter in diameter located at the center of the steel support. The autoradiograph and the fact that Aniansson used only one source in all the experiments lead to the conclusion that errors resulting from the characteristics of the source were minimal.

Aniansson found the range in air by the same method he used for liquids. His mean range for Po^{210} alpha particles in air, corrected to normal temperature and pressure, was 37.87 millimeters. This value is 1.4% shorter than the accepted value[28]. Aniansson claims that this difference resulted from a thin layer of aluminum deposited on the crystal surface and a layer of the crystal that was made insensitive by the grinding process. Therefore, he added a corresponding correction to the value of the mean range he had found graphically.

The thin layer of aluminum was added to reduce any possible energy transfer from the liquid to the crystal. This energy transfer mechanism is suggested and discussed by Birks[31]. Aniansson felt that some energy might be given to the crystal by the action of the molecules excited as a result of the irradiation process. After a series of experiments Aniansson concluded that this energy transfer to the crystal was negligible; however, in order to avoid alteration of the crystal the aluminum layer was left on its face.

Therefore, it appears that the major systematic errors associated with this measurement of the mean range of Po^{210} alpha particles in liquid water resulted from the layer of aluminum and the insensitive zone of the crystal. The mean range reported by Aniansson is 39.88 microns, which is 2.7% higher than that calculated by means of Bragg's law. He reported 1.469 for the integral stopping power, S, a value which agrees fairly well with that given by DeCarvalho and Yagoda.

7. Palmer and Simons

The latest published report of the range of 5.3 MeV alpha particles in liquid water appeared in 1959 as a result of the work of Palmer and Simons. They evaluated the range-energy relationship of alpha particles in liquid water, air, and water vapour using Po^{212} (8.78 MeV) alpha particles and nuclear emulsion techniques. They were able to measure, also, the differential and integral stopping powers of water, as shown in Table 2.

Their experiment can be discussed in two parts. The first part, which is of greater interest here, involves the evaluation of the range of Po^{212} alpha particles in liquid water as well as the relation between path length traversed and residual energy. The latter permits a range-energy curve to be constructed. The second part of their experiment deals with the factors involved in determining range-energy relationships for alpha particles in water vapour and air.

The first part describes the measuring techniques used in studying the penetration of alpha particles from Po^{212} through water layers of known thicknesses and into photographic emulsions. Since the range-energy relationship for alpha particles in the C-2 emulsion used was well-known[32], then the energy of a particle entering the emulsion could be determined, and, hence, the loss of energy on passing through a given amount of water could be found. The loss of energy in the emulsion and the corresponding length of the path through water could then be used to determine the range in water of alpha particles with energy equal to that lost in the emulsion after a definite value had been established for the total range of Po^{212} alpha particles in liquid water.

Table 2. Differential and integral stopping power values for liquid water at various alpha particle energies.

Energy (MeV)	Experiment*		Theory**	
	s_W	S_W	s_W	S_W
8	1.48	1.33	1.47	1.47
7	1.44	1.31	1.47	1.47
6	1.41	1.30	1.48	1.47
5	1.38	1.29	1.50	1.47
4	1.36	1.24	1.55	1.46
3.5	1.37	1.22	1.56	1.44
3	1.29	1.20	1.55	1.42
2.5	1.18	1.20	1.50	1.40
2	1.06	1.22	1.40	1.39
1.5	.98	1.30	1.33	1.40

*Measured by Palmer and Simons
**Calculated by Platzman (ref. 18)

Such data for different thicknesses of water led to a range-energy curve.

The Po^{212} was deposited on a portion of a highly polished steel cylinder and covered with formvar. A 100 micron C-2 research plate, also covered with a thin film of formvar, was the emulsion employed.

A thin layer of water was placed between the two formvar films, so there was water between the surface of the cylinder and the emulsion. By means of an x-ray shadow of the cylinder the line of contact of the cylinder on the plate was determined. Then Palmer and Simons were able to find a path length T in water corresponding to the measured residual track length L in the emulsion. To each of these residual track lengths a correction was applied for losses in the formvar film. Figure 12 shows a diagram of their experimental arrangement.

