"THE INVESTIGATION OF RADIATIONS OF LOW SPECIFIC ACTIVITIES AND COGNATE PROBLEMS"

by

Henry W. Wilson,

Department of Natural Philosophy,

University of Glasgow.

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789 East Eisenhower Parkway
P.O. Box 1346
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CONTENTS.

PREFACE

This thesis describes the work of the writer in the field of low intensity beta- and gamma-ray spectroscopy. This work was carried out between October 1947 and September 1951. Part 1A contains a theoretical introduction to the subject and part 1B a discussion of the experimental techniques used in this field. The remainder of the thesis, parts 2,3,4,5 and the appendix, is concerned with the investigations carried out by the writer and the conclusions reached as a result of these investigations.

The introduction is rather longer than is perhaps usual. This is due in part to the wide scope of the investigations and although the matter included in the discussion of the different methods of analyzing beta- and gamma- radiations is not new, it represents an analysis by the writer with special regard to his own field of weak activities. Moreover it should be noted that the introduction contains a certain amount of original work (the theory of the "Wall Effect", experiments on calibration) which seemed to fit most naturally into part 1.

The study of the unstable nickel isotopes started in the proposal of Dr. S.C.Curran to investigate the reported low energy positron emission of Ni^{59}. This earlier work, done in collaboration with Dr. Curran, resulted in the discovery of the low energy negatron emitter Ni^{63}. Later the writer resumed, on his own initiative, the investigation of nickel to elucidate the mode of decay and period of Ni^{59}. In this way the capture decay of Ni^{59} was discovered with no evidence of positron emission. Regarding the work on Hg^{203}, the investigations were extended by the suggestion of Dr. Curran that the integrating properties of the proportional counter would prove useful in elucidating the decay. The writer was wholly

responsible for undertaking and prosecuting the work on samarium and europium, on the "Wall Effect", Part 1B, Sections 8,9 and 10 and the experiments on calibration, Part 1B, Section 11. The work on rubidium and neodymium was part of a general investigation of low intensity of sources carried out in the department. The work described in the appendix was carried out at the suggestion of Dr. J.B. Birks.

At a late stage in the examination of the samarium and europium activities the writer asked Mr. G.M. Lewis to collaborate in coincidence studies (Part 4, Section 6) since he had suitable scintillation counting equipment. Also the writer was assisted by Mr. D. Dixon in the later stages of the work on samarium and europium andin the work on rubidium. The remaining investigations described in Parts 1B, 2, 3, 4 and in the appendix were carried out entirely by the writer.

References in the text are given in the style now used by British journals of physics, viz. by giving the name or names of the author or authors followed by the date of publication. The full references are given at the end.

Finally I should like to thank Professor Dee for his kind interest and support in the course of this work, Dr. Curran for his guidance and for many helpful suggestions and Mr. J.T.Lloyd and the workshop staff for their assistance in the construction of apparatus.

H.W.W.

PART 1. INTRODUCTION.

A. Theoretical.

1. Historical Difficulties in Beta-Decay.

The study of beta-rays emitted by radioactive substances has been actively and continuously carried out since the discovery of radioactivity in the closing years of the last century. An immense amount of experimental data has resulted from this study accompanied by equally extensive theoretical investigations and yet it is only within recent years, since the war, in fact, that the theory of beta-decay about to be described briefly has become so trusted that disagreement of experimental results with theory makes the data suspect rather than the theory (at any rate, when the beta-transition is known to be "allowed").

The phenomenon of beta-decay raised several theoretical problems of considerable difficulty especially after Chadwick's results of 1914. Earlier, von Baeyer and Hahn (1910), by magnetic analysis of beta-rays from a radioactive source, showed that the beta-rays possessed homogeneous electron groups or "lines". Von Baeyer, Hahn and Meitner (1911, 1912) and Rutherford and Robinson (1913) using the more accurate 180^{0} focusing spectrometer of Danysz, discovered and investigated many such "lines". Between 1910 and 1914, only this line structure was investigated or, apparently, expected.

However in the latter year, Chadwick (1914) first showed that the beta-spectrum of RaB possessed, in addition to a prominent line spectrum, a continuum of beta-particles having a well defined upper energy limit and found that the number of particles in the continuum far exceeded the number in the monoenergetic groups. The difficulty which this discovery

raised, and others which became apparent as time went on, occupied the attention of many physicists for almost twenty years and were as follows.

I. Let us consider the decay of an isotope A into another isotope B, with the emission of a negative electron, or negatron, i.e. $A \rightarrow B + \beta^-$. We will show later that this is energetically possible, if the atomic mass of A is greater than the atomic mass of B, and the energy released is given by the difference of these masses, since, by Einstein's energy relation, mass and energy are equivalent and, in a nuclear reaction, are interchangeable. It is found that the energy corresponding to this mass difference is equal (within experimental error) to the upper limit of the beta-spectrum. However by far the greater number of the emitted electrons have an energy less than this, and, in fact, the average energy is nearly always less than half the maximum energy. Evidently the electrons must lose energy somehow, or else the law of conservation of energy does not hold in this case. Although the latter possibility was seriously suggested by so great a physicist as Bohr, there was naturally a great reluctance to accept this explanation. Ellis and Wooster (1927) carried out their calorimeter experiment in an endeavour to find evidence of the former explanation. A RaE source was placed in a calorimeter which was surrounded by 1.2mm of lead which was sufficient to stop all ionizing radiations emitted by the source. The energy lost by the source and trapped by the calorimeter corresponded to an average energy of 0.35 ± 0.04 MeV per disintegration. The upper energy limit of the beta-spectrum was 1.2 MeV while the mean energy was estimated to be 0.34 MeV, agreeing with the value found by Ellis and Wooster. As a result we can say that, if the law of conservation of energy holds, the energy must be carried off by a radiation which is not trapped by the calorimeter.

II. Another difficulty was that the law of conservation of angular momentum did not appear to hold. An electron has an intrinsic spin $\frac{1}{2}$. In beta-decay an isotope decays to a neighbouring isobar, i.e. its atomic weight, considered approximately as a whole number, remains unaltered. Now it is to be expected theoretically and is known experimentally that a nucleus of even mass number will have a spin value which is an integer, and is generally zero. If and when beta-decay takes place, the nucleus will be left still with integral spin, as its mass number is unaltered. On the other hand the electron can carry off only half integral total angular momentum. This would leave the product nucleus with half-integer spin which as we have seen is not the case. A similar argument applies to the case of a nucleus of odd mass number, which possesses a half-integral spin value, as does the product nucleus. We will postpone further discussion of this difficulty.

III. If we consider the process of beta-decay as being the result of the process

$$n \rightarrow p + \beta^-$$

we are faced with an apparent violation of the law of conservation of statistics, since all three particles have Fermi statistics.

2. The Neutrino Hypothesis.

To remove these difficulties, Pauli postulated the existence of a new elementary particle, now called the "neutrino" (Italian: "the small neutral one") which is produced at the same instant as the beta-particle. It must be light, possessing no charge, have spin $\frac{1}{2}$ and Fermi statistics. Such a particle could carry off the surplus energy whenever the electron left the nucleus with less than the maximum energy. That it should have no charge is necessary in order to conserve charge in the process of beta-decay. Its spin of $\frac{1}{2}$ allows it to carry away the half-integral spin

which caused the second difficulty. It requires to be light in order to explain the agreement of the maximum energy of the beta-decay with the mass difference between the parent and product atoms which had been measured in some cases. We shall discuss experiments proving the lightness of the neutrino later, but it may be said here that the evidence is in favour of it having less than 1/300th. of the mass of an electron. Also direct experimental evidence of the existence of a neutrino will be given. It is easy now to understand the result of Ellis and Wooster, assuming the existence of this light neutral particle. Its interaction with matter is extremely small, Nahmias having placed the upper limit of probability of capture by a nucleus at one primary encounter in 300,000km of air. This corresponds to a cross-section of $\sim 10^{-30}$ cm.^{2} per atom. This means that practically always, the neutrino would go right through the calorimeter. A rough calculation shows that it would capture less than one in 10^8 neutrinos emitted.

3. The Energy Available for Decay.

Using this suggestion of Pauli's, Fermi (1934) formulated the first detailed theory of the process. He suggested that radio active decay took place as follows:-

Negatron emission:-

$$n \rightarrow p + \beta^- + \nu$$

where ν represents the neutrino. For theoretical convenience, one usually assumes that a neutrino is absorbed from a state of negative energy and that an "anti-neutrino" is emitted.

Positron emission:-

$$p \rightarrow n + \beta^+ + \nu$$

In this case a neutrino is emitted. It will be useful at this point to work out the mass-energy relations governing beta-decay.

Negatron emissions:-

The conversion of a neutron into a proton with the emission of a negatron and a neutrino is possible if

$$M_n(Z)^A - M_n(Z+1)^A \geq m + \mu$$

where $M_n(Z)^A$, $M_n(Z+1)^A$ are the nuclear masses of the parent and product nuclei respectively, m is the mass of an electron and μ is the mass of the neutrino, which may be zero.

Atomic, rather than nuclear, masses are quoted in tables however. These include the mass of the orbital electrons. Substituting we have

$$M_a(Z)^A - Zm - M_a(Z+1)^A + (Z+1)m \geq m + \mu$$

$$\therefore \quad M_a(Z)^A - M_a(Z+1)^A \geq \mu$$

The energy which is available for decay is given by

$$E = M_a(Z)^A - M_a(Z+1)^A - \mu$$

Positron emission:-

This is energetically possible if

$$M_a(Z+1)^A - M_a(Z)^A \geq 2m + \mu$$

as can be shown by an argument analagous to that for negatron emission. Here the energy available for decay is

$$E = M_a(Z+1)^A - M_a(Z)^A - 2m - \mu$$

In this case, there must be sufficient energy available to create two electrons. If there is not sufficient energy for this purpose a competing process will take place. This is usually called

Electron capture:-

Electron capture:-

Instead of emitting a positron it is possible for a nucleus to absorb an electron from the K-shell. The energy balance required for such a process is obtained in a similar fashion and is found to be

$$M_a(Z)^A - M_a(Z-1)^A \geq \mu + E_K$$

where E_K is the binding energy of the K electron. The right hand side is clearly much smaller than for the positron case and hence, when positron emission is impossible, electron capture may still be possible. If K capture is not possible, L,M,$\cdots$ capture is, in principle at least, so that the term E_K may in fact be omitted. However, as the energy available exceeds that necessary for positron emission, the latter process quickly becomes the more favourable one, since the probability of K-capture depends on the probability of a K electron finding itself in the nucleus, while for positron emission, the probability of sufficient energy being concentrated on one proton in the nucleus becomes much greater, as the energy available increases.

As a result of these considerations we believe that a pair of neighbouring naturally occurring isobars cannot both be stable against beta-decay and that the heavier isobars will decay into the lighter by one of the processes described above, However due to large spin changes the transition may be so highly "forbidden", i.e. the half-life may be so long, that it may not have been detected. Many such pairs (and even trios) of isobars are known to occur naturally, e.g.

$_{18}A^{40}$, $_{19}K^{40}$, $_{20}Ca^{40}$; $_{37}Rb^{87}$, $_{38}Sr^{87}$; $_{48}Cd^{113}$, $_{49}In^{113}$; $_{49}In^{115}$, $_{50}Sn^{115}$; $_{51}Sb^{123}$, $_{52}Te^{123}$; $_{70}Yb^{176}$, $_{71}Lu^{176}$, $_{72}Hf^{176}$; $_{75}Re^{187}$, $_{76}Os^{187}$. In all these cases, except for the Sb^{123} - Te^{123} pair, one of the isobars has been reported to be radioactive. However in some cases, the evidence is not very satisfactory

as yet, especially in the cases of Cd^{113} , In^{113} ; In^{115} , Sn^{115} ; Re^{187} , Os^{187}. The proportional counter technique employed by the writer will probably be used in the examination of these isobars at some future date.

4. The Fermi Theory of Beta-Disintegration.

This theory was first given in 1934 by Fermi (1934). An excellent summary of the theory, its application to experiment, and of experimental work up to the time of writing, was given by Konopinski (1943). The subject is also treated by many others, e.g. Bethe (1947), Moon (1949), Fermi (1950), Wu (1950) etc., to all of whom the writer records his indebtedness. It is not intended to derive the expression for the electron distribution but rather to indicate how Fermi tackled the problem, to state the result and to discuss it from the experimentalist's point of view.

Fermi formulated his theory on the assumption that the neutrino did exist and introduced a new interaction between the nucleon responsible for the β^+ or β^- decay, the electron and the neutrino. This interaction was chosen in analogy with the interaction between charges and the electromagnetic field in the production of photons from the atomic electrons However the beta-emission case is somewhat more difficult since an extra particle is involved.

If the Hamiltonian of the interaction between the nucleon and the electron-neutrino field is H, the number of beta-transitions per second is $\frac{2\pi}{\hbar} \left| \int \psi_{fin}^{*} H \psi_{in} \, d\tau \right|^2 . \rho(E)$

where $\rho(E)$ = the number of final states of the system per unit energy interval. ψ_{in} = initial state of the system = u_{in} = initial state of the nucleon. $\psi_{fin} = u_{fin} \times \psi_{elec} \times \phi_n$ = final state of the system. u_{fin} = final state of the nucleon. ψ_{elec} = final state of the electron. ϕ_n =

= final state of the neutrino.

Fermi assumed that H was given by

$$\int \psi^*_{fin} H \psi_{in}\, d\tau = g \int u^*_{fin} \psi^*_{elec} \phi_n u_{in}\, d\tau$$

This neglects relativistic corrections which are important only if the heavy nucleon has high velocity. The wave functions are to be evaluated at the position of the nucleon. "g" determines the strength of the interaction and is a constant which can only be determined experimentally. Its value is approximately 10^{-48} - 10^{-49} gm cm^5 sec^{-2}. It is usual in the final form of the equation to substitute "G" where $G = (g/mc^2)(\hbar/mc)^{-3}$. ψ^* corresponds to the emission of an electron and ϕ_n to the absorption of a neutrino, which is equivalent to the emission of an anti-neutrino.

The form of interaction chosen by Fermi is known as the "Polar Vector Interaction". Other forms are possible, obeying the criterion of relativistic invariance. A list of these forms follows:

1. Scalar $S=(\psi^* \beta \phi)(u^*_{fin} \beta u_{in})$
2. Polar Vector $V=(\psi^* \phi)(u^*_{fin} u_{in}) - (\psi^* \underline{\alpha} \phi)(u^*_{fin} \underline{\alpha} u_{in})$.
3. Tensor $T=(\psi^* \beta \underline{\sigma} \phi)(u^*_{fin} \beta \underline{\sigma} u_{in}) + (\psi^* \beta \underline{\alpha} \phi)(u^*_{fin} \beta \underline{\alpha} u_{in})$.
4. Axial Tensor $A=(\psi^* \underline{\sigma} \phi)(u^*_{fin} \underline{\sigma} u_{in}) - (\psi^* \gamma_5 \phi)(u^*_{fin} \gamma_5 u_{in})$
5. Pseudo Scalar $P=(\psi^* \beta \gamma_5 \phi)(u^*_{fin} \beta \gamma_5 u_{in})$

β, $\underline{\alpha}$, and γ_5 are Dirac operators and $\underline{\sigma}$ is the spin operator. Although Fermi chose the V form by analogy with the electromagnetic field case, the others are equally possible. Indeed any linear combination of these would be possible. As will be shown later, there is reason to believe that the "Tensor" form is a better choice than the "Vector" (Gamow and Teller, 1936).

