The β-ray Spectra of Rubidium^{86}, Strontium^{89}, Ekatantalum, and Protactinium*

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The theory of the action of a recently developed β-ray spectrograph is discussed briefly and an account is given of several improvements. In particular a baffle system of greater resolving power and a G-M counting system of greater stability are described in detail. The β-ray spectra of rubidium^{86}, strontium^{89}, ekatantalum, and protactinium were measured. The use of thin collodion for the backing of sources and windows of counter permitted accurate measurements down to 50 kev. The upper energy end points were found to be: rubidium^{86}—1.60±.03 Mev; strontium^{89}—1.32±0.03 Mev; ekatantalum—230 kev. The Fermi and K-U plots of Rb^{86} and Sr^{89} are given.

A NEW solenoidal type β-ray spectrograph has been described recently by Witcher and co-workers[1] which has as its greatest advantage a good combination of resolving power and efficiency. The relatively high value of the efficiency (1 percent of the total number of particles emitted can be detected) has been found extremely useful for the study of many artificially radioactive substances of small intensities. One of the major advantages of this instrument is that radioactive sources covering fairly large circular areas may be utilized without decreasing the resolution seriously. This permits the use of extremely thin sources and minimizes dispersion due to scattering within the source material.

As originally constructed the resolution of the instrument (half-width of line divided by $H\rho=5$ percent) was satisfactory for continuous spectra, but the complete resolution of many line spectra such as ekatantalum required a baffle system of greater resolving power. Further work has therefore been done on this instrument to increase its usefulness by designing an alternate baffle system which could be substituted when stronger sources were available and when it was desirable to study a line spectrum in more detail. The new baffle system, besides reducing the spread of the beam, introduces some improvements in the geometry of the spectrograph, i.e., it enables the source, slits, and counter to be more accurately lined up and centered.

This type of spectrograph requires a special form of Geiger-Mueller counter which, in general, does not have the characteristics which make the ordinary counter a satisfactory detector for β-rays. Further work has therefore been done to improve the counter characteristics and to make it useful for work at low energies.

THE BAFFLE SYSTEM AND THE RESOLUTION OF THE SPECTROGRAPH

In this spectrograph a homogeneous magnetic field is used to focus the electrons. The field is produced by a solenoid and that portion of the solenoidal field is utilized which is homogeneous to within 1 percent. The path of an electron with an initial momentum making an angle α with the magnetic field is a helix of radius $(mcv/eH)\sin\alpha$ and pitch $(2\pi mcv/eH)\cos\alpha$, whose axis is parallel to the magnetic field. An electron emerging from a point source on the solenoidal axis will return to the axis after completing one revolution, the distance between the source and the point where it returns being equal to the pitch of the helix. If z denotes the displacement of the electron in the direction of the axis (the source taken as origin) and R the displacement of the electron from the axis in a direction perpendicular to it, then $R=(2mcv/eH)\cdot\sin\alpha\cdot\sin(eHz/2mcv\cos\alpha$. If we write $D=2mcv/eH$, then

$$R=D\cdot\sin\alpha\cdot\sin(z/D\cos\alpha).$$

For fixed z_1 and D_1 (at a plane distant z_1 from the source and for a fixed ratio of electron velocity to field strength), R will vary with the angle of emission α, and the mathematical condition that will give a point of focus at $R=R_1$, is that $dR/d\alpha=0$. The symmetry of the field about the

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[1] Witcher, O'Conor, Haggstrom, and Dunning, Phys. Rev. **55**, 1135A (1939); Witcher, Phys. Rev. **60**, 32 (1941).