Measurements were made on six different plates, and the track lengths associated with each plate were grouped according to values of T. The averages of T and L were found for each group and a graph of T versus L drawn for each of the six plates. In all cases the curves obtained were straight lines with essentially the same slope. A typical plot appears in Fig. 13. The mean of these six straight lines was expressed as a single line intercepting the L axis at

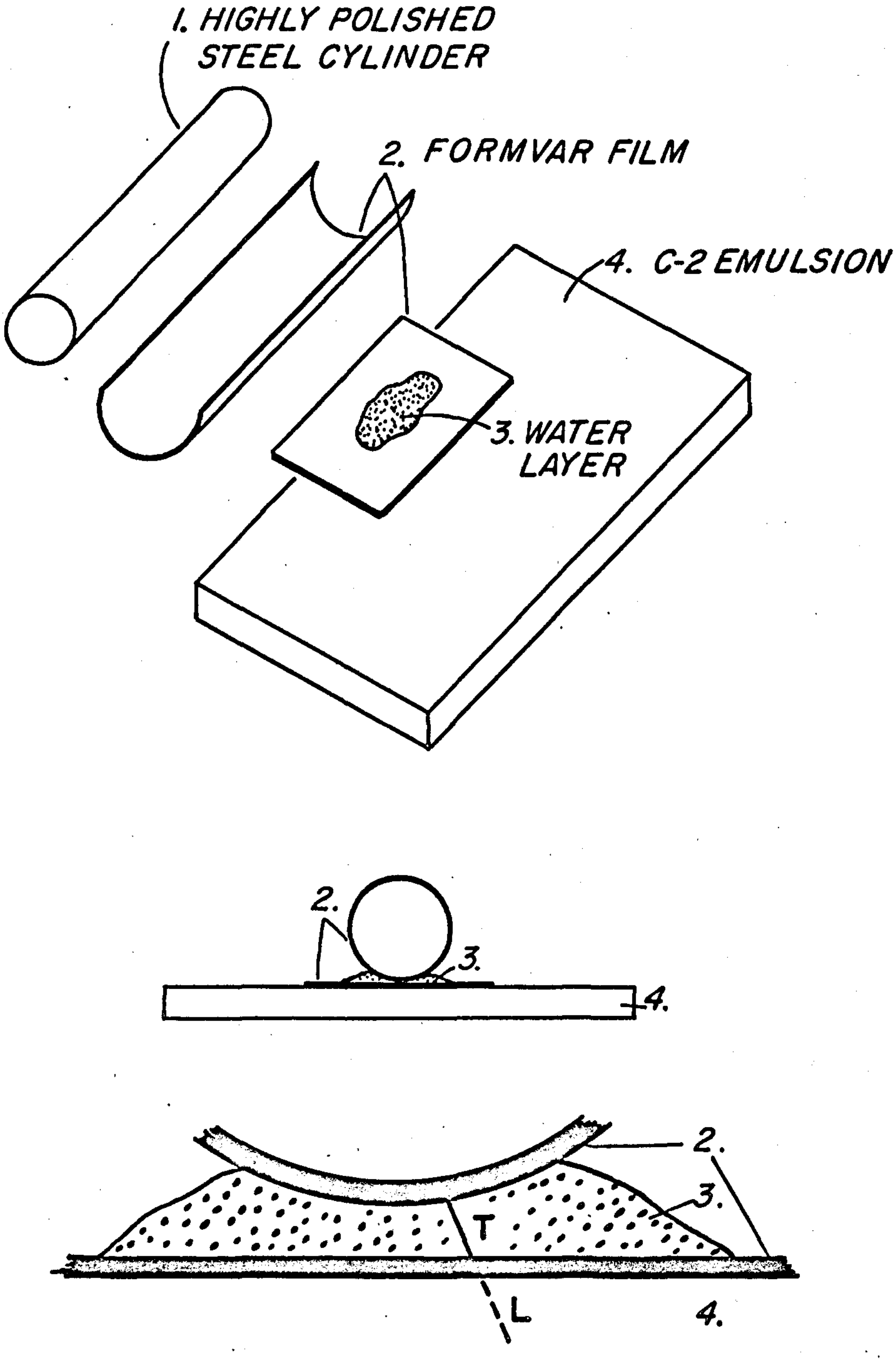

Figure 12 Experimental set-up used by Palmer and Simons.