In the first instance we shall assume, for simplicity, that the wave

functions of the neutrino and the electron are plane waves i.e. $\phi_n = \exp\left[-i(\underline{p}_n \cdot \underline{r}/\hbar)\right]$ and $\psi_{elec} = \exp\left[-i(\underline{p}_e \cdot \underline{r}/\hbar)\right]$ where $\underline{p}_n$, $\underline{p}_e$ are the momenta of the neutrino and the electron. This is true for the neutrino since it has very little interaction with anything, but in the case of the electron, is equivalent to neglecting the Coulomb energy of the electron. This is small compared with the disintegration energy if Z is small and the energy of the electron not too small. On this assumption, we can write the probability of a transition occurring between W and W+dW as

$$\frac{G^2}{2\pi^3}\,\frac{mc^2}{\hbar}\left|\int u^*_{fin}\, u_{in} \exp\left[-i(\underline{p}_n+\underline{p}_e)\underline{r}/\hbar\right] d\tau\right|^2 pW(W_0-W)^2\, dW$$

where p is the momentum in mc units and W is the total energy (including rest energy) in mc^2 units. W_0 is the maximum electron energy. W and p are expressible in terms of each other by the relation $p^2 = W^2 - 1$

If p_e and p_n are of the order of magnitude mc, the exponent is of the order $\frac{R}{\hbar/mc} \sim 1/100$. Hence the exponential is approximately unity and the matrix element reduces to

$$M = \int u^*_{fin}\, u_{in}\, d\tau$$

On this picture, if $\Delta I = 0$, $M \neq 0$ and this is an "allowed" transition. If M=0, the transition is said to be forbidden. The exponential must be expanded as a power series. The order of forbiddeness is the number of the first term which gives a non-vanishing matrix element. Since $\frac{R}{\hbar/mc} \sim 1/100$, the transition probabilities are reduced by approximately a factor of 10^4 per order.

If we include a term F(Z,W) which takes account of the Coulomb field of the nucleus we can write that the probability of emission of a beta-particle of total energy between W and W+dW in time dt is

$$N(W)\, dW.\, dt = \frac{G^2}{2\pi^3}\,\frac{mc^2}{\hbar}\,|M|^2\, F(Z,W)\, pW(W_0-W)^2\, dW.\, dt$$

Let us now consider the effect of the different parts of this equation. We see that $pW(W_o-W)^2$ represents the probability per unit energy interval of sharing the total energy W_o between the electron and neutrino such that the former gets W and the latter $W_o - W$. This factor is zero when W=0 or $W=W_o$. Also since pW alters slowly as $W \to W_o$, the shape of the spectrum is affected mainly by the factor $(W_o-W)^2$, i.e. the spectrum approaches the energy axis parabolically.

The effect of the Coulomb field of the nucleus is to attract negatrons and repel positrons. As a result of this factor there are relatively more negatrons and fewer positrons at the low energy end of the spectrum. F(Z,W), usually termed the "Fermi function", is given by

$$F(Z,W) = \frac{(s+1)2(2pR)^{2s-2}\, e^{\pi\alpha ZW/p}\, |\Gamma(s+i\alpha ZW/p)|^2}{[\Gamma(2s+1)]^2}$$

where Z is the atomic number of the product nucleus, and is negative for positron emission, R is the nuclear radius in units $h/2\pi mc$, $s=(1-\alpha^2Z^2)^{1/2}$ and α is the "fine structure constant". In order to calculate ft values and to draw Fermi plots, as will be described later, it is necessary to calculate the value of F(Z,W). Approximations which may be used in different cases are given below. (Feister, 1950).

The variable part of the function may be written

$$p^{2s-2}\, e^{\pi y}\, |\Gamma(s+iy)|^2$$

where $y = \frac{\alpha ZW}{p}$.

(i) Fermi in his original paper used a form of very limited applicability. For Z= 82.2, it took the form

$$F(82.2, p) \approx \frac{4.5 + 1.6p}{p}$$

(ii) Kurie, Richardson and Paxton (1936) gave the "Non-relativistic approximation".

$$F(Z,W) \approx \frac{2\pi y}{1 - e^{-2\pi y}}$$

This useful for $Z < 30$, $p \gg 1$. It gives one per cent accuracy for $Z \leqslant 10$ and for energies between 100 KeV and 2.8 MeV.

(iii) Bethe and Bacher (1936) gave another form which, as written by Longmire and Brown (1949a), runs

$$F(Z,W) \approx \frac{2\pi y}{1-e^{-2\pi y}} \left[\frac{(1+p^2)(1+4\alpha^2 Z^2)}{4} - 1 \right]^{s+1}$$

This is accurate to one per cent for $Z \leqslant 84$.

(iv) Nordheim and Yost (1937) provided a fourth approximation

$$F(Z,W) \approx \frac{2\pi y}{1-e^{-2\pi y}} (1-2s) p^{2s-2}$$

This gives one per cent accuracy up to $Z = 20$, and may be used for Fermi plot purposes up to $Z=35$.

Feister states that he plans to calculate $F(Z,W)$ for negatrons and positrons for all values of Z between 1 and 100, and of p between 0.05 and 7 (W between 0.67 KeV and 3.1 MeV). This was to have been completed by the end of 1949, but so far it has not appeared.

A further refinement which has not yet been taken into account is the screening effect of the atomic electrons, the effect of which has been studied by Longmire and Brown (1949, a,b) and Reitz (1950). The first two authors showed that the effect was negligible for S^{35}, but of more importance in the case of Cu^{64} and RaE. According to Reitz, the effect on electrons is negligable but is more important in the case of positrons, especially for $E < 300$ KeV and $Z > 25$.

5. ft Values.

Before going on to consider to what extent the Fermi theory has been supported by experiment, it will be well to say something about the "ft" values so often referred to in work in this field.

In 1933, an attempt was made by Sargent to fit the natural beta-emitters into some system similar to that of Geiger and Nuttall for

alpha-emitters. (Sargent,1933). On plotting the logarithm of the decay constant against the logarithm of the maximum beta-ray energy, he found that the points fell, very roughly, on two fairly well defined straight lines. Those lying on the upper one were later referred to as "allowed" transitions and those on the lower as "first forbidden". Only AcB appeared to lie off the curves to any considerable extent. It is interesting, in passing, that ten years later Konopinski agreed exactly with the classification resulting from the original Sargent Diagram. This quite empirical classification can be extended to the whole periodic table. In extending it in this way it is customary to plot it for short ranges of Z. A good example for low Z is given by Moon,(1949, p.45) and for short regions of high Z, by Feather and Richardson (1948).

After Fermi's theory was published it was possible to consider the matter theoretically. For allowed transitions the total transition probability, i.e. the reciprocal of the life-time is obtained from the relation.

$$\frac{1}{t} = \frac{G^2}{2\pi^3}\,\frac{mc^2}{\hbar}\,|M|^2 f(W_0)$$

$$\text{where } f(W_0) = \int_1^{W_0} F(Z,W)\, pW(W_0-W)^2\, dW$$

For simplicity, if we neglect $F(Z,W)$, $f(W_0)$ can be integrated fairly easily, remembering that $p = (W^2-1)^{\frac{1}{2}}$. The result varies rapidly with W_0 and has the approximate value $W_0^5/30$ for $W_0 \gg 1$ and $0.216(W_0-1)^{7/2}$ for $W_0 \sim 1$.

It is evident that if the calculation, including the factor $F(Z,W)$ were carried out exactly, and if $|M|^2$ were to remain constant, the product ft would be a constant equal to $\frac{2\pi^3}{G^2}\,\frac{\hbar}{mc^2}\,\frac{1}{|M|^2}$.

The chief difficulty lies in calculating M. However for "mirror-nuclei transitions", M must be near unity since the wave functions are so alike. (Mirror-nuclei are two neighbouring nuclei such that in one the number of protons exceeds the number of neutrons by one, and in the other the reverse is true).

Using recently published data on the mirror nuclei transitions, $H^3 \rightarrow He^3$, $C^{11} \rightarrow B^{11}$, and $Sc^{41} \rightarrow Ca^{41}$, the life-times of which are 12.46 yr, 20.5 min and 0.87 sec, we find that the ft values are 1150, 3890 and 2190 respectively. This is remarkable agreement in view of the fact that the life-times vary by a factor of about 10^9. In the case of the lightest mirror-nuclei, it is possible to calculate the value of G ($\sim 10^{-11}$) and therefore g ($\sim 10^{-48}$ erg cm). The smallness of the coupling between the electron-neutrino field and the nucleon is the cause of the slowness of beta-decay compared to most other nuclear reactions.

The value of ft is not constant. It rises with increasing Z and, if the transition is "forbidden", it rises by a factor of roughly 100 for each degree of "forbiddenness". Approximate values are given below. (Moon, 1949).

Z	Allowed	First Forbidden	Second Forbidden
$Z < 20$.	3×10^3	2×10^5	3×10^7
$20 < Z < 80$	8×10^4	2×10^6	5×10^7
$Z > 80$	2×10^6	2×10^7	10^9

If one uses this technique to give an indication of the degree of forbiddenness, the calculated ft value should be compared with ft values in the same region of Z to eliminate the effect of Z-change.

However the conclusions arrived at thus should be considered tentative and used only as confirmatory evidence.

Since the publication of a recent paper by Feenberg and Trigg(1950) the simplest method of calculating f has been to use the graphs which they provide. A very complete list of ft values based on the most accurate data available has been published by Feingold (1951).

6. The Verification of Fermi's Theory.

We shall now consider the various ways in which the theory can be tested.

(i) The comparison of the shape of beta-ray spectra as measured with the predicted shape.

(ii) The predictions made by the different forms of the theory (Fermi, Gamow-Teller, hereafter abbreviated as G-T) with regard to life time and degree of forbiddenness. This may serve to distinguish between the different forms of interaction.

(iii) The existence of the neutrino. If this can be shown experimentally to exist, the fundamental basis of the theory will have been made so much more certain by direct evidence of the postulated mode of decay.

To compare experiment with theory it is usual to draw a "Fermi" or "Kurie" plot. (Both terms are in use and there is not a consistent way of using them. In what follows, the term "Fermi Plot" will be used and will be understood to take into account the Coulomb effect). Considering the expression for the energy distribution of the beta-particles, it is evident that if $\sqrt{N(W)/pWF(Z,W)}$ is plotted against the energy of the particles a straight line will result provided $|M|^2$ remains effectively constant. (For forbidden decays $|M|^2$ may vary with energy). This straight line will cut the energy axis at $W = W_o$. This plot, then, does

two things. It shows whether the shape is the same as that predicted by Fermi theory for an allowed spectrum and it gives a more accurate value for W_o than can be obtained by inspection of the spectrum (due to the parabolic approach).

The calculation of $F(Z,W)$ is often laborious even when the approximations mentioned previously are employed. The tables promised by Feister will therefore be very acceptable indeed. However mention should be made of a very useful method of making the plot due to Bleuler and Zünti (1946). They put the negatron spectrum into the form

$$N(W)dW = C[Z]W^2(W_o - W)^2\theta[Z,W]dW$$

and give curves showing θ as a function of W for $40 \leq Z \leq 90$ and $Z\theta$ as a function of W for $0 \leq Z \leq 40$. $Z\theta$ is given for low Z because, as $Z \to 0$, $\theta \to \infty$ but $Z\theta$ remains finite. To make a "Bleuler-Zünti plot", one plots $\frac{1}{W}\sqrt{\frac{N(W)}{\theta}}$ against W, since $C(Z)$ is constant for a given isotope. For positrons θ must be multiplied by $e^{-Z/P}$, where P is the value of $Z\theta$ for Z=0. This method of making the plots was used by the writer in the examination of the spectra of Hg^{203}, Sm^{151} and Eu^{155}.

If the transition is not allowed, it may show a departure from the allowed shape, although many first forbidden spectra do show an allowed shape. In the event of departure from allowed shape, correction factors, given first by Greuling (1942), are used which, if the spin and parity changes assumed are correct, will lead to straight line plots. However in the case of higher degrees of forbiddenness there is often an unknown variable, the ratio of the matrix elements, which can be chosen only arbitrarily to fit the spectrum shape obtained.

In the early days, spectra which should have given straight line Fermi plots in fact did not. Konopinski and Uhlenbeck(1935) introduced a

modified form of the theory to explain this. In the equation for the electron energy distribution, the $(W_o - W)^2$ factor was replaced by $(W_o - W)^4$. However a "Konopinski - Uhlenbeck" plot predicted too high a value of W_o and more refined experiments of Tyler (1939) on the positron spectrum of Cu^{64}, of Feldman and Wu (1949) on Y^{91}, P^{32} and RaE and of Albert and Wu (1948) on S^{35} showed that the poor spectrum shapes previously obtained were due to the use of thick sources. For example, in the case of S^{35}, 1 and 2 $\mu gm/cm^2$ sources gave straight line Fermi plots but a 5 $\mu gm/cm^2$ showed departure from linearity at low energies. Much other work has also demonstrated the necessity of using not only sources of small average source thickness but also that the sources must be uniform. Sources possessing local variations of thickness of, say, 100 to 1, (as is, or was, common) lead to distortion of the spectrum.

The first decay which gave reasonable agreement with Fermi theory was In^{114}. The plot was linear from the end-point at 1.99 MeV to about 400 KeV. The lower end of the spectrum is affected by the presence of conversion electrons emitted in the decay of the 50day excited state of In^{114}. Since then considerable evidence has come forward supporting the Fermi distribution for allowed spectra. Indeed as the experimental techniques got better, it began to seem as though the allowed shape was universal. For a long time only the spectrum of RaE appeared to have a forbidden shape and its departure from the allowed shape was evidently real. Recently, however, a large number of forbidden shape spectra have been found. Before mentioning the evidence from this work it is necessary to mention the variation in the theory introduced by Gamow and Teller.

7. Gamow-Teller Selection Rules.

There is good evidence that the selection rule for allowed transitions, $\Delta I=0$, is not always obeyed. For example, in the K-capture disintegration of Be^7

$$Be^7 + k \rightarrow Li^7 + \nu$$

Li^7 is produced both in the ground state and in a 440 KeV excited state. If one assumes that $|M|$ is equal for the two modes of beta-decay, the calculated ratio of the intensities of the decay to the ground state and the decay to the excited state is ~10:1. Since this agrees with the experimentally measured intensity, there is good reason to believe that both transitions are allowed (this being assumed in making the calculation). However the excited and ground states of Li^7 are not expected to have the same spin values. It is likely that $I=\frac{1}{2}$ for the excited state and $I=\frac{3}{2}$ for the ground state. Thus ΔI cannot be zero for both transitions. This suggests $\Delta I=1$ may also give an allowed transition.