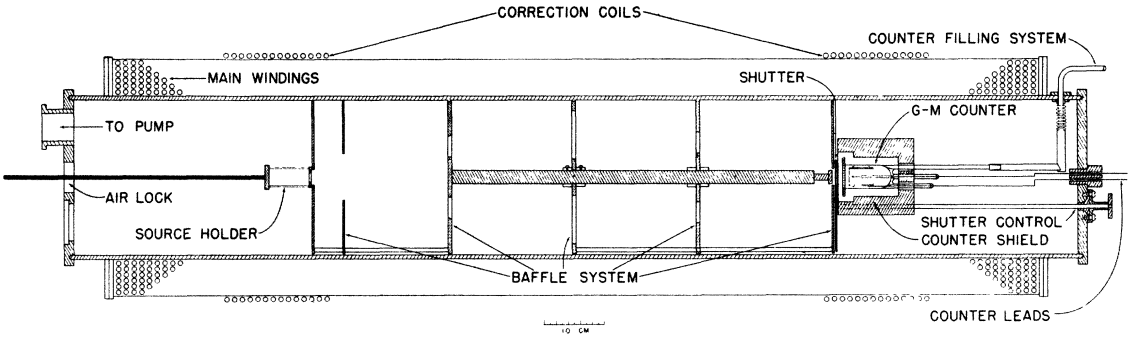

FIG. 1. Solenoidal β-ray spectrograph.

axis then leads to the focusing of all electrons with the same velocity and an angle of emergence α in a ring of radius R_1, in the plane $z=z_1$. A point focus is obtained if R_1 vanishes concurrently with $dR/d\alpha$. This requires that α be equal to 0, and since in this case there is no separation of the different velocities the focal ring must be of finite radius.

A diaphragm situated at a distance z_1 from the source and having an annular slit of radius R_1, will permit the focused electrons to pass through the instrument. A system consisting of a source and one such diaphragm would allow other electrons besides those which are focused at the slit to pass through, and an additional diaphragm is necessary to determine completely the helix within the limits set by the finite widths of the slits, if it can be assumed that no electron makes more than one revolution. By having a rod of material situated along the axis for most of the length of the spectrograph, the possibility of an electron making more than one revolution is ruled out.

FIRST BAFFLE SYSTEM

While only two slits are necessary, the baffle system originally designed contained a large number of slits which outlined the paths taken by the electrons of maximum and minimum velocity and angle of emergence that were allowed to pass through the spectrograph. An inner series of disks which are supported on a rod coincident with the axis outlines the curve $7.16 \sin (z\pi/90)$. The brass wall of the solenoid at its central point together with the inner diameters of two rings on either side of the center lies on the sine curve $12.5 \sin (z\pi/90)$. This baffle system, though more complicated than necessary and especially difficult to study exactly when computations of the average efficiency of extended sources are being made, has been designed to reduce scattering. There has been no evidence of any error due to scattering when this baffle system has been used. It was constructed so that the electrons were focused 86 cm distant from the source; the inner radius of the annular slit at that plane was 1 cm, and the outer radius was 1.5 cm. The value of D for which the efficiency of the spectrograph is a maximum is $94.7/\pi$. The emergent angles of the electrons which constitute the beam vary between 12° 36′ and 23° 30′. The solid angle at maximum efficiency is 1 percent and the half-width of the resolution curve (the plot of efficiency against D) is 5 percent. The choice of this geometry which leads to a good combination of resolution and efficiency under the practical restrictions of the size and power of the available solenoid has been discussed previously by Witcher.[1] He has also discussed the theoretical and experimental resolution curves for an extended circular source 2 cm in diameter.

SECOND BAFFLE SYSTEM

In the new baffle system the number of defining slits has been reduced to three. Two additional diaphragms whose slits are so large that they do not intercept the beam have been introduced to eliminate scattering. The annular ring slits are carefully centered with respect to the axis by means of bars connecting the outside ring to the inner disk. The β-ray source is in the form of a circular area 0.5 cm in diameter centered on the

axis of the instrument. The Geiger-Mueller counter has an open area greater than that of the last slit and is situated 1 cm behind the diaphragm where the electrons come to a focus.The maximum solid angle is obtained for $D=30$ and for this value of D the angles α range from 8.9° to 18.7°. The half-value of the resolution curve divided by the $H\rho$ value at the peak is 2.5 percent. Figure 1 shows the spectrograph with the new baffle system in place.

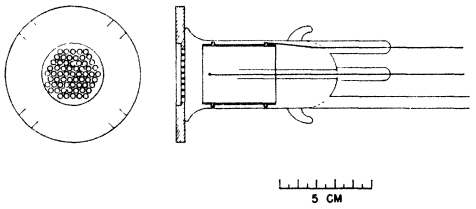

FIG. 2. G-M counter for use with spectrograph.

SOURCE

While the geometry of the instrument determines the efficiency for any point on an extended source, the intensity for any given source is a function of the diameter of the source. The resolution becomes greater as the diameter of the source is decreased. Sources up to 2 cm in diameter may be used.