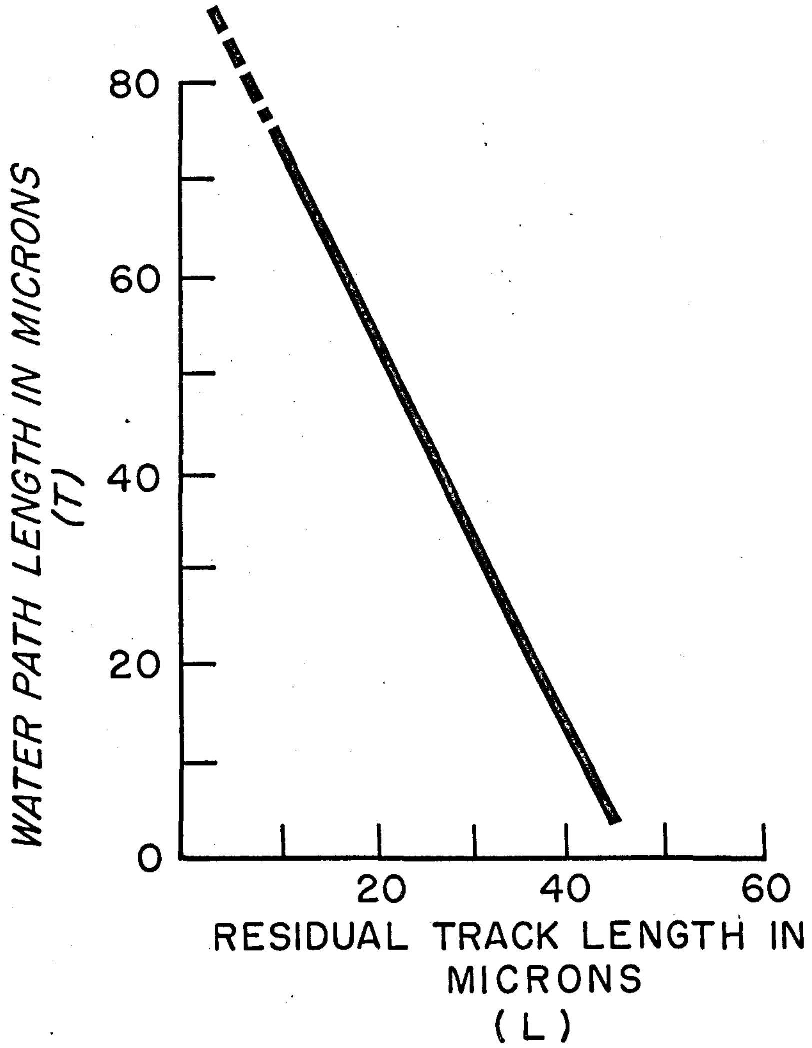

Figure 13 A typical T versus L plot for one of the six plates.

$L = 47$ microns, the range of Po^{212} alpha particles in C-2 emulsion. However, in order to verify their value of 98.2 microns for $L = 0$, which was found by noting where the extrapolated line crossed the T axis, an experiment was conducted to determine the mean range of Po^{212} alpha particles in water by comparing the range in wet emulsion with that in dry emulsion.

Palmer and Simons claim that, if a volume v of dry emulsion swells to a volume V when a volume of water W is absorbed and if the ranges of the particle in dry emulsion, wet emulsion, and water are L, R, and x respectively, then the following relationship holds

$$\frac{1}{R} = \frac{v}{VL} + \frac{W}{Vx} \tag{18}$$

from which x may be calculated. Therefore, the experimental procedure had to yield values for v, V, R, W, and L to permit x to be found.

Careful determinations were made of the needed quantities making sure that all of them were measured under the same conditions. When these values were inserted into equation (18) a mean range of (96.6 ± 1.7) microns for Po^{212} alpha particles in liquid water was found. The two values, 98.2 and 96.6 microns, differed, but, because Palmer and Simons felt that linear extrapolation at lower energies was not necessarily valid, they adopted a mean range of 96.6 microns for Po^{212} alpha particles in liquid water at 20° C.

It was then possible to construct the range-energy curve shown in Fig. 14. This was done by converting a residual path length L in the emulsion to residual energy E by use of the range-energy relationship for C-2 emulsion. The value of T corresponding to this L was read from Fig. 13. The mean range of alpha particles of the energy E was found by subtracting this T from 96.6 microns.