Further evidence comes from the reaction

$$He^6 \rightarrow Li^6 + \beta^- + \nu$$

We can consider Li^6 to be composed of an alpha-particle (I=0) plus a deuteron (I=1). Therefore the spin of Li^6 is expected to be I=1. This is in agreement with the experimentally determined value. In a similar way we can think of He^6 as being made up of an alpha-particle plus two neutrons. The spin of the double neutron, in the ground state, should be zero. Hence we expect the spin of He^6 to be zero, like all nuclei having even numbers of protons and neutrons, whose spins have been measured. Thus the spin change in the beta-decay is $\Delta I=1$ and, on Fermi theory, the transition is forbidden. However the half-life of the decay corresponds to a "super-allowed" transition ($ft \sim 10^2-10^3$, t in seconds). Also the Fermi

plot obtained by Brown and Perez-Mendez (1949) is straight from $E_o > 3$MeV to 150 KeV. Similar situations arise in the decays of C^{10}, F^{18} and Na^{22}. It appears therefore that the selection rules for allowed transitions should be altered to permit a spin change of $\Delta I = \pm 1$. Gamow and Teller (1936) showed that this was possible if one assumed a tensor interaction instead of the polar vector form chosen by Fermi. Thus for allowed transitions, $\Delta I = 0, \pm 1$. A physical interpretation of the rules is that, according to Fermi theory, the neutrino and the electrons always leave with opposite spin whereas, in the G-T theory, they may leave with parallel or opposite spins.

Consider, however, the following reaction

$$Be^{10} \rightarrow B^{10} + \beta^- + \nu$$

On the same reasoning as before, since Be^{10} is equivalent to He^6 plus an alpha-particle and B^{10} is equivalent to Li^6 plus an alpha-particle, we expect a spin change $\Delta I=1$. Experimentally, however, the decay is found to be forbidden. This can be explained by taking into account the parity changes involved. According to the G-T theory, $\Delta I=0,1$ (and 2), with parity change, is first forbidden, while $\Delta I=0, 1$ without parity change, is allowed. Presumably there is a parity change in the decay of Be^{10}.

The selection rules for the vector and tensor forms of interaction are given by Konopinski and are reproduced below. "Yes" indicates that there is a parity change, "No" that there is not.

Interaction	Allowed	First Forbidden	Second Forbidden
V (Fermi)	$\Delta I=0$ (no)	$\Delta I=0$, 1 (no $0\leftrightarrow 0$) (yes)	$\Delta I=1,2$. (no $1\leftrightarrow 0$) (no)
T (G-T)	$\Delta I=0$, 1 (no $0\leftrightarrow 0$) (no)	$\Delta I=0$, 1, 2 (yes)	$\Delta I=2,3$ and $0\leftrightarrow 0$ (no)

Skyrme (1950) gives the selection rules for all forms of interaction.

Further evidence for G-T rules comes from fuller consideration of the simplest mirror nuclei such as n,p; H^3, He^3 and of the simple nuclei He^6, Li^6, the relative sizes of the nuclear matrix elements of which can be calculated. (Wigner, 1939). Good agreement between the values of M^2ft for the cases of H^3 and He^6 is obtained, if M^2 is calculated assuming a tensor (G-T) interaction, but not if a vector(Fermi) interaction is assumed. The values are given below. The value of M^2ft for the case of the neutron covers a wide range due principally to the range in quoted life-times (9-30 min). If we assume the value of M^2ft to be approximately that given for the other decays, by the G-T interaction, one can predict a half-life of 10-12 min for the neutron.

Radioactive Nucleus	$\frac{ft}{(exptl.)}$	$\frac{M^2ft}{(G\text{-}T)}$	$\frac{M^2ft}{(Fermi)}$
n	$\left\{\begin{matrix}875\\2916\end{matrix}\right.$	$\left\{\begin{matrix}656\\2186\end{matrix}\right.$	$\left\{\begin{matrix}219\\728\end{matrix}\right.$
H^3	1125	844	271
He^6	584	877	0

The same shape is predicted by all forms of interaction for allowed spectra. It was therefore realised that to distinguish between Fermi and G-T interactions, it was necessary to investigate forbidden spectra. However, as has been mentioned earlier, a large number of first forbidden transitions gave an allowed shape and it was not till recently that forbidden shapes, other than that of RaE, were observed. The first to show a

definitely forbidden shape was Y^{91} in the decay

$$Y^{91} \longrightarrow Zr^{91} + \beta^- + \nu$$

The spin change was believed to be 2 and the parity change "Yes". According to Fermi rules $\Delta I=2$ is second forbidden but it can be first forbidden by G-T rules. If ΔI is one unit greater than the degree of forbiddenness, as is permissible on G-T rules, a unique energy dependence differing from an allowed shape by a factor

$$\alpha \sim (W^2 - 1) + (W_0 - W)^2$$

is predicted. The "allowed" Fermi plot was definitely curved, but use of the α-factor resulted in a perfectly straight line. This provided further evidence of the correctness of the G-T choice of interaction. Further work showed that the decays of Cl^{38}, K^{42}, Y^{90}, Sr^{90}, Sr^{91}, Sb^{124}, Sn^{123}, Sn^{125} and Cs^{137} were in the same class. The nuclear shell model (Haxel, Jensen and Suess, 1950) predicts $\Delta I=2$ for each of these disintegrations. All required the α-factor correction. Also it was shown that the value of ft $(E_0^2 - 1)$ was always $\sim 10^{10}$, which was in agreement with theoretical expectations.

The decay of Be^{10} will be considered next. The spin change is $\Delta I = 3$ and the half-life is 4.4×10^{13} sec. On Fermi rules this would be third forbidden, while on G-T rules it could be second forbidden. Use of the second forbidden D_2 factor

$$D_2 = \frac{1}{90}\left[3(W_0 - W)^4 + 10(W^2-1)(W_0-W)^2 + 3(W^2-1)^2\right]$$

which was predicted by Marshak (1949) on the basis of the G-T interaction gave a straight Fermi plot. The spectrum shape is again unique. Similarly the use of the unique D_3 factor

$$D_3 = (W_0-W)^6 + 7(W_0-W)^4(W^2-1) + 7(W_0-W)^2(W^2-1)^2 + (W^2-1)^4$$

in the case of K^{40} gives good agreement from 1.38 MeV to 500 KeV

(Alburger, 1950). This infers that the decay of K^{40} is third forbidden on G-T rules. Change of parity is predicted by the shell model. This is in agreement with the selection rules and spectrum shape. The polar vector interaction requires that the decay be fourth forbidden (since the spin change is four) and is therefore ruled out.

So far it might seem that either of the G-T interactions, tensor or axial vector (for both give rise to G-T rules), fit all the facts. However this is not so, notably in the cases of Cl^{36} and RaE. The shape of the former is fitted by no single interaction, but Wu and Feldman(1949) have shown that it can be fitted by a linear combination of the scalar and tensor interactions, or of the axial and polar vector interactions. It happens that these linear combinations also fit the α-type spectra discussed earlier. However final conclusions await the verification of the spin change and spectrum shape. According to Wu (1950) useful information regarding the necessity of using such linear combinations would be gained by the study of a decay where the spin change is equal to the degree of forbiddenness. Such a decay is provided by Rb^{87} which has a spin change of three and is at least third forbidden by half-life considerations. Wu states that this is the only case known (presumably for $\Delta I > 1$). This activity has been studied by the author assisted by Mr. D.Dixon. It will be discussed later.

8. Evidence for the Neutrino Hypothesis.

The direct evidence for the existence of the neutrino can arise, theoretically at least, in two ways.

(i) The inverse beta-process. This is a process of the form

$$n + \upsilon \rightarrow p + \beta^-$$

Of course, to detect this experimentally it is necessary that the neutrons

be in some nucleus, e.g.

$$Li^7 + \nu \rightarrow Be^7 + \beta^-$$

The incident neutrinos would have to have sufficient energy to supply the mass difference between Be^7 and Li^7. The cross-section for such a process is extremely small however. If we take the cross-section for hitting the nucleus as approximately 10^{-24} cm^2 and the probability of beta-decay in the nucleus as 10^{-20}, the cross-section for the process is $\sim 10^{-44}$ cm^2 which is far too small to detect.

(ii) The Recoil Method. This method offers more hope of success, however. It is evident that the energy, and momentum, of the recoil in the nucleus will depend on whether or not a neutrino is emitted along with a given electron. A neutrino travelling in the same hemisphere as the electron will cause the nucleus to have a higher momentum than if only an electron of the same energy were emitted and conversely if the neutrino travels in the opposite direction, it will cause the nucleus to travel with smaller momentum.

The energy of the neutrino, E_ν, is given by

$$E_\nu = E_o - E_\beta$$

Its momentum, p_ν, is given by

$$\underline{p}_\nu = \underline{p}(\text{nucleus}) + \underline{p}_\beta$$

The energy and momentum are related by

$$p_\nu^2 c^2 = E_\nu^2 + 2E_\nu \mu c^2$$

where μ is the neutrino mass. If this is zero we have

$$p_\nu c = E_\nu$$

There are different ways of using these relations in experiments designed to confirm the existence of the neutrino. These experiments are for the most part rather ingenious and will be described briefly below.

One of the earliest attempts was due to Leipunski (1936). His work on the recoil of charged B^{11} ions arising from the decay of the 21 min positron emitter, C^{11}, gave evidence of recoils more energetic than those expected in the absence of a neutrino. The experimental results were very uncertain however.

Crane and Halpern (1938,1939) used Cl^{38} which is a conveniently long-lived source (37 min) having high energy beta-rays (up to ~ 5 MeV) which is of course an advantage. The Cl^{38} was put into a cloud chamber and photographs were taken with a short delay after expansion. This allowed the ions to diffuse and the energy of recoil was estimated by counting the number of drops. The momentum of the beta-particle was measured by its curvature in a magnetic field. Their results were inconsistent with the recoil being due to the emission of a beta-particle alone and therefore supported the neutrino hypothesis.

The next experiment to be mentioned is that of Allen (1942). This followed a suggestion of Wang (1942) that if the recoils due to Be^{7} (a K-capture source) were measured, the recoil energy should be mono-energetic, since in each decay the neutrino should carry off the entire decay energy (apart from a negligable K X-ray recoil). The mass of Be^{7} is 7.01916 and that of Li^{7} is 7.01822. Thus the energy available for the transition is 0.00094 Mc^{2}. Since the neutrino is very light practically all this energy appears as kinetic energy and its momentum is thus 0.00094 Mc (neglecting the neutrino mass). This is equal to the momentum of the recoiling Li^{7} nucleus, whose energy is therefore ~ 60 eV. Allen devised as ingenious method of detecting and measuring the recoil energy of these nuclei, which he found to have a maximum value of ~ 48 eV.

There is a gamma-ray of energy 0.45 MeV but this could give rise to a recoil of only 5.6 eV. To confirm that the recoils were not due to the gamma-rays, Allen showed that there were no coincidences between the recoils and the gamma-rays.

In a fourth attempt, Jacobsen and Kofoed-Hansen (1948) used the radioactive gas Kr^{88} which decays with a 2.7 hr period to Rb^{88}. Rb^{88} decays in turn to Sr^{88} with a half-life of 18 min. The recoil energies of the Rb^{88} ions were measured as a function of a varying negative voltage. The Rb^{88} ions were detected by allowing them to deposit on a foil, the activity of which was afterwards measured. This gave a maximum energy of recoil of 51.5 eV in good agreement with the value calculated from the maximum beta-ray energy, 2,4 MeV. This in itself, of course, does not prove the existence of a neutrino., but gives more confidence in the results. The experimental curve they obtain is different from that expected for decay without neutrino emission. Correction for the various errors in the experiment make the result more conclusive.

A recent experiment of Smith and Allen (1951) on the Be^{7} decay gave recoil nuclei of maximum energy 56.6 $\pm$ 1.0 eV. They did not succeed in getting a monoenergetic group of recoil energies.

9. The Mass of the Neutrino.

That the mass of the neutrine is very small can be seen from several experiments. Firstly, the agreement between the upper energy limit of certain spectra agrees very well with the mass difference of the nuclides concerned. Haxby, Shoupp, Stephens and Wells (1940) showed, by consideration of the decay of N^{13} into C^{13}, that the mass of the neutrino must be less than 1/10th. of the mass of the electron.

Secondly, the shape of the spectrum of tritium near the end-point is

very sensitively dependent on the mass of the neutrino. Curran, Angus and Cockroft (1949 b,c) have shown that the mass is less than 1/300th. of the electron mass. Experiments of Hanna and Pontecorvo (1949) give a value of less than 1/500 th., while Cook, Langer and Price (1948) showed as a result of their investigation of S^{35}, that the mass was less than 1/100th. of the mass of an electron.

Thirdly, if the neutrino mass were an appreciable fraction of the mass of the electron, the energy considerations given earlier would lead one to expect a greater number of stable neighbouring isobars, since there would have to be a certain mass difference in order to create the neutrino. Good reviews of the experimental verifications of the neutrino hypothesis have been given by Pontecorvo (1949) and Crane (1948). These articles go into more detail than has been possible here.

To sum up, the situation in the theoretical side of the field of beta-decay has been considerably improved in recent years and especially in the period during which this research was being prosecuted. The reasons for, and the result of the various investigations carried out by the writer will be considered later, but it may be said here that the results were generally in agreement with the predictions of Fermi theory, using the G-T interaction. These and the large number of other experiments which have been performed have therefore justified the basic neutrino hypothesis of Pauli. Experiments to detect the neutrino have been, at least, in favour of its existence though none can yet be considered to offer certain proof.

B. Experimental.

1. Instruments and Techniques used in Beta- and Gamma-Ray Spectroscopy

There are several quite distinct methods of analysing the radiations emitted by radioactive substances. It is proposed to describe and compare these different methods which are listed below:-

I. Absorption - simple, critical and coincidence.

II. Cloud Chambers.

III. Magnetic Spectrometers.

IV. Electrostatic Spectrometers.

V. Proportional Counters.

VI. Scintillation Counters.

VII. Electron Sensitive Emulsions.

VIII. Other Methods of Measuring Gamma-Ray Energies.

There are separate sub-divisions in the above categories. Also sometimes a technique in one group will be associated with a technique in another. For example, absorption experiments may be carried out in which the particles absorbed are in coincidence with others lying in a given energy interval selected by a magnetic spectrometer.

I(a). Simple Absorption.

A good review article on the technique of simple absorption has been given by Glendenin (1948). This method of analysis requires relatively simple apparatus, is sensitive and can give quick though somewhat rough measurements of particle and photon energies in cases of fairly simple decays. It has been much used in the course of the present research for preliminary identification and examination of radioactive sources. For beta-ray energy measurements, the set-up consists merely of a source, a series of absorbers and a detecting device. This device, in the case of

stronger sources, may be an electroscope. Alternatively a Geiger counter is employed. The absorbers are inserted as close to the detecting device as is convenient and the counting rate, or intensity of ionisation, is noted as the absorber thickness is increased. As the thickness of the absorbing foils used in beta-analysis is rather small, it is customary to weigh the foils and express their thickness as an areal density - in mgm per cm^2. It is found that if homogeneous electrons are being examined, a plot of intensity against absorber thickness yields a fairly straight line which, at the limit of electron range, turns quite sharply to a horizontal line which may correspond to the background intensity, or perhaps to background plus gamma-radiation. On the other hand, if a beta-spectrum is being investigated, it is sometimes found that graphing the logarithm of the intensity against absorber thickness yields a fairly straight line with a reasonably defined end-point. Because of the approximate straightness of these lines, it used to be the custom to quote the relative "hardnesses" of beta-spectra in terms of the apparent absorption coefficients which the slope of the lines yielded. In many cases the absorption curve had a distinct curvature and the initial slope was used. This method is now rather uncommon.

At the present day it is customary to read off the end-point of the curve and estimate from some range-energy relationship the corresponding electron energy. The writer has usually employed a graph given by Glendenin (1948, fig.12). This seems in general to lead to consistent results. A formula due to Flammersfeld (1946) has also been found successful. It can be used in the range 50 KeV $\leq$ E $\leq$ 3 MeV and runs

$$E = 1.92\,(R^2 + 0.22R)^{\frac{1}{2}}$$

where R is the range in gm/cm^2 and E is in MeV.