The source, which is made by dropping the radioactive material in the center of a film of collodion of the order of 0.7-μ thickness, supported by an aluminum ring, is placed in position by fitting the ring into a diaphragm which has a central hole. Scattering from the collodion film support should be negligible for electrons above 30 kev.[2]

To minimize the small residual scattering that might take place by multiple scattering from the brass wall of the instrument, a sheet of fiber board 1/64″ thick is rolled to cover the wall of the solenoid between the source and the final slit.

GEIGER-MUELLER COUNTER

Inasmuch as the solenoidal spectrograph focuses the electrons in an annular ring, the electrons must enter the counter at one end and the cylindrical electrode must therefore be at least 3 cm in diameter. To keep the background of the counter low, the length of the cylinder was made relatively small (4 cm) and the central wire was covered by glass tubing leaving an active region of only 2-cm length exposed. The effect of this restricted geometry is that the pulses generated are small. Previously, the counter window was mounted on a metal grill and it was found necessary to apply a potential intermediate between that on the central wire and the cylindrical electrode to prevent spontaneous discharges through the counter to the grill. The counter characteristics have been improved by substituting a plastic grill with 60 percent open area ($\frac{1}{8}$″ holes) which results in increased stability and a wider plateau.

0.5-mil thick Cellophane windows are satisfactory for use down to 1900 $H\rho$ and approximate corrections can be made down to 800 $H\rho$. For accurate spectra below this energy, thinner collodion windows were used to close the counter. The film is made by dropping a half-and-half solution of collodion and amyl acetate on distilled water. The size of the holes in the grill determines the strength, i.e., the thickness necessary to withstand the pressure in the counter. In order to obtain a film that was tight over the comparatively large area of 10 cm^2, a film made of 12 superimposed layers of original film was necessary. Since each film is approximately 0.08μ thick the total thickness is about 1μ.

The collodion film when laid over the open area of the grill extends only about 2 mm beyond the open area and the pressure in the counter is sufficient to seal the thin film to the grill. A thin film of stopcock grease provides a satisfactory seal between the ground edge of the counter and the ground plastic grill, the two being held together by means of rubber bands that pass over hooks on counter and grill. A counter gas mixture of 9 cm of argon and 1 cm of alcohol has been found satisfactory, a five-gallon ballast flask being connected to the system to stabilize the pressure over a long period against possible porosity in the thin films. The operating characteristics of the counter are now very satisfactory with the threshold at 800 volts and a plateau of more than 150 volts. The counter is shown in Fig. 2.

Such a counter closed with a thin window of approximately 1-μ thickness of collodion will give

[2] A. Flammersfeld, Zeits. f. Physik **112**, 727 (1939).

accurate spectra for electrons ranging down to 50 kev without appreciable corrections.[2] Since the collodion films will vary somewhat in thickness, it is not possible to extrapolate accurately to zero thickness by placing one, two, and three additional thicknesses in the path of the beam by means of a shutter. It is therefore necessary to determine the region in which the spectra are not distorted by absorption in another way. This is done by placing one additional film in the path of the beam, varying the energy of the beam, and determining the point at which the intensity of the beam is not reduced by the film. For all energies above this point, the spectra are accurate.

COUNTER STANDARDIZATION

As a check on the sensitivity of the counter, a source of uranium oxide electroplated on copper is placed in one of the shutter windows. The use of this β-ray source has been found preferable to a radium γ-ray standard outside the instrument, especially when it is necessary to compare the operation of the counter over long periods of time. It is thus possible to measure the decay of a spectrum with accuracy. The counter is completely shielded from the uranium when the shutter is turned through 180°, and the background of the counter, which is about 20 per minute, is not affected by the presence of the uranium in the spectrograph.

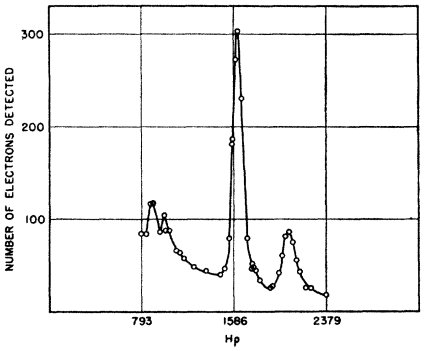

FIG. 3. Uncorrected spectrum of $_{91}$ekatantalum^{233}.