Although their experiment did not yield directly the mean range of Po^{210} alpha particles in liquid water, their range-energy curve does give a mean range value for 5.3 MeV alpha particles in this medium. Palmer and Simons report a mean range value of 46 microns and give differential and integral stopping powers as 1.29 and 1.39, respectively. This value for the mean range is the highest yet reported for this energy; in fact, it is about 15% higher than the mean range reported by Aniansson. Their stopping power values are, correspondingly, the lowest reported.

It is this high range value that causes one to look for possible sources of error. Two sources of error appear to lie in determining accurately L values less than fifteen microns and in accounting for air between source and plate. The presence of air would, of course, give range values that

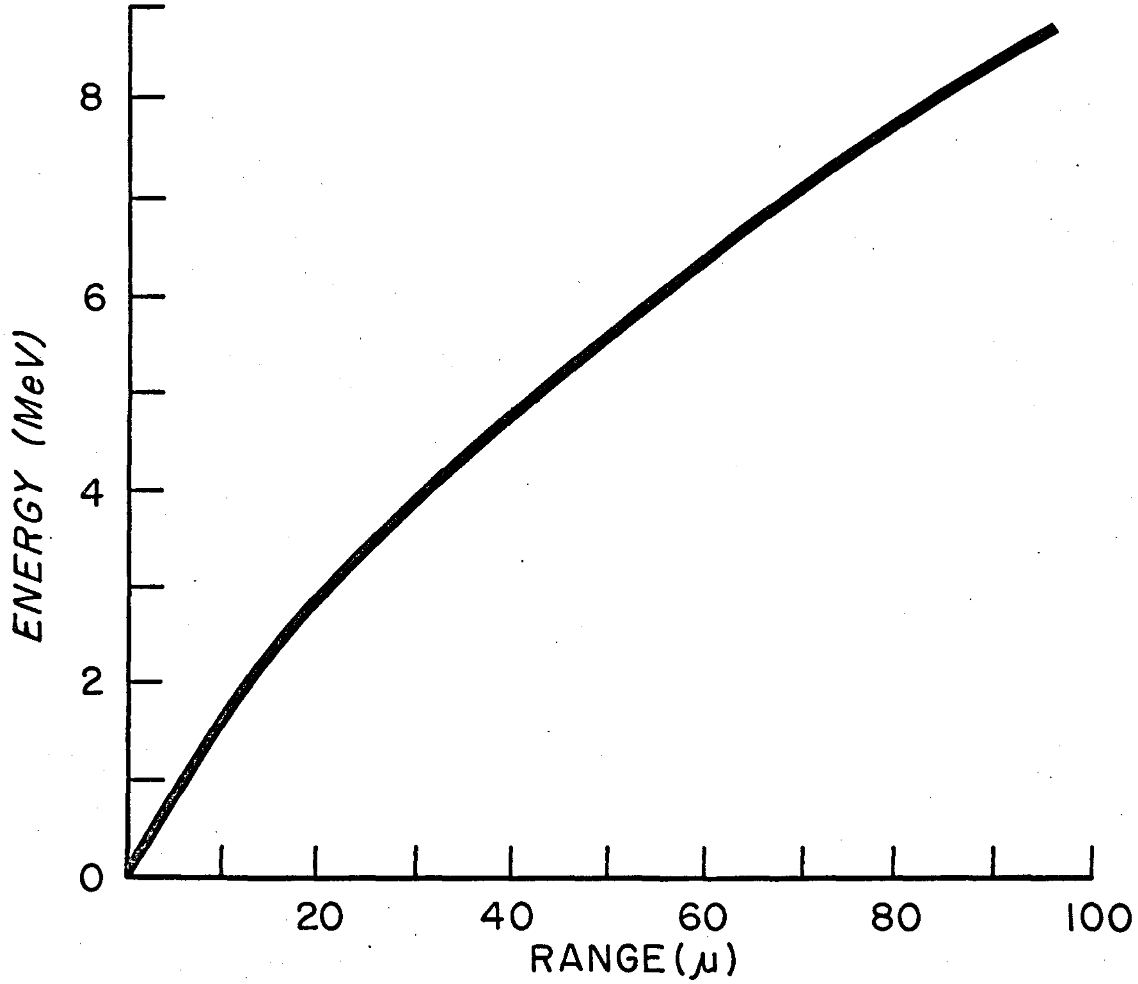

Figure 14 Range-Energy relation for alpha particles in liquid water found by Palmer and Simons.