Feather gave a rule for determining beta-ray energies in 1930 (Feather, 1930). Using the more recent constants (Feather, 1938) it takes the form

$$R = 0.54E - 0.160$$

where R and E have the same significance as before. He describes a technique which appears to give a more precise end-point energy and tells one something about the shape of the spectrum. As usually carried out, the absorption curve is compared with that of a standard and well known source in an absolutely standard experimental arrangement. He himself chose RaE since its spectrum was the only one in 1938 whose shape was known to be measured reliably. However in view of its unusual spectrum shape, it might not be his choice now, since other more convenient spectra have been accurately measured.

When a beta-spectrum is complex, it is often possible to identify only the highest energy component end-point if the corresponding transition is of the same order of intensity as the other transitions. However occasionally two, and even three, end-points can be seen (e.g. Hg^{203} absorption curves; see later). The maximum energies in the case of beta-spectra must not be too close together if this is to be possible.

In the case of gamma-rays one always plots the logarithm of the intensity against absorber thickness, since the intensity is reduced exponentially. From the graph one deduces the half-thickness, or mass absorption coefficient, and the energy is determined from tables or graphs (e.g. Glendenin, 1948, figs. 15, 17; Handbook of Chemistry and Physics, 1949). If two or more gamma-rays are present, their energies can be measured only if they are well separated. If this is so, two or more regions of distinctly different slope are observed and by subtracting

the intensity corresponding to the highest energy gamma-ray, one can graph the logarithm of the intensity of the second gamma-ray against thickness and thus measure its energy. However in the case of sources emitting many gamma-rays, as was the case with neutron irradiated samarium (see later), this method fails completely. Another disadvantage of the method is that, if the photon energy is high, the absorption coefficient alters slowly with energy and even starts to rise at high energies, which means that two different absorbers must be used to give an unequivocal result.

For gamma-rays the coincidence method due to Curran, Dee and Petrizilka (1938) may be employed. The gamma-rays impinge on a thin converter which emits Compton and photo - electrons. These pass across a thin walled counter into a second thin window counter. The coincidence rate of the counters is plotted against increasing thickness of absorber placed between the counters. At the end-point, the absorber thickness, plus air gap and window thickness, is measured and the energy of the gamma-ray deduced. This method reduces the difficulty of measuring a gamma-ray energy to that of measuring a beta-ray energy by absorption and is simple and sensitive. However it still fails for two gamma-rays close in energy.

To sum up, the absorption method has important advantages and disadvantages. It is very sensitive and quick, and can therefore be used for preliminary examination of new radioactivities and to check the existence of suspected activities in a radioactive source. The technique was used for both purposes in the course of this research. It is also very simple and requires very little apparatus. However it does not normally yield accurate energy values, its energy resolution is low and it is easy to draw wrong conclusions on the basis of absorption methods alone.

I(b). Critical Absorption.

This method, which applies only to the measurement of gamma-rays, may be used up to ~100 KeV. It makes use of the fact that the absorption coefficient of any element is very much higher for energies slightly greater than its K absorption edge energy than for energies somewhat less. To use this method a series of absorbers of increasing atomic number, whose K edge energies are in the region of the gamma-ray energy, are inserted in turn between the source and the detector. As Z increases, the intensity falls slowly and then at some value, Z, say, rises sharply. The energy of the gamma-ray lies between the K absorption edge energies of the elements of atomic numbers Z and Z-1. This method determines the energy to ~5% for lower energies and to ~2-3% for higher energies.

I(c). Coincidence Absorption Methods.

Under this heading will be considered methods distinct from the coincidence method of measuring gamma-ray energies due to Curran et al., mentioned earlier. Coincidences between gamma-rays and beta-rays, or between gamma-rays and gamma-rays, or between beta-rays and conversion electrons from a source are investigated and the general technique is an important tool in the unravelling of complex spectra. Good reviews of the technique are given by Dunworth (1940), Mitchell (1948), Curran and Craggs (1949, pp. 158-161) and others. We shall not go into the subject in detail, but shall consider a few special cases which will demonstrate the technique and reveal something of its usefulness. For example, consider a gamma-decay scheme such as is shown in fig.47(A). If the source in question is placed between two gamma-counters giving N_1, N_2 counts per minute in each counter, the accidental coincidence rate N_c, due to events occurring within the finite resolving time, τ, of the circuit, will be

$$N_c = 2N_1N_2\tau$$

But if an appreciable fraction of the nuclei decay to the ground state by the 15 KeV-85 KeV transition, an additional number of coincidences will be observed, provided that the 85 KeV transition follows the 15 KeV with a half-life not appreciably greater than the circuit resolving time. It is usually much less. It is possible to calculate from the coincidence rate and the efficiencies of the counters, the fraction of nuclei de-exciting by each mode of decay. Also if absorbers are placed in front of one counter, the resulting absorption curve may be split into two regions corresponding to the 15 KeV and the 85 KeV gamma-rays, provided there is not too much internal conversion of either. Alternatively the gamma-ray counters may be of the scintillation type so that one can select a particular gamma-ray energy, say 85 KeV, and show by absorption, or by pulse size analysis, that the gamma-rays in coincidence have an ebergy of 15 KeV. Fuller details of the experimental technique are to be found in the references given previously.

The examination of the beta-gamma coincidence rate can often lead to further information about the complexity of a beta-spectrum. Let us consider again in the decay of fig.47(A), but for simplicity, let us neglect the 15 and 85 KeV gamma-transitions. Often it is difficult, or even impossible, to detect or measure the energy of the lower energy beta-transition by absorption or other means. However if the source is placed between a gamma-ray counter and a beta-ray counter, the only coincidences observed, excluding the accidental and background ones, are due to coincidences in time between the beta-particles of the 150-KeV transition and the gamma-rays, again providing the half-life of the gamma-transition is not longer than the resolving time of the circuit. By placing absorbers in

front of the electron counter, one can measure the energy of the beta-rays in coincidence with the gamma-rays. It is customary to graph the ratio

$$R = \frac{\text{Number of beta-gamma coincidences}}{\text{Number of beta-particles detected}}$$

against absorber thickness as the number of beta-particles recorded per minute is reduced by the absorbers. The thickness of absorber giving zero coincidence rate (after subtracting the accidental and background rate) gives the maximum energy of the lower energy transition (here 150 KeV). It should be noted that if a spectrum consisting of a beta-ray followed by a gamma-ray is examined in this way, the ratio R will remain constant as the absorber thickness increases.

If the beta-ray counter is replaced by a magnetic spectrometer, the maximum energy and spectrum shape of the lower energy transition can be examined free from the higher energy beta-rays. This way of determining these properties is more precise than by deducting them from analysis of a Fermi plot.

In coincidence work it is an advantage to have a strong source since (a) data is accumulated more quickly (b) the effect of backgrourd is less. However a strong source gives rise to a relatively much higher accidental coincidence rate. Dunworth (1940) has shown that little advantage is gained by having a source with a disintegration rate, N_o , greater than that given by

$$N_o = \frac{1}{2\tau}$$

where τ is the resolving time of the circuit and is generally 1 μsec, provided τ does not vary with counting rate. It is usually measured by placing the two counters well apart and irradiating each with a separate strong source. If the individual counting rates, N_1 , N_2 , and the

coincidence rate, N_c , are measured, τ can be evaluated by use of the equation

$$\tau = \frac{N_c}{2N_1 N_2}$$

When using a magnetic spectrometer, the source strength must be high which introduces further difficulties of sources thickness, etc. For example if we consider a semi-circular spectrometer of resolution 1% and transmission 1%, the efficiency for beta-ray counting is 10^{-4}. A gamma counter is unlikely to have an overall efficiency greater than 10^{-3}. In a source giving, say, 10^6 disintegrations per second and giving a gamma-ray for each beta-particle this would give only one true coincidence per 10 seconds. Obviously one would wish to use a more efficient spectrometer. The answer would appear to lie with the scintillation counter, which detects gamma-rays much more efficiently and has a higher solid angle for beta-particles. Even in the case of simple coincidence-absorption investigations it is well worth while replacing the gamma detecting Geiger counter by a scintillation detector as was done by Mr. Lewis and the author in the study of the decay schemes of samarium and europium. This is believed to be one of the first experiments in which a scintillation counter and a Geiger counter were used in coincidence.

Finally it should be pointed out that if the decay scheme is more complicated than those discussed, it is still possible to solve it by absorption methods, as has been pointed in the review articles already mentioned. A good method, probably, is to select different energies of gamma-rays by the scintillation spectrometer and measure the intensity and energy of the beta-rays in coincidence with these gamma-rays.

II. Cloud Chambers.

This well known technique, originally used by C.T.R. Wilson, has been employed to a considerable extent in beta-rays spectroscopy, though not so much in recent years as formerly. In the course of time it has been improved and its facilities extended. In beta-ray spectroscopy it is possible to use the source in gaseous form, if this is chemically and physically suitable, which is of great value since it eliminates source thickness and backing problems; or as a solid source. In the latter case the source may, if necessary, be spread over a relatively large area, which is a help if the source is weak. However the efficiency of detection is rather low. The actual efficiency during an expansion may be ~10% but since the "dead-time" of a cloud chamber is very long compared to the sensitive period, the overall efficiency is much lower. The method is of value since it gives the nearest approach we may ever have of a picture of the motion of the decay electron, and even of the recoil atom. (The photographic emulsion technique shares some of the properties of a cloud chamber and will be mentioned later). One can measure directly (a) the point of origin of the particle or particles (this can be of value in complex decay, pair production, etc.), (b) the range, if not too great for the chamber, (c) the density of ionisation along the track and (d) the radius of curvature in a magnetic field which gives the momentum and sign of charge of the particle. In preliminary investigations it may yield less misleading results than other techniques.

However it has disadvantages, too. One, the low efficiency, has already been mentioned. Another is the time and labour involved in getting good statistics. As the number of tracks per exposure is limited by consi-

derations of ease of measurement, a large number of exposures is necessary and the time involved in measuring the radius of curvature of a large number of tracks (which is the best way of deducing the momentum, or energy, of the particle) is very considerable. The result is that in most work done by this technique, the number of particles per momentum interval was too low for accurate comparison of experiment with theory, or to determine the end-point accurately or, in some cases, to determine whether the spectrum was complex. Another difficulty in attaining high accuracy is due to the scattering of electrons in the gas, as has been pointed out by Gupta and Ghosh (1946). These authors give a theoretical treatment of the effect, which can lead to fairly serious errors in the measurement of curvature. Further information on the cloud chamber technique is given in the paper just referred to.

III. Magnetic Spectrometers.

The use of a magnetic field to separate particles of different energies was first made in the early experiments of von Baeyer and Hahn, previously mentioned. In their arrangement, a vertical slit was placed between a radioactive source and a photographic plate. A uniform magnetic field acted parallel to the slit. This constituted a crude non-focusing spectrograph. Since particles were deflected to different extents according to their momenta, a spectrum was obtained on the plate. The resolving power was sufficient to observe electron conversion lines superposed on continuous spectra and to measure the corresponding momenta.

In 1913, Danysz suggested the well-known 180° focusing spectrometer which has remained the most important type in general use to the present day. (Danysz, 1913). This utilises the property that, if a bundle of rays of equal radius of curvature is emitted from a source into a fairly small

solid angle, the rays tend to focus at a point distant 2ρ along a line perpendicular to the mean direction of emission of the beta-rays, where ρ is the radius of curvature.

This idea was fruitfully exploited by Rutherford and Robinson (1913) who measured the energy of many electron lines with an accuracy far exceeding that of von Baeyer and Hahn. Other workers, notably Ellis, improved the apparatus to such an extent that energy measurements could be made to a relative accuracy of 1 in 1000 and an absolute accuracy of 1 in 500 (Ellis and Skinner, 1924). Among other things this involves the measurement of the field strength to that degree of accuracy and such work has scarcely been bettered to-day. However it has to be noted that the naturally radioactive sources which were used have generally a very high specific activity which permitted the use of very thin sources of small area, which we shall see is desirable.

In the usual arrangement the source consists of a wire or narrow strip of thin aluminium, nylon film or other material which is coated with a thin layer of source. The source and source support must be thin and the latter made of low Z material, to avoid distortion of the spectrum shape. In front of the source is placed a slit whose long dimension is parallel to the magnetic field and perpendicular to the median plane of the path of the beta-particles. Its purpose is to limit the solid angle of emission of the particles. The narrower the source and slit, the better will be the resolving power of the instrument, at the expense, however, of intensity. The particles then pass through a system of baffles (to stop scattered electrons) and are detected by a photographic plate or by a Geiger counter, which has a slit in front of the window. In the first case, which is the original method, many energies are recorded at one time, thus giving a

spectrum between two widely distributed energies. Used thus, the instrument is sometimes called a "spectrograph" by analogy to the optical spectrograph. When a Geiger counter and slit are employed, a particular momentum range is selected. This instrument may be termed a "spectrometer" since it is acting as a monochromator. Both methods have their advantages and disadvantages. The plate has the useful property of being able to collect data over a wide energy range simultaneously and can do so over a long period, integrating the intensity. Also no lifetime corrections need be made if the spectrum is contained on one plate. For high accuracy, especially with weak sources, it is an advantage to use a permanent magnet since the field strength is not likely to vary as an electro-magnetic field might. However it is not possible by this method to measure relative intensities accurately. This was not of such moment when interest was concentrated in the measurement of electron line energies and the relation of the values to the energy levels of the nuclei involved but now that interest is, to a much greater extent, concerned with the examination of the shapes of beta-spectra this disadvantage becomes important. It may be, however, that the use of electron sensitive emulsions, enabling the tracks of individual electrons to be distinguished, will alter the position. The use of these emulsions will be considered later.

When the counter technique is used, however, the sensitivity is much increased and, since each electron entering the counter gives a count, it is possible to make quantitative measurements on the intensity distribution of the spectrum. However there are the difficulties of having to measure each momentum interval separately, of having to allow for the decay of the source, of greater electrical complication and of window thickness. The latter, like the question of source thickness, is very important. The

presence of a window allows only a fraction of electrons in the lower energy region to enter the counter and thus the spectrum shape in the lower energy region is distorted to a greater or less extent. This problem has been tackled in several ways. The obvious way is to reduce the window thickness to a minimum. Recent workers have used windows as thin as 0.03 mgm/cm^2 (Langer and Cook, 1948), which corresponds to an energy of about 5 KeV and probably affects the spectrum shape up to ~15 KeV. Langer and Cook also tried to use an electrostatic potential of 25,000 volts between the exit slit and the counter window in order to give the electrons sufficient energy to pass through the window. The attempt was not completely successful.

Absolute measurement of energy in a beta- or gamma-ray spectrometer is both laborious and difficult. However when it is necessary, the semi-circular spectrometer is the best of the magnetic resolvers for the purpose, since it utilises a uniform field which can be measured absolutely and thus the energy of a particle of a given radius of curvature can be determined absolutely. However it is more common to measure energies relative to a standard electron line, e.g. the ThB F-line (241 KeV) and the ThC" gamma-ray (2.62 MeV). The flux is usually measured by the flip coil or the rotating disc techniques.