THE SPECTRA OF EKATANTALUM AND PROTACTINIUM

A. V. Grosse, E. T. Booth, and J. R. Dunning[3] have reported on the preparation of a β-emitting isotope of the naturally radioactive protactinium. Thorium in the form of a solution freed from the naturally radioactive protactinium is irradiated by slow neutrons from the Columbia cyclotron. The irradiated thorium shows a strong β-activity having a half-life of 25 minutes which could be explained by the formation of Th^{233} by resonance capture in an (n, γ) reaction which disintegrates by β-emission into an isotope of protactinium—$_{91}$ekatantalum^{233}.

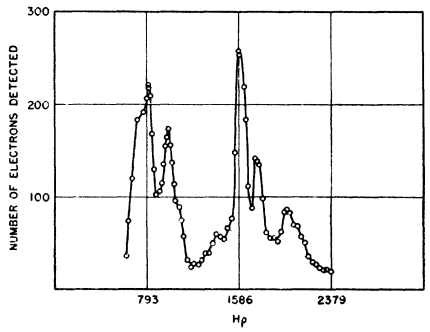

FIG. 4. Uncorrected spectrum of $_{91}$protactinium^{231}.

The chemical separation of ekatantalum from the irradiated thorium gives a product which is again β-active, tests on an ionization chamber giving the half-life as 27.4±0.3 days. The decay of this substance by the emission of an electron would give rise to a new nucleus, presumably an unstable isotope of uranium, $_{92}$U^{233}. To establish definitely that the ekatantalum emitted nuclear electrons, the β-ray spectrum was examined.

The source after preparation by Dr. Grosse was placed over a 2-cm diameter area on a thin collodion film about 0.7μ thick. The layer of material was very thin so as to produce no effect from back scattering above the energy at which the accuracy of the instrument is limited by absorption in the Cellophane counter window. The comparison of the spectra obtained from two sources whose thickness and backing were different, established that errors due to back scattering were not apparent in the region of the spectrum reported on. The counter window was of 0.5-mil Cellophane.

The spectrum of ekatantalum shown in Fig. 5 has been obtained from the experimental results plotted in Fig. 3 by correcting for decay, division of ordinates by H to allow for the varying resolu-

[3] A. V. Grosse, E. T. Booth, and J. R. Dunning, Phys. Rev. **59**, 322 (1941); G. T. Seaborg, J. W. Gofman, and J. W. Kennedy, Phys. Rev. **59**, 321 (1941).

tion of the instrument, and by correcting for absorption in the Cellophane window. This last is done by placing one, two, and three additional layers of Cellophane in front of the counter window and extrapolating to zero thickness. The correction curve as a function of the energy is shown in Fig. 6 and gives the factor by which the actual intensity must be multiplied to yield a corrected spectrum. These corrections have been applied down to $H\rho = 800$ although it is difficult

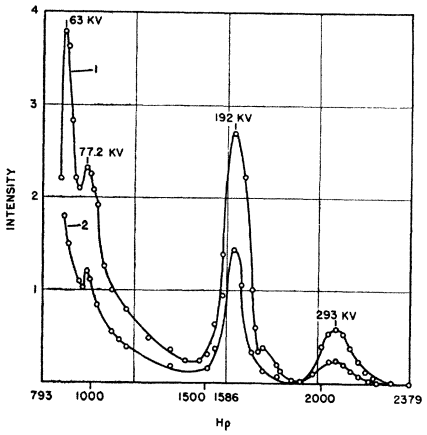

FIG. 5. Corrected β-ray spectrum of $_{91}$ekatantalum^{233}.

to establish just where the approximation becomes to poor to apply.

The spectrum of ekatantalum contains at least four γ-ray conversion lines at 63 kev, 77.2 kev, 192 kev, and 293 kev. Two of these lines, those at 192 kev and 293 kev, appear to be complexes of unresolved lines. The spectrum above 230 kev seems to consist of conversion lines alone; the lines below 230 kev appear to be superimposed on a continuous background. The spectrum has been followed with an accuracy of 2 percent on the peaks and the points have been taken too close together to admit of any undiscovered line of any intensity in the portion of the continuous background between 1000 and 1500 $H\rho$. If we rule out the possibility of this portion of the curve having been formed by a large number of lines all of small intensity, it can be concluded that the ekatantalum emits nuclear electrons, and that a uranium isotope $_{92}U^{233}$ is formed. The maximum energy end point is masked by the line at 192 kev. By subtracting this line from the continuous spectrum, the end point of the continuous spectrum appears to lie in the neighborhood of 230 kev.