are too small. Errors in T values due to distortion (wrinkling) of the formvar and the measurement of the small angles needed to evaluate the distances travelled in water by the alpha particles would also arise. Some errors would arise from weighing the amount of water absorbed by the emulsion and in determining the thickness of the emulsion. These last two items must be known in order to use equation (18), and the mean range of Po^{212} alpha particles in water as obtained from this equation is the most important single determination in this experiment. No doubt the use of formvar introduced some uncertainties. It is difficult to imagine a manufacturing technique that would yield a sheet of the material that did not vary in thickness by at least several microns. Furthermore, it appears that Palmer and Simons used the same value for the stopping power of the formvar to convert its thickness to an equivalent thickness of emulsion regardless of the emulsion path length L.

However, the errors associated with measuring small L values, with weighing amounts of water absorbed by the emulsion, and with measuring the thickness of the emulsion would seem to have been small, since the difference in the two independent measurements for the mean range of the Po^{212} alpha particles was less than 2%. Likewise it should not be possible to associate large errors with the use of the range-energy relationship for C-2 emulsion. In fact Rotblat states that one can rely upon the range-energy relation for C-2 emulsion to convert ranges into energy values to within 1% error in the energy.

It nevertheless appears that careful experimental evaluation of the uncertainties mentioned would be worthwhile.

VII. DISCUSSION

In Michl's work there appear to be two sources of error. These derive from the lack of sensitivity of the film and the probable presence of Bi^{210}. Either one of these could in itself render a measurement doubtful, but the possibility of having both of them present in any determination would tend to make the result quite dubious. Platzman[18] feels that it is virtually impossible today to assess the accuracy of Michl's results. This opinion is shared by the author, who believes that Michl's measurement should be regarded as approaching the true value only.

In DeCarvalho and Yagoda's measurement there exist at least two significant sources of uncertainty. One of these is associated with the fading of the latent image[33]. The other of these involves the difficulty of determining accurately the decrease in range resulting from loss of energy to the silver bromide and from the direction of the particle necessary to form a track.

The fading of the latent image is important, because the presence of the alpha tracks on the emulsion was the sole means these workers used to determine the range. Therefore, a reduction in the visibility of the tracks, especially near the end of the range, would certainly affect the accuracy of the final result.

A certain amount of energy is needed to enable the silver bromide grain in the emulsion to form a latent image. The energy to form this image must come from the Po^{210} alpha particle. This means the alpha particle must lose energy to the emulsion as well as to the water. Hence, the image is formed at the expense of the range of the alpha particle. This fact is the reason why range measurements performed with nuclear emulsion in liquid water in the manner of DeCarvalho and Yagoda would be expected to yield range values shorter than the true range. This type of error would also be present in Michl's work.

In the experiment of DeCarvalho and Yagoda an alpha particle, in order to be detected, must travel in a direction slightly below the horizontal. It is assumed that these authors made an appropriate correction.

The Ross effect[34] can appear in any experiment in which nuclear emulsions are used. This effect deals with changes in the size and positions of very small adjacent images. As the gelatin drys the images move closer together. The maximum magnitude of this effect is about 1.6% for two images originally 100 microns apart. DeCarvalho and Yagoda recognized and minimized this possible source of error by selecting a very weak developer.

There do not seem to be any significant sources of error in Aniansson's experiment except the one that apparently can be attributed to a thin insensitive layer of the crystal and the thin aluminum coating on the crystal. Aniansson's correction for this factor seems to be quite reasonable. Graphical analysis probably contributed a small amount of uncertainty. If diffusion of the polonium into the backing material had been appreciable, the junctions of the inclined lines on his graph with the horizontal lines would have been rounded.

Fano[35] points out that, when an alpha particle approaches a boundary between two media, it causes a charge to be induced on the surface approached. This induced charge causes the particle to lose slightly more energy in the neighborhood of the boundary than would normally be expected. This effect is very tiny, and it is extremely doubtful that it produced a noticeable reduction in Aniansson's range value.

Aniansson's use of mechanical apparatus, the "mikrokators," to measure the minute distances between source and crystal, could raise doubt. However, Aniansson certainly demonstrated the reproducibility possible with these devices. In a later paper[15] Aniansson reports that he measured the range of Po^{210} alpha particles in twenty-two hydrocarbons with an uncertainty of 0.012 micron. The apparatus used is essentially the same as that employed for his water measurement.