In the case of gamma-rays, it is usual to provide more shielding, usually lead, between the source andthe detector than is necessary for beta-ray work. Two techniques have been employed. In the first the gamma-rays are allowed to pass through a low Z absorber of thickness sufficient to stop all the beta-rays from the source. The Compton electrons ejected by the photons and going in the forward direction are detected. Their energy is related to that of the gamma-ray by the relation

$$E_e = \frac{E_\gamma}{1 + mc^2/2E_\gamma}$$

where E_e is the maximum electron energy and E_γ is the quantum energy. A continuous spectrum of electrons is obtained by this technique and the energy limit is used in calculating E_γ .

Alternatively the source and low Z absorber may be surrounded by a thin high-Z foil. The photons eject photo-electrons from this foil and, provided it is thin enough, these give sharp lines superposed on the Compton background corresponding to conversion in the K,L and, sometimes, M shells. Also the photoelectrons arising from internal conversion in the source itself may be measured. By measuring either internal or external conversion lines an accuracy of 1% or better is obtainable and a separation of gamma-rays of energy difference less than 10% can be effected (Ellis and Skinner, 1924, Siegbahn, 1946).

A theoretical treatment of the spectrometer has been given by Li (1937) and by Geoffrion (1949) and is reviewed by Persico and Geoffrion (1950). It is not proposed to repeat it here. One finds that, using the Rayleigh criterion, the resolution R, defined as the reciprocal of resolving power, is given by the general expression

$$R = \frac{\delta p}{p} = \frac{1}{4}\left[\frac{s+w}{\rho} + (\alpha^2 + \beta^2)\right]$$

where s and w are the source and detector slit widths respectively, ρ is the radius of curvature, α and β are the semi-angles subtended bythe slit at the source and p is the momentum. It can further be shown that the intensity factor, or fraction of electrons leaving each square centimetre of the source which arrive at the collector is proportional to $\rho^2 R^{5/2}$. Thus one wants a large radius of curvature and an increase of resolving power of the instrument involves a much greater reduction in the intensity. A typical instrument might give 1% resolution and 0.1% transmission, thus

giving an efficiency (or ratio of transmission to resolution) of one tenth. It is actually possible to attain an effiency of one eighth. In order to examine weak sources there seems no reason why the resolving power should not be considerably decreased with a correspondingly large increase in transmission.

An interesting modification of this spectrometer was made by Dzelepow, Kopjova and Vorobjov (1946) who, to increase the trsnsmission, used six semi-circular spectrometers of fairly low resolving power in one vacuum chamber in order to measure the very weak activity of K^{40}. However the accuracy of their results appears to be suspect in view of later work and theoretical predictions.

Msgnetic Lens Spectrometers.

The property of "helical focusing" was first proposed by Kapitza(1924) and tried by Tricker (1924). In this method the source is a small disc placed on the axis of a uniform solenoidal magnetic field and particles of a given energy emitted at a small angle to the axis come to an approximate focus on the axis after describing one complete helical turn. Witcher(1941 showed that, in fact, the best focus was a ring whose centre lay on the axis and whose plane was perpendicular to it. He constructed a spectrometer using this principle, a theoretical treatment of which was given by Persico (1949). This method has two advantages over the "lens" type spectrometers developed from it, that (a) the calculations of path, and energy, are rigorous and (b) sources of fairly large area can be used (about 16 times that of a short lens spectrometer of the same performance). Also the instrument is easily adjusted and is less sensitive to magnetic disturbances. The performance figures of Witcher's apparatus were 1% transmission and 5% resolution. However, the power consumption was large

and the apparatus was of large dimensions.

The short coil lens spectrometer uses essentially the same method of focusing but employs a coil of limited dimensions situated midway between the source and the detector. A ring baffle is situated at the centre to select a given angular spread of electrons. First suggested by Klemperer (1935), the theory was worked out by Deutsch, Elliott and Evans (1944). Apart from spherical aberration, the lens may be treated as a simple optical lens. Typical performance figures worked out theoretically were T $\sim$0.4% and R $\sim$2.5% which are slightly better than those of Witcher's spectrometer. However Witcher's apparatus was not as efficiently designed as it might have been and probably the only advantage which the short lens spectrometer possesses over the uniform solenoidal field method is that it is more economical to build and operate.

Another variation is the "long lens spectrometer" introduced by Siegbahn (1943) and used by Slätis and Siegbahn (1949) and by Agnew and Anderson (1949). Spherical aberration is reduced by this design.

It may be pointed out that to distinguish between positive and negative electrons in the lens type spectrometers it is necessary to insert helical baffles which select only particles travelling with the correct direction of rotation.

The lens type spectrometers, in general, give a higher transmission at the expense of resolving power. The relation between intensity and resolution, R, is of the same form as before, i.e. the former is proportional to $R^{5/2}$. This type of spectrometer appears most useful when relatively weak sources are being handled, but for precise work the semi-circular spectrometer or the double focusing instrument (about to be described) is to be preferred.

Double Focusing Spectrometer.

The semi-circular spectrometer provides focusing in the plane perpendicular to the magnetic field only. It would obviously be of value if rays emitted at an angle to this plane were also focused to the same point as the particles in the plane. Svartholm and Siegbahn (1946) showed that a magnetic field of the type used in betatron design would have the desired property. The field has the form

$$H = H_0 \left(\frac{r_0}{r}\right)^n, \qquad 0 < n < 1.$$

If $n = \frac{1}{2}$, the radial and axial frequencies are equal and the image point is at an angular distance of $\sqrt{2}\pi$ from the object. Shull and Dennison (1947 a, b) have considered the field expression in the more general form

$$H = H_0 \left[1 - \alpha\left(\frac{r-r_0}{r_0}\right) + \beta\left(\frac{r-r_0}{r_0}\right)^2 - \dots\right]$$

For double focusing, α must be $\frac{1}{2}$, to provide radial and axial focusing. If β has the value $1/8$, second order radial aberration is eliminated, if $3/8$, second order axial aberration is eliminated. The compromise value of $\beta = \frac{1}{4}$ appears to give the smallest image.

This type of spectrometer has several advantages. It has a resolving power as good as that of the semi-circular instrument but has a much better transmission. A resolution of 0.3% and a transmission of 0.3% are easily obtained. The efficiency, which is about unity, is higher than that of the other types. It has twice the linear dispersion of the semi-circular instrument of the same radius. There is, however, the difficulty of obtaining the correct shape of pole piece and, unlike the lens spectrometers, the relation between current and momentum is not linear, though, of course, the field-momentum relation is.

Spiral Orbit Spectrometer.

Another interesting magnetic resolver is the so-called "Spiral Orbit" spectrometer developed principally in Japan and the University of California. (Miyamoto, 1942,1943,Sakai,1950, Sagane and Giles,1951) and used by Sagane, Gardner and Hubbard (1951) to measure the energy spectrum of the electrons from μ-meson decay. In this apparatus, particles are emitted from a source in the centre of circular pole pieces, the field strength between which is non-uniform in such a way that particles of a given momentum are focused in a "stable orbit". Since the particles can leave at any angle in the horizontal plane and with a fairly large vertical angular spread, the solid angle or transmission, is very high indeed. Typical figures quoted by Miyamoto for an actual spectrometer are given below

$H\rho$ (gauss-cm)	1666	1925
$\Delta H\rho / H\rho$	7%	5%
Solid Angle	5.8%	5.8%

The theory shows that, provided the area of the source is small, or the diameter of the magnet is large, the efficiency is higher than for the other types of spectrometers.

Each type of spectrometer has its advantages and disadvantages. These have already been pointed out, but one may say that the semi-circular spectrometer is the simplest in principle and is best suited for absolute energy measurements. Thus it is likely to remain the basic instrument in use. The lens spectrometer is useful if a large transmission is required though at the expense of resolution. The double focusing instrument gives a resolution comparable to that of the semi-circular spectrometer but has a higher transmission. In comparison with other

techniques, the magnetic methods are precise, can be used at high energies but require relatively strong sources.

Before leaving the subject of magnetic spectrometers, mention should be made of the pair spectrometer used to measure the energies of gamma-rays in the energy region above ~5 MeV (Walker and McDaniell,1948). In this region, the most important interaction of gamma-rays with matter is pair production and if the sum of the momenta of the pair electrons is constant, so is the sum of the energies and, therefore, the energy of the original photon giving rise to the pair is constant. This property is utilised by the spectrometer in the following way. Gamma-rays are allowed to produce pairs in a thin sheet of metal of relatively large area. The positive and negative electrons of each pair travel through an angle of 180° in opposite directions under the influence of a magnetic field. Banks of counters are situated on either side and a coincidence circuit is arranged in such a way that all coincidences due to the two electrons of each pair, which have a constant energy sum, record on a given scaler. With the eight counters of Walker and McDaniell (four on each side) it was possible to select and record seven different energy intervals simultaneously. The instrument build by Rae at Glasgow University has five counters on either side giving nine channels. The coincidence circuit and counters are arranged so that <u>any</u> pair of counters can be used to record a given energy pair, provided they have the correct separation = $2(\rho_1+\rho_2)$ where ρ_1 and ρ_2 are the radii of curvature of the positron and negatron respectively. This means that a large area of radiator can be employed.

IV. Electrostatic Spectrometers.

The simplest form of electrostatic spectrometer is that used by Watt and Williams (1946) to determine the end-point energy of the tritium beta-spectrum. A voltage between the source and the counter opposed the passage of electrons from one to the other and this voltage was increased until no particles were able to get into the counter. Actually, since the window thickness was ~12 KeV (measured by accelerating electrons from a filament to a sufficient energy just to penetrate the window) they had to use as "zero" voltage, a value of + 12 KeV. This method gave an end-point energy of about 11 KeV instead of the more recently accepted value of ~18 KeV. Evidently it is not an accurate method, at least in this energy range. Also it cannot be used conveniently for higher energies because of the high voltages necessary. It does not give the spectrum shape since the electrons travel at different angles relative to the field and only the component of velocity parallel to the field is measured.

It is possible to build an electrostatic spectrometer on the lines of the magnetic instrument, but as it possesses no particular advantage over the latter there does not appear to be much point in constructing such an instrument. The field, and therefore voltage, across the deflecting electrodes must be very high. However such instruments have been used by Backus (1945) in the study of Cu^{64} and by Rogers, McReynolds and Rogers(1940) in the measurement of m/e and v for various electron lines of RaB. The theory of the instrument has been given by Hughes and Rojansky (1929). They show that, using cylindrical section electrodes, the best focus is at $127^{\circ}11'$ and not at 180° as in the case of the magnetic spectrometer.

However a considerably modified form of the instrument of Watt and Williams has been built by Hamilton and Gross (1950) and used by them in

their study of the low energy (0-30 Kev) region of the S^{35} spectrum (Gross and Hamilton, 1950). Here the source was placed at the centre of a sphere and the particles passed through a variable retarding potential between two spherical grids concentric with the outer conductor which acted as a collector for the electrons. The current arriving at this electrode was measured as the retarding potential was increased to its maximum of 30 KeV. This method eliminated the angular distortion effect of the Watt and Williams apparatus. The transmission, or solid angle, was very high ($\sim 2\pi$, not 4π, since the collector and grids were actually hemi-spherical) but this was offset by the fact that current, rather than individual particle, detection was employed.

V. Proportional Counters.

The use of proportional counters for the examination of beta-spectra, X-rays and soft gamma-spectra was started in Glasgow in 1947. This technique has several great advantages, as follows: (i) It is very well suited to the examination of spectra at low energies, since sources can be put inside the counter thus avoiding window difficulties. Also radiations of energy <1 KeV can be measured with considerable accuracy as will be shown later, provided an extremely thin source and source backing is employed e.g. a gaseous source. (ii) Source and source mount thickness difficulties have often been avoided by the use of the source in a gaseous form. (iii) The solid angle is always very high, being usually 2π or 4π, generally the latter. This permits its use for weak radioactivities which could not otherwise be analysed. (iv) A large source area can be utilised, $\sim$ 1000 cm^2 in the bigger counters, so that even 1 gm of source yields a source thickness of only 1 mgm/cm^2. (v) The fact that the counter is operating as a spectrometer has a great advantage over methods using

Geiger counters, with regard to "background". The "background" spectrum of a large counter operating at high pressure (~5 atmospheres) is spread out, more or less uniformly, over a range of energy ~500 KeV or more. Thus if one considers a K-capture source whose radiation is concentrated in a range of less than say 5 KeV, the sensitivity of the counter is ~100 times better than in the case of a Geiger tube. The same is true for gamma- or X-radiations or for conversion electrons. This means that the presence of a weak source giving rise to such radiations might give no appreciable increase in counting rate over the "background" value but still give good and easily measured peaks. This was the case, for example, in the discovery of Ni^{59}, as will be described in Part 2. In the case of beta-spectra, the position is somewhat altered but is still very favourable. For example if the beta-spectrum is spread over a range of 50 KeV, the ratio of counting rate with the source present the rate with it absent is increased by a fraction ~10 and if one considers only the region near the end-point, the factor is much greater. There are other, less important, advantages some of which will appear in the work about to be described. More recent work in Glasgow has extended the facilities of these counters in two ways (a) the extension of the upper energy limit which can be measured and (b) the elimination of "end-effect". As the writer employed the proportional counter technique to a great extent in the work described in this thesis, it is proposed to postpone further discussion of it until the remaining two techniques have been described.

VI. Scintillation Counters.

The scintillation technique may be considered to be a development of one of the earliest methods of detecting radiations, viz. the flourescent screen, which was first used to detect X-rays and cathode rays. However,

combined with a microscope, it was shown to be capable of detecting single events, for example the arrival of individual alpha-particles. Much of this work is associated with Lord Rutherford who, for example, used the method to detect the protons arising from the first artificial disintegration of the nucleus (Rutherford 1919). In the modern technique the scintillations arising from some phosphor, instead of being observed by eye, are detected by the very sensitive photomultiplier tube and recorded on a scaler or displayed on an oscilloscope screen. In 1944, Curran and Baker (1944, 1947) first used this technique to detect alpha-particles. Zinc sulphide, used by these workers, has a high efficiency for the detection of alpha-particles provided the sulphide layer is thin. This is necessary since zinc sulphide absorbs the radiations which it emits. The use of larger crystals which did not have this disadvantage has provided a very useful tool in nuclear physics. If it is large, and transparent to its own radiations, particles can be wholly stopped in the crystal giving a light pulse the size of which is a function of the energy of the particle. Also higher energy gamma-rays can be detected with an efficiency unapproached by gas counters, because of the increased stopping power of the crystal.

There are several important properties which a phosphor used for scintillation counting should possess. It should be possible to grow large, uniform crystals which have high efficiency for the conversion of particle or gamma-energy loss into radiation in a spectral region in which the photomultiplier tube detects efficiently. It should have a short resolving time(short afterglow), high transparency to its own flourescent radiations, good stability and, for some purposes, high density and containing an element of high atomic number.

Crystals having the desired properties have been developed, notably

anthracene, naphthalene, stilbene, sodium iodide (thallium activated) etc. The first three have very short resolving times ($<10^{-8}$ sec) and are very useful in the detection of electrons and soft gamma-rays. Because of the high atomic number of iodine and the high density of sodium iodide this material is very efficient for the detection of high energy gamma-rays. It has a resolving time of $\sim 10^{-7}$ sec. Also the phosphor may be in the form of a solution, in a suitable solvent, of a flourescent material.

It is obviously an advantage if the phosphor responds linearly to increasing energy. Hopkins (1951) showed anthracene is linear in the region 100 KeV-1MeV but not at lower energies. West, Meyerhof and Hofstadter (1951) showed sodium iodide is linear from 2 KeV - 500 KeV, while the work of Bannerman, Lewis and Curran (1951) showed that it is linear to $2\frac{1}{2}$ MeV.