A source of naturally radioactive protactinium $_{91}Pa^{231}$ prepared by Dr. Grosse was examined in order to compare it with its isotope and thereby establish independently the purity of the ekatantalum. Although Figs. 3 and 4 (plot of number of electrons detected against current or corresponding $H\rho$) show a similarity in the positions of some of the most intense conversion lines, the relative intensities and positions of the lines differ markedly. The spectrum of protactinium obtained checks as far as the resolved lines are concerned with the spectrum obtained by L. Meitner.[4]

To check further the purity of the sources and the modes of decay, both the ekatantalum and protactinium were remeasured after 27 days. The curve 2 of Fig. 5 shows that all parts of the ekatantalum curve decay at the same rate. The half-life of the ekatantalum measured on the spectrograph by taking an average of the decay at various points along the curve was 26.0 ± 0.5 days which checks well with the observed decay period for the bulk material. Since there was no

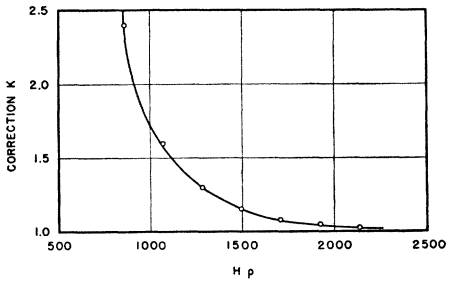

FIG. 6. Correction factor for absorption in 0.5-mil Cellophane window.

observable change in the shape of the ekatantalum curve, it was not possible to make any discovery as to the mode of decay of the product uranium nucleus. The natural protactinium showed no decay at the second measurement as is to be expected if the sample is pure. The spectra shown in Figs. 3 and 4 are the experimental plots of the electron intensity against the current or corresponding $H\rho$.

[4] L. Meitner, Zeits. f. Physik **50**, 15 (1928).

THE SPECTRUM OF RUBIDIUM[86]

Helmholz, Pecher, and Stout[5] have succeeded in producing a radioactive product with a period of 19.5 days by the bombardment of Sr with 16-Mev deuterons by the (d, α) reaction. They definitely established this product to be rubidium. By comparing their results with those of Snell[6] (who found an 18-min. and 18-day period after the bombardment of Rb^{85} and Rb^{87} with slow neutrons giving rise to the isotopes Rb^{86} and Rb^{88}), they concluded that the radioactive Rb was Rb^{86}, since this is the only Rb of the two that could be formed by the Sr (d, α) reaction, Sr^{88} being the heaviest Sr.

Three microcuries of Rb^{86} were prepared in the form of Rb Cl for examination in the spectrograph. The solution was dropped onto a collodion film about 0.7μ thick resulting in an invisible layer of Rb Cl when dry. The spectrum was first studied with a counter window of 0.4-mil thick Pliofilm, and a careful study made of the upper energy region. For the low energy region a counter window of approximately 1-μ thick collodion was employed. The curves from these two measurements were reduced to the same intensity

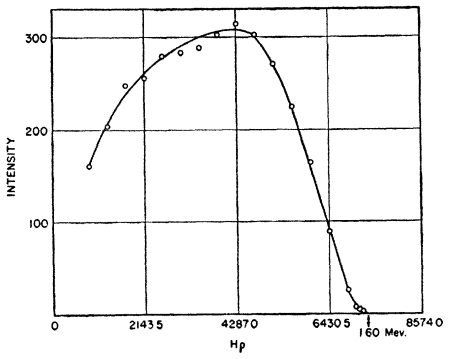

FIG. 7. β-ray spectrum of rubidium[86].

at a point above the energy at which absorption occurs in the Pliofilm window.

Figure 7 shows the corrected spectrum of Rb^{86}. It is seen that the upper end point of the experimental spectrum lies at $H\rho = 7288$. After correcting this upper energy end point for the finite spread of the resolution curve the value 1.60 Mev ±.03 is obtained as the true end point. The value obtained by Helmholz, Pecher, and Stout from absorption measurements is 1.56 Mev calculated from Feather's rule.