There is a fairly long list of possible sources of error that might be written for Palmer and Simons' experiment. Even the small areas of the formvar film through which alpha particles passed could have exhibited non-uniform thickness. It is possible that air was entrapped by the formvar. The conversion of formvar thickness to equivalent emulsion thickness was no doubt subject to uncertainty. Wrinkling the formvar film was probably quite difficult to avoid. The need to measure small angles to determine track lengths could have led to appreciable uncertainty. The weight of the cylinder that held their source might well have made the thickness of the water layer through which the alpha particles passed non-uniform. The experiment these men undertook was a very difficult one.

It was necessary for Palmer and Simons to use the range of Po^{212} alpha particles in liquid water to arrive at the points on their range-energy curve. Their two independent measurements yielded 98.2 microns and 96.6 microns for this range, and they chose to use the latter value in constructing the range-energy curve. Yet another worker[36] used a scintillation detector to measure this range and obtained (101 ± 2) microns. If Palmer and Simons had used this figure, their indicated range for Po^{210} would have exceeded the similar results of Wilkins and Aniansson even more. It seems that a repetition of Palmer and Simons' experiment would be well worthwhile.

There are other factors discussed in the literature that could account for uncertainties in range measurements. For example, it has been reported[37] that four alpha particle groups are emitted by Po^{212}. These are tabulated below.

Group	Energy (MeV)	Relative Intensity
α_0	8.776	10^6
α_1	9.489	40
α_2	10.417	20
α_3	10.536	170

It is possible that the presence of these higher energy groups could have affected the results of Palmer and Simons, but the relative intensities of the higher energy groups are quite small. Their detection would, of course, yield excessively high range values.

One author[38] detected a track 300 microns long on an emulsion exposed to Po^{210}. He speculated that an alpha particle had engaged in a nuclear reaction with the material on which the source was deposited and caused the emission of a proton.

Chang[39] claimed that he observed thirteen alpha particle groups in the decay of Po^{210}. Twelve of these were less energetic than 5.3 MeV. Attempts to reproduce Chang's work have failed, and the consensus of opinion seems to be that the lower energy groups resulted from the diffusion of the Po^{210} into the backing material.

Several other factors reported in the literature might be mentioned. It has long been known that hydrogen peroxide forms when alpha particles pass through liquid water. It has been shown[40] that the amount of hydrogen peroxide formed is too small to require a correction to range measurements.

The electrons ejected from the absorber atoms as a result of interactions with alpha particles are called delta rays. Lea[41] claims that nearly one-half of these delta rays possess initially energy in excess of 100 eV. Although an electron with this amount of energy can travel about one micron in liquid water, this fact should have caused no difficulty in the experiments reported here. The delta ray track in an emulsion does not begin at the point where the electron was ejected, and, when the track does become visible, its grain density is quite different from that of the alpha particle track. The use of a discriminator in work involving scintillation detection should preclude counting delta rays.

The value for the mean range of Po^{210} alpha particles in liquid water has steadily increased over the years from

Michl's low figure to the surprisingly high value reported by Palmer and Simons.

It appears that it would be fruitful to repeat the experiments of Aniansson and Palmer and Simons in order to learn why their values are so disparate.

Aniansson has demonstrated the reproducibility of his method, and, furthermore, his result is quite close to that of Wilkins. Aniansson was forced to make a correction to his result, probably because the grinding process used to make his scintillating crystal flat was too severe. Perhaps this difficulty could be eliminated by using a plastic phosphor such as NE-102, a product of Nuclear Enterprises of Canada. Aniansson himself suggested that in future work the measurement of distances between the source and crystal might be carried out by means of an interferometer. This idea is certainly desirable and feasible. The factors that should be watched with utmost care in a repetition of Palmer and Simons' work have already been enumerated earlier in this discussion.

VIII. ACKNOWLEDGEMENTS

The author wishes to express sincere appreciation to Dr. L. M. Diana for his guidance, advice, and constructive criticism throughout the entirety of this work and to the entire physics staff of the University of Richmond for helpful discussions. He wishes to express gratitude to his mother without whose constant encouragement and financial assistance this work would not have been possible.

The author thanks the libraries of the University of Richmond, Medical College of Virginia, and Virginia Institute for Scientific Research for assistance in finding and obtaining various papers needed for this account.