A review of work done using NaI(Tl) crystals has been given by Hofstadter and McIntyre,(1950a) . Bannerman et al. give a useful discussion of different ways in which the scinillation technique can be used. It has been used more for the detection and measurement of gamma-radiations than for beta-rays though uses in both fields will be discussed. Gamma-rays interact in three ways. (i) Photoelectric effect. This effect, which is important at energies, $\gtrsim 0.25$ MeV, gives a single peak if moncenergetic photons are captured. High Z materials are more efficient than low Z. (ii) Compton process. The electrons scattered by this process have a wide range of energies with a well defined upper energy limit but are homogeneous in energy for a given direction, a fact that is made use of in one method of examining gamma-rays. (iii) Pair production. This process becomes possible at energies exceeding 1.02 MeV and is of

importance at energies greater than ~2 MeV.

The technique can be used down to fairly low energies but does not compete with the proportional counter in the low energy region due partly to the high background counting rate, (which can be much reduced by working at low temperatures, using liquid air or solid carbon dioxide etc.) and also due to the poor resolving power expected on the basis of the relatively low number of primary electrons released at the photo-sensitive cathode.

Beta- or gamma-spectra are most simply measured by the single crystal technique. In the case of beta-rays the source can be held near the crystal or even imbedded in it as was done by Bannerman in the examination of Hg^{203}. Good (1951) used a potassium iodide crystal (an excellent phosphor) to investigate the shape of the beta-spectrum of K^{40}. This was similar, in principle, to the technique of using radio-active gases in proportional counters. Probably this would be a good method of investigating the natural, weak activity of Rb^{87}. The crystal should not be too small in order to minimise the effect on the spectrum shape due to the loss of electrons emitted near the surface. Perhaps the liquid scintillation technique might be applied here.

In the case of gamma-rays it is usual to estimate the energy from the "Compton edge" of the energy distribution in the case of low Z crystals. If sodium iodide is used, a good photoelectric peak can often be obtained which yields a more accurate value. It is customary to calibrate with sources emitting one, or a small number, of gamma-rays e.g. Co^{60}. To count beta-rays against a background of gamma-rays, one can use a very thin anthracene crystal which will not capture gamma-rays efficiently but will record every incident electron.

If a radioactive isotope possesses several gamma-rays it is very

difficult, or impossible, when using the single crystal technique, to distinguish between the peaks due to the different gamma-rays because of the overlapping Compton distributions, one of which exists for each energy. This is especially so if the energies are not widely separated. In such a case, and indeed in all gamma-ray work, a method described by Hofstadter and McIntyre (1950b) is useful. It makes use of the fact that the electrons associated with photons scattered by the Compton process at a particular angle are homogeneous in energy. Also at angles near 180°, the rate of change of energy with angle is small. For example, between 150° and 180°, the energy increases by ~2% for a gamma-ray of 1 MeV. If a collimated beam of gamma-rays is allowed to impinge on a crystal I and the photons scattered at a particular angle, usually as near 180° as possible, are detected by a second crystal II (shielded from the direct gamma beam), then the pulse distribution of electrons in I, which are in coincidence with the gamma-pulses in II, will give well defined peaks corresponding to each gamma-ray energy which stand out clearly with little or no background since there are no longer continuous overlapping Compton distributions. The energy of the gamma-ray is easily calculated from the electronic energy. Hofstadter and McIntyre used this technique to measure the gamma-rays emitted by Co^{60}.

The usefulness of the method in coincidence studies has already been mentioned and will be obvious for three important reasons. Firstly, its efficiency for gamma-ray detection, compared to a Geiger counter, can increase the sensitivity of the technique by a factor of from ten to a hundred. Secondly, the fact that the scintillation counter is also a spectrometer, means that a particular energy interval can be selected by kick-sorter and only the gamma-rays (or beta-rays) in coincidence with the

particular event selected, are studied. Also the resolving times of scintillation counter coincidence arrangements can be made very much smaller than for the Geiger counter case, because of the much shorter resolving times of the crystals. This cuts down the random coincidence rate, permits the use of stronger sources and allows one to measure much shorter lifetimes of excited states. It is not intended at this point to go into any detail regarding coincidence techniques as these have already been mentioned. However Bannerman et al. have discussed some of the uses of the scintillation detectors in this field. They also suggest an interesting application of the scintillation technique to the "pair spectrometer". The two arrays of counters are replaced by two large crystals or liquid phosphors the output voltages of which are fed to a fast coincidence circuit and the amplitudes added. If the spectrum of the added amplitudes is analysed, the gamma-ray spectrum is obtained. This method avoids the complicated circuitry associated with the Geiger counter method and, too, is equivalent to the use of a very large number of Geiger counters, the accuracy being limited mainly by the resolving power of the scintillation counters.

The integrating properties of the crystal technique are similar to those of the proportional counter. If a source is placed inside a crystal the energies of all events in coincidence are added and this can be a considerable advantage. It is intended to discuss this matter at some length in the section dealing with the radioactivity of Hg^{203}, since the property was used to good effect in that case, with both proportional and scintillation counters.

To sum up, the scintillation spectrometer technique is very sensitive, has a high efficiency for the capture of high energy radiations, has a short

resolving time and, using "light pipes" can be put in confined spaces. On the other hand, at low energies, it does not compare favourably with the proportional counter, to which it is closely akin, in resolving power, accuracy of measurement or background counting rates.

VII. The Electron Sensitive Photographic Emulsion.

The development of the G5 emulsion by Ilford Ltd., the NT2a and NT4 emulsions by Kodak, Ltd. and the NTB3 emulsion by Eastman Kodak, Inc., all of which are electron sensitive, suggests new ways of examining beta-radiations. Three techniques suggest themselves.

Firstly, one may replace the plate of a magnetic plate spectrograph by such an emulsion. By counting the number of electrons actually recorded at different parts of the plate, an accurate measurement of relative intensities can be made. It will be recalled that the measurement of the density of an ordinary photographic plate does not give accurate intensity values. Also the method is more sensitive since each individual electron is counted, as in the counter type spectrometer. If a permanent magnet spectrograph were used, a plate might well be left in it for several weeks before the increasing background became too serious, especially if the apparatus were stored underground.

Secondly, one might place a thin layer of a weak radioactive source over the surface of the emulsion and measure the track lengths produced by the decay electrons. Alternatively the plate might be soaked in a weak solution of the source, provided this had no deleterious effect on the properties of the emulsion. This would eliminate source thickness difficulties and would be equivalent to the use of a radioactive gas in a proportional counter. The chief advantages of this method are:(a) Its great sensitivity. Calculations by the author show it to compare favour-

ably with the most sensitive of the other techniques and he is investigating its possibilities. (b) Its simplicity. However it has several disadvantages, principally that the resolving power is not likely to be better than ~20% due to range straggling and scattering. Also the analysis is tedious though probably not much more so than in some of the other techniques. A. Bonetti (1950) has actually used such a technique in the examination of Rb^{87}. He used a 0.05 gm/cm^2 source exposed 0.3 mm above a 200 micron G5 plate for seven days. He got an end-point not inconsistent with that obtained by Libby, i.e. 130 KeV, but pointed out this could be low due to (a) poor geometry (b) thick source (c) high background - he used one month old plates. He suggests selecting electrons going in a particular direction and using fresh plates with emulsions of thickness up to 1 mm, or even more. In order to relate the range of the electron to its energy, the results of Zajac and Ross (1949) may be used. This method would be specially useful for weak long-lived sources such as those in which the writer is interested.

Finally the technique at present under investigation by Reynolds at the Radiation Laboratory, University of California, is of interest. A source containing gravimetrically minute quantities of one or more activities is analysed by a mass spectrometer. The plate on which the separated masses fall is pressed against the surface of a G5 plate. The radioactive isotopes show up as lines of electron tracks on the plates and he hopes to analyse the ranges and hence find the maximum energy of the electrons for each active mass. His first results seem promising.

VIII. Other Methods of Measuring Gamma-Ray Energies.

Brief mention should be made of two other methods of measuring gamma-ray energies. The method of diffraction by a crystal is well known.

Usually, now-a-days, a curved crystal and photomultiplier detector are employed since the focusing property of the former and the sensitivity of the latter make the technique much more sensitive. Nevertheless, it requires a very strong source and cannot be used much above 1 MeV. However, in the region in which it is useful it is a very accurate technique.

Also, the nuclear photo-effect has been used for the measurement of high energy gamma-rays. Deuterium is commonly used for gamma-rays between 2.2 MeV (threshold) and ~10 MeV (at which energy other processes begin to take place and also the cross-section falls off). Gibson et.al.(1947) impregnated an Ilford nuclear plate with heavy water and measured the ranges of the protons produced in the plate due to gamma-radiation from the (p, α) reaction in flourine.

This concludes the review of the various methods of analysing the radiations emitted by radioactive sources. Now follows a fuller description of the proportional counter technique.

2. The Proportional Counter Technique.

The earliest form of proportional counter was that devised by Rutherford and Geiger in 1908 (Rutherford and Geiger,1908). This might be considered as a development of the ionization chambers, or electroscopes, which had been used previously. Their apparatus consisted of a cylindrical cathode inside which was a co-axial anode wire. When a sufficiently high voltage was applied across the wire and cylinder it was found that the entry of a single alpha-particle gave an observable deflection on the quadrant electrometer used as a measuring instrument. It was found possible to count alpha-particles at a rate of three to five per minute. The technique was later improved when Geiger and Rutherford (1912) replaced the quadrant electrometer by the faster and more sensitive string electrometer. The

pulses were recorded on photographic film at rates up to 1,000 per minute. These counters had the property, often useful, of being able to detect alpha-rays in the presence of intense beta- and gamma-ray background, because of the larger pulses of the alpha-particles. Further development by Geiger and Klemperer (1928) showed that, by raising the voltage sufficiently, pulses due to beta-, as well as alpha-, particles could be obtained. Provided it was not raised too much, beta- and alpha-particles could still be distinguished. In the period up till 1948, proportional counters were used mainly for the detection of alpha-particles, other energetic nuclei and neutrons. In 1948, Curran, Angus and Cockcroft (1948, 1949a) extended the technique to beta-ray energy measurement. A good description of early work in this field is given by Rutherford, Chadwick and Ellis (1930). More recently Lewis (1942) and, especially, Korff (1946) and Curran and Craggs (1949) have written very useful and detailed monographs which deal fully with proportional counter theory and technique. A very readable review by Curran (1950) presents a clear picture of the development of the present technique and how it is applied to the measurement of beta- and soft gamma-ray energies. Also Korff (1950a, 1950b, 1951) has reviewed the technique in three articles in "Nucleonics". The first deals with the theory of the proportional counter, the second with its design and use while the third deals with the associated circuits, especially the amplifier. An earlier review was given by the same author (1942) and a general review of Corson and Wilson (1948) includes a section on the proportional counter as does the book "Ionization Chambers and Counters" by Rossi and Staub (1949). The theory of the action of the proportional counter was given originally by Rose and Korff (1941) and is given fully by Curran and Craggs (1949).

3. Description of the Gaseous Discharge.

Let us consider the basic type of counter consisting fundamentally of a cylindrical cathode and co-axial anode wire. Fig. 1, which shows the type of counter used in early work in Glasgow, will indicate what is meant. Here the cathode consists of a thick aluminium foil rolled into a cylindrical form and placed inside a cylindrical glass vessel. If a variable e.m.f. is applied between the cathode and the anode of the counter, inside or near which is placed a source of radiation, and if we measure the current or charge collected by the wire in a given time as the e.m.f. increases, the following events take place.

I. The current increases with voltage, because as the field strength increases, the ions formed inside the counter by the radiation travel faster (have a higher mobility) and suffer less recombination.

II. The current then becomes stationary as voltage increases. This saturation effect is due to the fact that recombination is now negligable and the current is now equal to the number of ions originally formed per second. The ionization chamber operates in this region. Such a chamber can be used to measure the current produced by a source of radiation, or the charge collected in a given time if the source is weak. It can even be used to detect the passage of single particles of high ionizing power if it is connected to a very sensitive amplifier. The size of the pulse, dV, appearing on the central wire is given by

$$dV = ne/C$$

where e is the electronic charge, 1.6×10^{-19} coulombs, C is the capacity of the chamber and associated circuit and n is the number of ion-pairs formed. The height of the pulse produced is usually governed by the number of the electrons only since they possess a much higher mobility than

do the positive ions and are therefore collected in a much shorter time. A simple calculation, taking C as $\sim 10\,\mu\mu F$ shows that an alpha-particle from RaA (6.0 MeV) will give rise to a pulse of about 2.7×10^{-3} volts which is well above the noise level of a good amplifier, say, $\sim 10^{-5}$ volts. Thus the passage of a densely ionizing particle is fairly easily measurable. However it would be very difficult to measure the pulses produced by beta-particles passing through the chamber.

III. If the voltage is further increased, the current, or pulse height due to individual events, starts increasing again, due to ionization by collision. In a cylindrical counter the field increases rapidly as the distance from the wire decreases. The equation relating the field strength E_r at a distance r is

$$E_r = \frac{V}{r} \times \frac{1}{\log_e r_2/r_1}$$

where V is the applied voltage, r_2 and r_1 are the radii of the cathode cylinder and of the wire respectively, In the case of a proportional counter having, typically, r_2 and r_1 equal to 2.75" and 0.002" " respectively and operating at a typical voltage of 4000 volts, the field varies from 79.7 volts per cm at the cathode to 96 Kilovolts per cm at the wire. The threshold of the proportional region is the voltage at which an electron is able in its last mean free path length before hitting the wire, to cause an ionizing collision. As the voltage increases further, electrons commence making such collisions at greater distances from the wire. Each collision provides an additional electron which in turn can make further ionizing collisions. The average number of electrons collected at the wire due to a single initial electron is called the gas amplification factor and is usually denoted by the symbol A. The voltage of the pulse on the wire is given by

$$dV = Ane/C$$

where the other symbols have the same meaning as before. In the ionization chamber region, A has the value unity. A usually lies in the region $1-10^4$ for accurate proportionality, provided suitable gas mixtures are used. Practically any gas, or gas mixture, will give satisfactory proportionality up to a value of A of 100. The property of the cylindrical counter, that the field which is strong enough to cause multiplication of electrons by ionizing collisions exists only at very short distances from the wire, is a very useful one for true proportional counting. It means that electrons of any given energy originating in any part of the counter apart from an infinitesimal volume around the wire form the same number of electrons by multiplication. This property is shared by a spherical counter, having a small spherical ball at the centre to collect the electrons. However, the cylindrical form is the easier to realise in practise. In the case of a parallel plate counter, it is obvious that the pulse size will be bigger, the further the original electron is from the positive plate.