THE SPECTRUM OF STRONTIUM[89]

Sr^{89} is produced by bombarding Sr^{88} with deuterons in the (d, p) reaction. A source of 2.78 microcuries of this Sr^{89} was prepared for study.

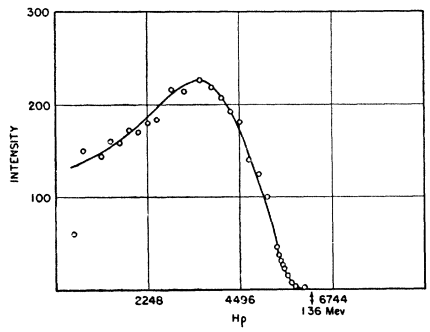

FIG. 8. β-ray spectrum of strontium[89].

The solution was prepared in the lactate form and after placing a drop of the concentrated solution on the center of the 0.7-μ thick collodion film the source after drying was invisible. There is little probability that an error in the spectrum has occurred due to back scattering from the source.

As in the case of rubidium the spectrum was first examined with a 0.4-mil Pliofilm window closing the counter and a second run was taken at the low energy end with a collodion film of approximately 1-μ thickness on the counter. The source strength was such as to give a counting rate of only 144 counts per minute at the peak. The counting period was such as to give an accuracy of ±2 percent on most of the points of the curve except at the upper and lower energy end of the spectrum. Figure 8 shows the spectrum thus obtained. The upper energy end point when corrected for the finite spread of the resolution curve is 1.32 Mev ±0.03. The half-life of this substance is 55 days.

[5] A. C. Helmholz, C. Pecher, P. Stout, Phys. Rev. **59**, 902 (1941).
[6] A. H. Snell, Phys. Rev. **52**, 1007 (1937).

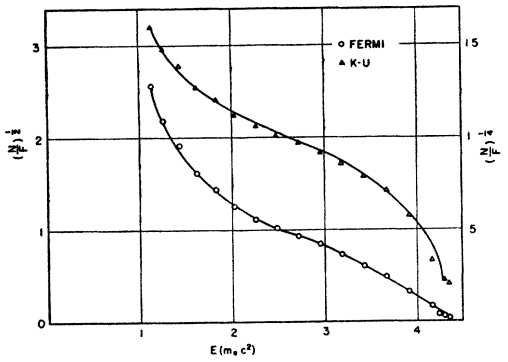

FIG. 9. Fermi and K-U plots for rubidium[86].

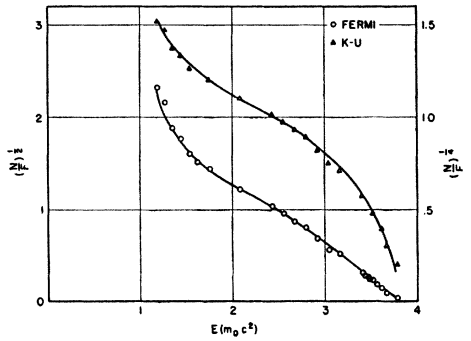

FIG. 10. Fermi and K-U plots for strontium[89].

THE SHAPE OF THE SPECTRA

Since both rubidium[86] and strontium[89] lie on the second forbidden Sargent curve, it is not to be expected that the Fermi plot for these elements be straight lines. To facilitate comparison between these and other doubly forbidden spectra such as P^{32}, the Fermi and K-U plots of Rb^{86} and Sr^{89} are shown, in Figs. 9 and 10. N represents the intensity and $F(\eta, Z)$ is the Fermi function for low Z

$$F(\eta, Z)=\eta^2(2\pi y/1-e^{-2\pi y});\quad y=Z[(1+\eta^2)^{\frac{1}{2}}/137\eta].$$

It is seen that toward the upper energy end points of both of these spectra the Fermi plot is very nearly a straight line but at low energies it is convex toward the axis and lies above the extended straight line.

I wish to express my sincere thanks to Professor J. R. Dunning and Dr. E. T. Booth for their constant suggestions and advice during the course of this work, and to my co-workers, Dr. C. M. Witcher and J. O'Conor, S. J., for their cooperation and assistance in the research. I am indebted to Dr. A. V. Grosse and Dr. J. M. Kenny for their aid in procuring radioactive sources.