IX. BIBLIOGRAPHY

1. Osborne, R. V.: Nature 199 (1963) 295.
2. Tso, T. C.: Science 146 (1964) 1043.
3. Radford, E. P., and Hunt, V. R.: Science 143 (1964) 247.
4. Stannard, J. N., and Casarett, G. W.: Radiation Research Supplement 5, (Academic Press, New York, N. Y., 1964).
5. Hill, C. R.: Health Physics 9 (1963) 952.
6. DeCarvalho, H. G., and Yagoda, H.: Phys. Rev. 88 (1952) 273.
7. Cowan, F. P., Phillips, L. F., and King, R. J.: Health Physics 10 (1964) 34.
8. Zirkle, R. E.: Advances in Biological and Medical Physics, 5 (Academic Press, New York, N. Y., 1957) p. 111.
9. White, F. A., Rourke, F. M., Sheffield, J. C., Schuman, R., and Huizenga, J. R.: Phys. Rev. 109 (1958) 437.
10. Michl, W.: Sitz. Akad. Wiss. Wien 123 (1914) 1965.
11. Appleyard, R. K.: Proc. Cambridge Phil. Soc. 47 (1951) 443.
12. Appleyard, R. K.: Nature 163 (1949) 526.
13. DeCarvalho, H. G.: Phys. Rev. 78 (1950) 330.
14. Aniansson, G.: Phys. Rev. 98 (1955) 300.

15. Aniansson, G.: Trans. Roy. Inst. Technol. (Stockholm) No. 178 (1961).

16. Palmer. R. B., and Simons, H.: Proc. Phys. Soc. 74 (1959) 585.

17. Halliday, D.: Introductory Nuclear Physics (J. Wiley & Sons, New York, N. Y., 1950) p. 125.

18. Platzman, R. L.: Symposium on Radiobiology (J. Wiley & Sons, New York, N. Y., 1952) p. 139.

19. Bragg, W. H.: Studies in Radioactivity (MacMillan Science Monographs, MacMillan Publishing Co., New York, N. Y., 1912) chapter 5.

20. Brandt, W.: Health Physics 1 (1958) 13.

21. Thompson, T. J.: Thesis University of California Radiation Laboratory - 1910 (1952).

22. Gray, L. H.: Proc. Cambridge Phil. Soc. 40 (1944) 85.

23. Allison, S. K., and Warshaw, S. D.: Revs. Modern Phys. 25 (1953) 787.

24. Ellis, R. H.: Phys. Rev. 97 (1955) 1043.

25. Webb, J. H.: Phys. Rev. 74 (1948) 511.

26. Figgins, P. E. (editor): National Academy of Science - National Research Council Nuclear Science Series 3037 (1961) p. 5.

27. Yagoda, H., and Kaplan, N.: Phys. Rev. 73 (1948) 634.

28. Holloway, M. G., and Livingston, M. S.: Phys. Rev. 54 (1938) 18.

29. Philipp, K.: Z. Physik 17 (1923) 23.

30. Overman, R. T., and Clark, H. M.: Radioisotope Techniques (McGraw-Hill Book Co., New York, N. Y., 1960) p. 173.

31. Birks, J. B.: Scintillation Counters (Pergamon Press, New York, N. Y., 1960) chapter 7.

32. Rotblat, J.: Nature 167 (1951) 550.

33. Boyd, G. A.: Autoradiography in Biology and Medicine (Academic Press, New York, N. Y., 1955) p. 33.

34. Mees, C. E. K.: The Theory of the Photographing Process (The MacMillan Co., New York, N. Y., 1954) p. 1041.

35. Fano, U.: Annual Review of Nuclear Science 13 (Annual Reviews, Inc., Palo Alto, Calif., 1963) p. 56.

36. Palmer, R. B.: Proc. Phys. Soc. 78 (1961) 766.

37. Kaplan, I.: Nuclear Physics (Addison-Wesley Publishing Co., Reading, Mass., 1958) p. 265.

38. Andreev, V. N., and Sirotkin, S. M.: Nuclear Science Abstract 18:7708 (1964).

39. Chang, Y.: Phys. Rev. 69 (1946) 60.

40. Storey, W. H., and Shalek, R. J.: Bull. Am. Phys. Soc., S2, 1 (1956) 86.

41. Lea, D. E.: Actions of Radiation on Living Cells (Cambridge University Press, Cambridge, Great Britain, 1947) p. 27 and p. 82.