This region is called the "proportional region" because the output pulse, dV, arising from the initial formation of a number of ions, n, is accurately proportional to n. In other words, a proportional counter not merely distinguishes between particles of different ionizing powers, i.e., alpha-particles, protons, electrons, etc., but can also be used to measure energies. The method can be extended to electrons, even of very low energy, as can easily be demonstrated by calculation. Consider the case of a well designed amplifier having a noise level at the first grid of, say, $\sim 2\text{-}3 \times 10^{-5}$ volts. Assuming that one can measure fairly accurately the energy of a particle giving rise to a pulse of approximately three times that value, that the capacity of the system is $\sim 10 \mu\mu F$ and that the gas amplification is ~ 100, the energy of the corresponding radiation is only

~ 1.5 KeV. By increasing the gas amplification to 10^4, as can easily be done by the use of suitable gases, an energy of only 15 eV is obtained. Of course such an electron could not produce even one ion-pair, but this shows that the method is capable of detecting single electrons. Indeed Curran, Cockroft and Angus (1949) measured the pulse distribution from single slow electrons liberated by light from the aluminium cathode of a proportional tube. This gave information regarding the statistical fluctuations in the gas amplification. They also showed that it is possible to obtain gas gains in excess of 10^5 and remain within the proportional region, provided the initial amount of ionization is small.

IV. If the voltage is further increased the counter operates in the so-called "region of limited proportionality". The value of A is about 10^4-10^7, though these figures depend on the gas employed. In this region, the pulse height is no longer proportional to energy, since A does not increase so rapidly for energetic as for less energetic particles. This is mainly because the positive ion sheath begins to have an appreciable effect. Also the gain goes up more rapidly because of liberation of photoelectrons at the cathode, electrons produced by positive ion bombardment of the cathode and ionization by the positive ions. This region can still be employed usefully to differentiate between particles of widely different energies.

V. Further increase of voltage brings us to be "Geiger region". Here pulses of constant size arise from all events which ionize the gas in the counter-even if only one ion pair is formed. In this region very great sensitivity is achieved, but all discriminating power, except by external means, is lost.

VI. Finally, if the voltage is increased beyond the end of the Geiger "plateau", where the counting rate stays roughly constant with increasing voltage, the counter enters a "region of continuous discharge". (Actually

the discharge is not really continuous, but consists of a large number of little pulses. In it the value of A generally exceeds 10^8). Finally it should be noted that these regions are not sharply defined but pass gradually from one to the other.

4. Theory of the Proportional Counter Action.

As has previously been mentioned, the theory of the action of the proportional counter has been developed by Rose and Korff(1941). On the basis of certain assumptions regarding the nature of the discharge, which will be mentioned briefly below, they derived an expression for A, the gas amplification factor. Since the derivation is given fully by Rose and Korff and also by Curran and Craggs (1949) it is not proposed to repeat it here. In the form given by Curran and Craggs, the expression runs:-

$$A = \exp\left\{2\left(\frac{V_A a r_1}{\log_e \frac{r_2}{r_1}}\right)^{1/2}\left[\left(\frac{V_A}{V_1}\right)^{1/2} - 1\right]\right\}$$

where V_A is the total voltage across the tube, V_1 is the voltage at which multiplication by collision begins, r_1 and r_2 are the radii of the wire and cathode cylinder respectively and a is the rate of increase of ionization cross-section with energy in cm^2 per volt.

The assumptions made are as follows:- (i) The positive ions formed in the discharge move in a field which is usually low and which decreases as they move. They do not therefore contribute appreciably to the number of ions formed by collision. They travel very slowly compared to the electrons and give rise to a space charge. In the proportional region this effect is small and is neglected in the theoretical treatment. (ii) Recombination of electrons and positive ions can be neglected since this is a very small effect. (iii) Statistical fluctuations in the number of ions formed in a given length of path of a particle of given energy are neglected. (iv) Strongly electro-negative gases and vapours, such as

O_2, Cl_2, Br_2, H_2O etc. capture electrons to form negative ions. The movement of such ions is very slow and the whole character of the avalanche is altered if such gases are present. It is assumed that this is not the case. (v) The effect of photons formed in the avalanche can be neglected. Especially when complex molecules are present, is this justified. If the effect is present, it gives rise to larger pulses than would otherwise be the case. (vi) The effect of secondary emission at the cathode surface due to positive ion and photon bombardment is neglected.

(v) and (vi), if present, tend to increase A, while (i), (ii) and (iv) tend to decrease A. The effect of (1) has been discussed by Montgomery and Montgomery (1940). The region of limited proportionality, where A does not increase as rapidly for high energy events as for low, is probably due to the voltage shielding effect of the positive ion space charge. The effect of (vi), secondary emission, becomes of importance in the case of monatomic or diatomic gases if A rises much above 100. However polyatomic gases do not show the effect, which appears to be due to the formation of metastable states in the noble and other gases. Such states may have lifetimes of $\sim 10^{-3}$ sec. instead of the normal lifetime for an excited atomic state, viz. $\sim 10^{-8}$ sec. Such atoms release photo-electrons from the wall with a relatively high probability ($\sim$ few per cent) and they cannot normally lose their energy by collision with similar atoms or molecules. However if a polyatomic gas is mixed with the gas the effect is much reduced, or even eliminated because the excitation energy is transferred by collision to the polyatomic molecule which predissociates instead of radiating or de-exciting at the wall. In our experiments, argon and methane was used; or, occasionally, argon and nitrogen. There are two slight disadvantages involved in using polyatomic gases (a) the operating

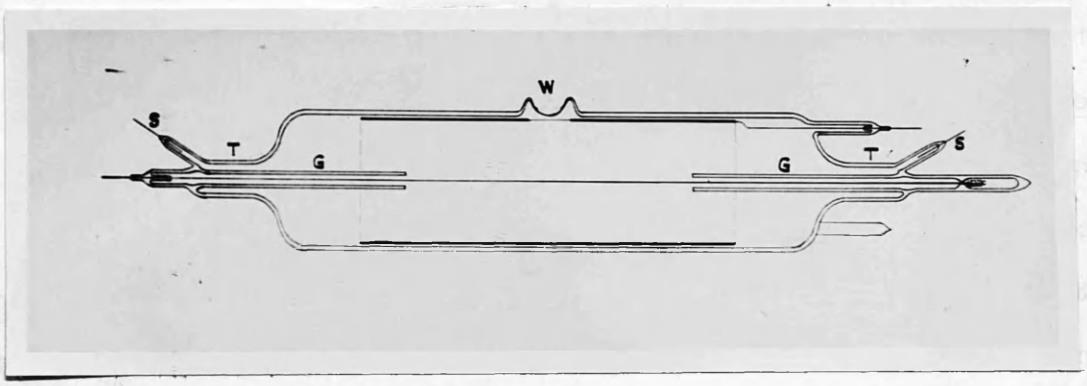


Fig.1. A glass-type proportional counter.

voltage is higher and (b) the life to the counter is limited, since the polyatomic molecules are dissociated in absorbing the excitation energy. As in this research it was customary to refill the counter before each experiment, the latter disadvantage was of no importance.

That the assumptions made are valid has been shown by experiments of Rose and Korff (1941) and of Curran, Angus and Cockroft (1949a). Generally, if the value of A is desired, it is measured experimentally rather than by use of the formula. In the normal use of the technique, however, one does not measure A at all but compares the pulse distribution due to the unknown radiations with that of some standard radiation as will be described in the section dealing with calibration.

5. Development of the Proportional Counter.

The counter shown in fig.1, which is reproduced by permission from the paper of Curran, Angus and Cockroft (1949a), was the type first employed in Glasgow for the examination of beta- and gamma-ray spectra, and is of a standard type. The cathode, a cylinder of sheet aluminium, is contained in a glass envelope which has a window of very thin glass W. There is a hole in the cathode at that position. The wire, generally 3 or 4 mil tungsten, is held under tension by a spring and is mounted as nearly axially as possible. The wire itself must be free of kinks and of uniform diameter. The wire is connected to the first grid of the head amplifier, while the cathode is connected to a source of high negative potential. The wire is protected by "guard Tubes", G, which are of glass covered with "Aquadag" and earthed. These prevent leakage of charge from the cathode to the wire and also keep the wire from "seeing" charged insulators, which would induce charges on the wire. The parts of the counter, T, are coated with "Aquadag" for the same purpose. A counter of this construction, if well

cleaned and pumped before filling and afterwards sealed permanently, maintained its properties for many months. However it was more usual, in the case of all-metal counters especially, to re-fill the counter before each run.

The dimensions of the counter must be large enough to prevent distortion of spectra due to the original electrons hitting the wall. With the first counters used, principally for the examination of the beta-spectrum of tritium, it was easy to ensure that no appreciable distortion arose from this cause. However as more energetic radiations were investigated, e.g. C^{14}, S^{35}, Ni^{63} etc.,it became necessary to increase the dimensions and the gas pressure. A doubling of pressure corresponds, of course, to a doubling of dimensions. This required that the counters be made of metal instead of glass. Pressures up to 5 atmospheres could be used easily. Above that pressure it became necessary to purify the gases rather carefully. Counters were built to operate at ten atmospheres. Also one can use gases of higher Z, and therefore of higher stopping power, e.g. krypton, xenon, etc. Unfortunately these gases are rather expensive, although experiments were made using xenon. Another difficulty arises in this case, however. In argon, a monoenergetic group of photons will give rise to a single peak because of the high "Auger effect", whereas in the case of the other gases named, where the effect is much smaller, there will be several peaks and the results will be more difficult to interpret.

A big advance in the energy range covered was accomplished by the use of magnetic fields to confine the particles within the gas. There are two ways of doing this. Firstly, one may place the counter in the fringing field of a magnet, the axis of the counter being parallel to the tangent to the circumference of the pole pieces, assumed to be circular. (Curran,

Cockroft and Insch, 1950). Electrons from a source situated within the counter are forced to circulate round the periphery of the poles. This method was used to examine the beta-spectrum of P^{32} which has an end-point of 1.7 MeV. The more common, though less powerful method, however, is to place the counter in a uniform magnetic field with the axis in the direction of the field, as shown in fig.31. In this arrangement, electrons with a component of velocity perpendicular to the field, which would otherwise strike the wall, are forced to travel in the direction of the field with a helical motion. Provided the path in the direction of the field is long enough, at the gas pressure used, to stop the most energetic electrons emitted by the source, no distortion due to the size of the vessel will occur. This method, due to Cockroft, is described briefly by Curran (1950). By these methods it is now possible to investigate beta- and photo-electron spectra with upper energy limits well in excess of 1 MeV.

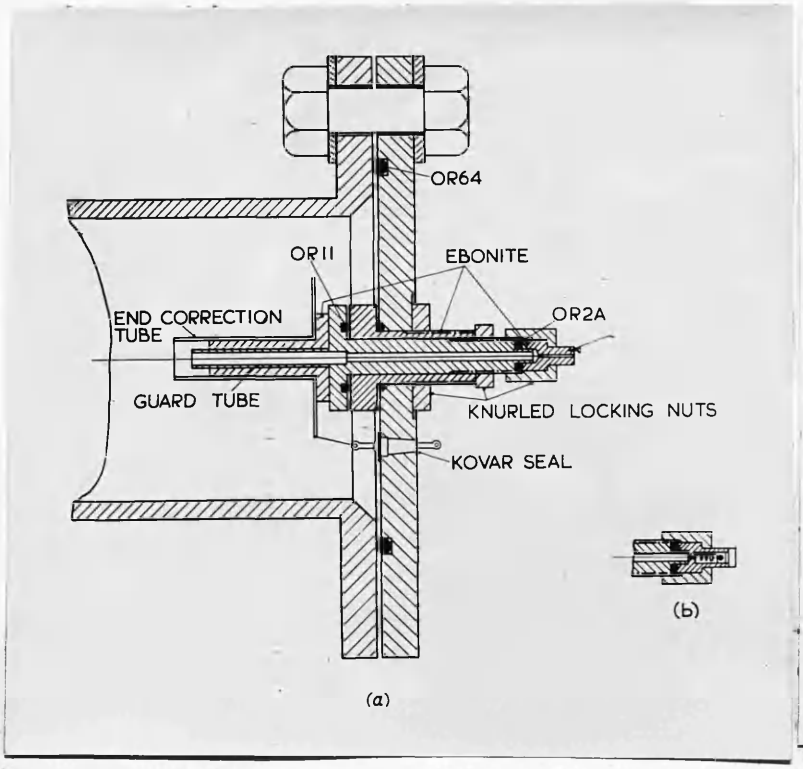
There is one unfortunate feature marking the simple proportional counter shown in fig.1, i.e. "end effect". The value of A increases from zero at the end of the guard tube to its maximum value at a distance which is usually of the order of the radius of the counter. If the source is located near the centre of the counter and the pressure is such that no particles can enter the region of non-uniform A, as was the case in the Ni^{63} investigation, or if one allows the radiations to enter by the window, this effect is of no importance. But if the source is in the form of a gas (as in the case of H^3, S^{35}, C^{14}, etc.) or if particles from solid sources can penetrate as far as the non-uniform region of the counter, it must be considered. Its effect is to give too many low energy pulses at the expense of high energy ones. There are various ways of minimising or eliminating this effect. Firstly, the counter may be made with a very large length-diameter

ratio, as in the earlier examinations of the tritium spectrum. Such error as remained was corrected by approximate calculation (Curran, Angus and Cockroft, 1949b). Another technique, introduced by Angus, Cockroft and Curran (1949) was to divide the wire into unequal lengths by a glass bead, and to subtract the spectrum obtained from the shorter portion from that obtained from the longer portion in the same length of time. This eliminates the end-effect but requires considerably more labour for the same accuracy than would be necessary if an end-corrected counter were used. The most satisfactory method of eliminating the effect is that due to Cockroft and Curran (1951) who placed "end-correction tubes" round and coaxial with the guard tubes and extending slightly beyond them. To these tubes is applied the voltage corresponding to their radial distance from the wire. The formula used is

$$\frac{V(\text{probe})}{V_A} = \frac{\log r_3/r_1}{\log r_2/r_1}$$

where r_1, r_2 and r_3 are the radii of the wire, cathode and end-correction tube respectively. In practice it is best to compare the pulse height due to monoenergetic X-rays fired through a window in the wall opposite the end of the end-correction tube with that due to X-rays fired into the middle region of the counter and adjusting the voltage till these are equal.

Two other methods have been described by Rossi and Staub (1949). In the first, which is probably applicable only to heavy particle detection, two sheets of glass placed perpendicularly to the wire, which is sealed in the glass, define an accurate end-effect-free counting volume since the potential distribution across the sheets is the same as that across the gas. In the second method the guard tubes and wire are made of equal, or approximately equal, radius. Hypodermic needle has been used for the guard tubes but this gives only approximate end-correction and the method is obviously



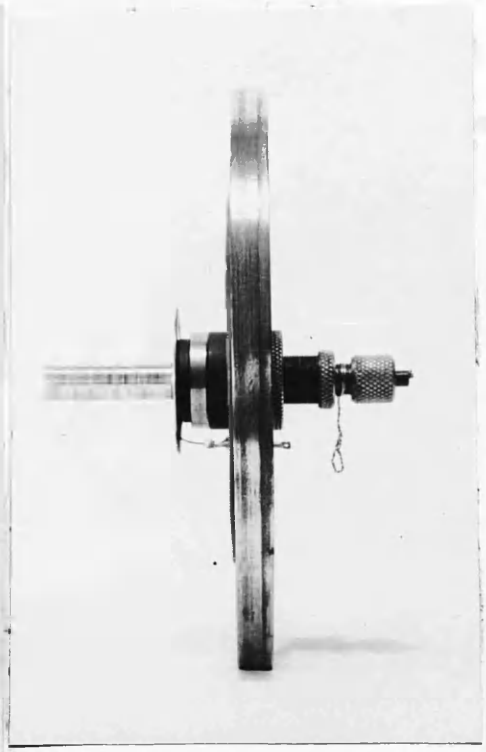


Fig. 2. Drawing of the end plate assembly of a typical proportional counter, designed to operate at pressures up to ten atmospheres.

Fig. 3. Photograph of end plate assembly.

difficult to apply.

Brief mention should be made to the "Maze" type counter in which the cathode consists of a layer of "Aquadag" on the outside of a glass envelope (Maze, 1946, Cockroft and Valentine, 1950). The construction of such a counter is simple but the main advantage is that the surface of metal exposed to the gas filling is very much reduced. Thus the counter is more easily cleaned and outgassed, and is less likely to become contaminated by radioactive sources; hence distortion of the spectrum due to source deposition on the wall is avoided. This is especially important when gaseous sources are employed. Finally the multiple wire counter, described by Rossi and Staub, should be mentioned. It provides a disc-shaped counter through which particles can be fired from end to end. The ends are two discs of wire mesh screens, while a grid of parallel wires is placed between the ends. The "drum counter" described by Cockroft and Curran (1951) could be used for similar purposes.

6. Construction of a Proportional Counter.

In fig.2 is shown a drawing of the end of a proportional counter incorporating the various improvements introduced from time to time. This particular counter was designed by the author for the study of the radiations of Hg^{203}. A 0.004" tungsten wire passes through a guard tube, through a narrow hole in an ebonite plug and is held by a small screw which is connected to the head amplifier by a stronger wire. The guard tube supports the end-correction tube. On the other end of the wire is sealed a small glass bead which presses against a spring as shown in fig.2(b). This ensures that the wire is kept taut. As this counter was designed to operate at a pressure of ten atmospheres, the end plates are securely held by six bolts and nuts at each end. The counter tube, end plates, end-correc-

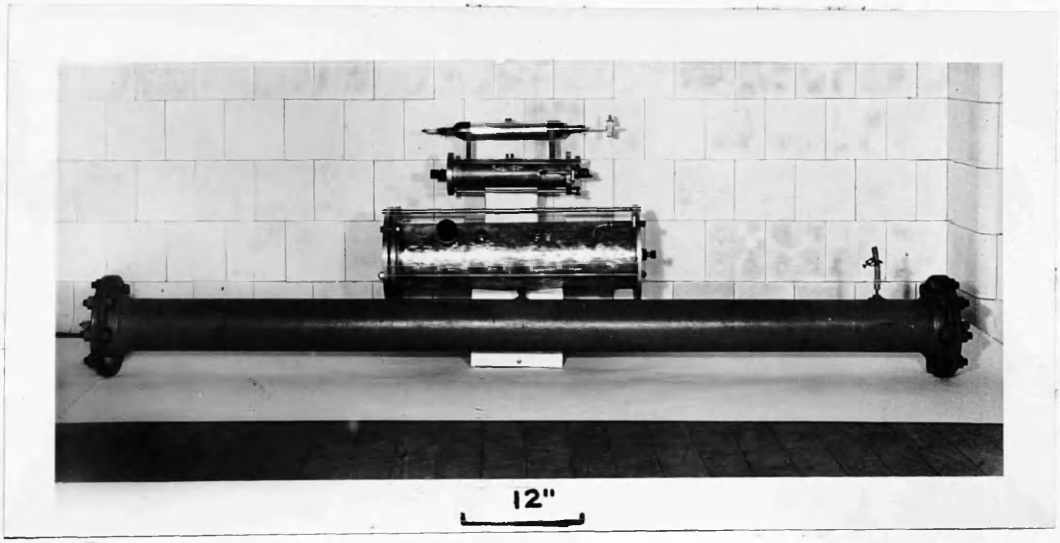


Fig.4. Various types of proportional counters.

tion tubes and guard tubes are all of steel and the other metal parts are of brass. Ebonite is used for insulation.

One important feature of the construction of this counter is that all joints are sealed with rubber O-rings. The code numbers indicated in the figure refer to the sizes. This construction has two advantages. In the more usual design of counter a certain amount of trouble was often experienced in making the counter sufficiently gas-tight. In this design, however, the counter was practically always gas-tight on first assembly. Also the time of assembly was very much reduced. A photograph of the end plate assembly is shown in fig. 3.

Finally, to give some idea of the range of size of proportional counters, fig. 4. shows four of the counters used by the author at different times. The longest is over six feet long. In each of these counters one can see clearly the windows through which are passed radiations from external sources, or X-raysfor calibration purposes. It was fairly standard to use 0.002" aluminium windows except when soft particle radiations were being examined. Then a much thinner glass window was used, as in the case of the counter shown in fig. 1.

7. Preparation of Sources.

It is always desirable, especially when soft radiations are being studied, to have a source which is as thin as possible and on as thin and low-Z support as possible. At first this was achieved by the use of gaseous sources. This proved possible in the examinations of H^3 (tritium gas), C^{14} (carbon dioxide) and S^{35} (hydrogen sulphide). However the method is by no means universally applicable, for (i) no gaseous compound may exist (ii) if one does, it may capture electrons strongly as was found to be the case with the vapour of nickel carbonyl and of, possibly, lead tetramethyl,

used in attempts to investigate the radioactivity of Ni^{63} and RaD; (iii) the gas may react with the materials of which the counter is made. When the gaseous method is employed it is important than the range of the highest energy electrons be small with respect to the diameter of the counter, since an electron which strikes the wall does not give up its full energy. This tends to give too many low energy electrons at the expense of high energy electrons.

A second method was to deposit a thin layer of the source on a thin backing material supported on a wire loop. As the reflection of electrons by the source backing tends to give rise to a type of distortion similar to that mentioned above, it is important to use backings made of low atomic number materials. Aluminium leaf ($\sim$0.16 mgm/cm^2) and nylon films ($\sim$10 μgm/cm^2) were commonly used by the writer for this purpose. The effect of the source support on the spectrum shape may be approximately estimated by use of electron back-scattering data due to Burtt (1949). The importance of using thin sources was mentioned earlier. The slowing down of electrons leaving the inside of the source gives rise to a distortion of the spectrum (Feldman and Wu, 1949). Langer, Motz and Price (1950) found that sources of thickness $<$11 μgm/cm^2 gave rise to deviations in the spectrum shape below 8 KeV.

Not only must a source be thin but it must be uniform in thickness. Langer (1949)showed that a source prepared by evaporation from a solution can easily have variations in thickness of 100:1. The best way of preparing a thin, uniform source is by evaporation of the source in vacuo, as described in the section dealing with the preparation of the mercury source. However Langer described another technique which gave sources of fairly good uniformity. This method has been much used by the writer and is as follows:

To a solution of the source is added ~5% by volume of unsulin (Burroughs Wellcome 80 units/cc). The solution is then spread over the source mount and evaporated to dryness, either in vacuo, or by infra-red heating.

Braden et.al. (1948) showed the importance of the effect of the charging of source mounts due to the emission of charged particles. In their experiments using Na^{24} the mount charged to 20 KeV in 300 hours. In a magnetic spectrometer it is essential that some means of conducting away the charge be introduced but in the proportional counter it is probable that collisions of the gas atoms with the source mount perform that duty. Nevertheless the author sometimes deposited a thin layer of carbon on nylon film supports.

In a third method of preparing sources the source material was distributed as a thin layer over a large area of aluminium foil which was then rolled in cylindrical form and placed inside the counter pressing against the wall. This method, which was used in the studies of Ni^{59}, Ni^{63}, Nd^{150} and Rb^{87} to be described later, is especially useful when the specific activity of the source is very low, as has been mentioned. However, there is one important consideration to be borne in mind when this method is used The maximum range of the particles must be fairly small compared with the radius of the counter as otherwise particles hitting the wall will not give up their full energy in the gas and this will lead to distortion of the spectrum shape. This effect will in future be referred to as the "wall effect". Since this method of supporting sources was much used in the course of this research, it was felt essential by the writer that the wall effect should be investigated mathematically. This treatment follows and it is shown that, as a rough guide, if the range of a particle of maximum energy is less than half the radius of the counter, the spectrum is not

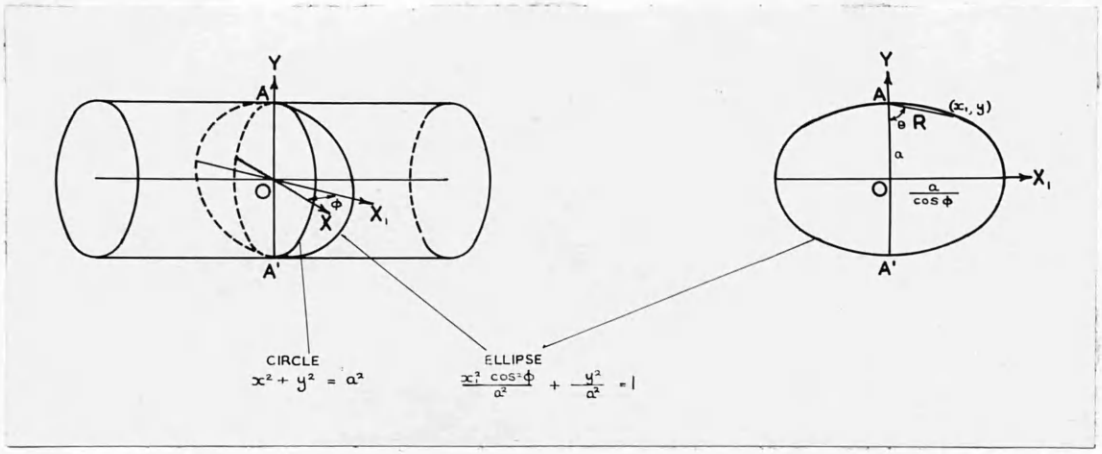


Fig. 5. Wall effect. Diagram representing a proportional counter with a source on the inside wall. A small element of area of source situated at A is considered in the text.

altered to any important extent. Also a method of correcting a spectrum thus affected is indicated.

8. The Effect of Wall Curvature on the Spectrum Shape (Wall Effect).

The first step is to find how many particles of a given range R hit the wall of the counter before giving up all their energy. Consider fig. 5(a) which represents the counter, of radius a. Consider a small area of the source situated at A and let AOA' be the perpendicular dropped from A passing through the wire. This line is taken as Y-axis while the X-axis is in a plane perpendicular to the axis of the counter as shown. Let OX' be the new axis when the plane YOX rotates through an angle ϕ. The plane then intersects the counter in an ellipse as shown in fig.5(b). Its equation is

$$\frac{x_1^2 \cos^2\phi}{a^2} + \frac{y^2}{a^2} = 1$$

$$\text{or} \quad x_1^2 \cos^2\phi + y^2 = a^2 \qquad \text{(i)}$$

Consider all particles of some fixed energy corresponding to a range R in the gas. Then, from fig. 5(b)

$$R^2 = (a-y)^2 + x_1^2 \qquad \text{(ii)}$$

We now proceed to find the value of θ such that all particles of the given energy emitted at an angle greater than θ in the YOX' plane will hit the wall. From the figure

$$x_1^2 + y^2 = R^2 + a^2 - 2aR\cos\theta \qquad \text{(iii)}$$

(ii) can be written

$$x_1^2 + y^2 + a^2 - 2ay = R^2 \qquad \text{(iia)}$$

From (iia) and (iii), it follows that

$$\cos\theta = \frac{a-y}{R} \qquad \text{(iv)}$$

Now (i) and (iia) are solved for y. From these equations

$$y^2(1-\cos^2\phi) + 2ay\cos^2\phi - a^2(1+\cos^2\phi) + R^2\cos^2\phi = 0$$

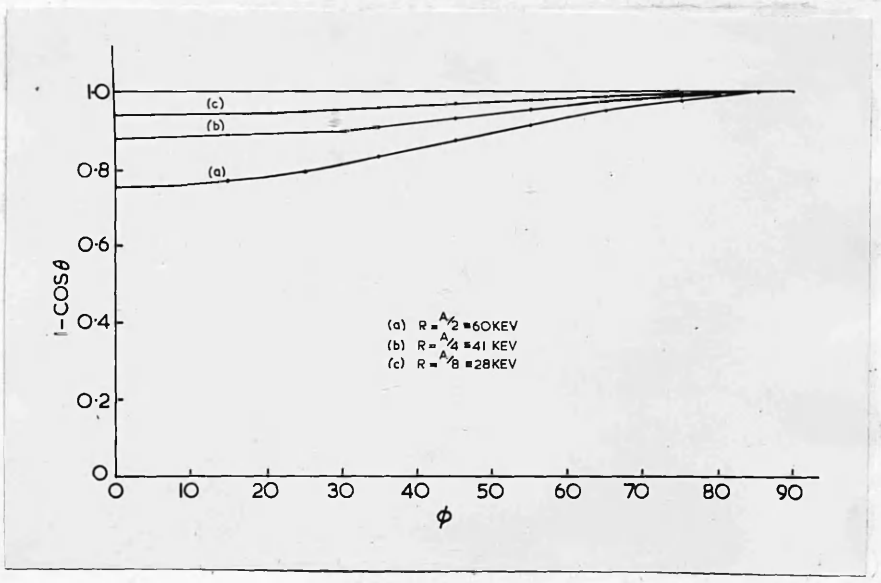


Fig. 6. Wall effect. The variation of θ with ϕ and R.

This equation is solved for y and, after simplification, we have

$$y = \frac{-a\cos^2\phi \pm \sqrt{a^2 - R^2\sin^2\phi\cos^2\phi}}{\sin^2\phi} \qquad \text{(v)}$$

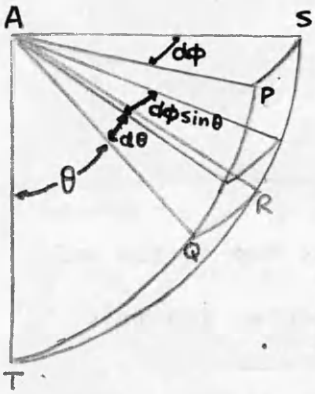
Substituting this value of y in (iv) gives

$$\cos\theta = \frac{a \mp \sqrt{a^2 - R^2\sin^2\phi\cos^2\phi}}{R\sin^2\phi}$$

Only the negative sign gives real values of $\cos\theta$, therefore

$$\theta = \cos^{-1}\left[\frac{a - \sqrt{a^2 - R^2\sin^2\phi\cos^2\phi}}{R\sin^2\phi}\right] \qquad \text{(vi)}$$

For any value of R and ϕ, this equation gives the limiting value of θ.



We must now consider the number of particles emitted into the volume between the two planes at angles ϕ and $\phi + d\phi$. The particles in the segment AQPSR of the unit sphere do not give up all their energy in the gas while those in AQRT do. We now determine the ratio of those not affected to the total number emitted i.e. the ratio of the area QRT to the area PST.

$$\text{Area of PQRS} = \iint dA = \int_0^{d\phi}\int_{\theta}^{\pi/2} \sin\theta \cdot d\theta \cdot d\phi = \cos\theta \cdot d\phi$$

$$\text{Area of PST} = d\phi$$

Therefore, the ratio

$$\frac{\text{Number of particles which give up their full value}}{\text{Total number of particles emitted}}$$

$$= \frac{d\phi - \cos\theta \cdot d\phi}{d\phi} = 1 - \cos\theta \qquad \text{(vii)}$$

where θ is given by (vi).

The way in which this ratio varies with ϕ and R is shown in fig.6 for the case of $R = \frac{a}{2}$, $R = \frac{a}{4}$ and $R = \frac{a}{8}$. These values corresponded, in the case of the counter and gas pressure used in the analysis of the radiations

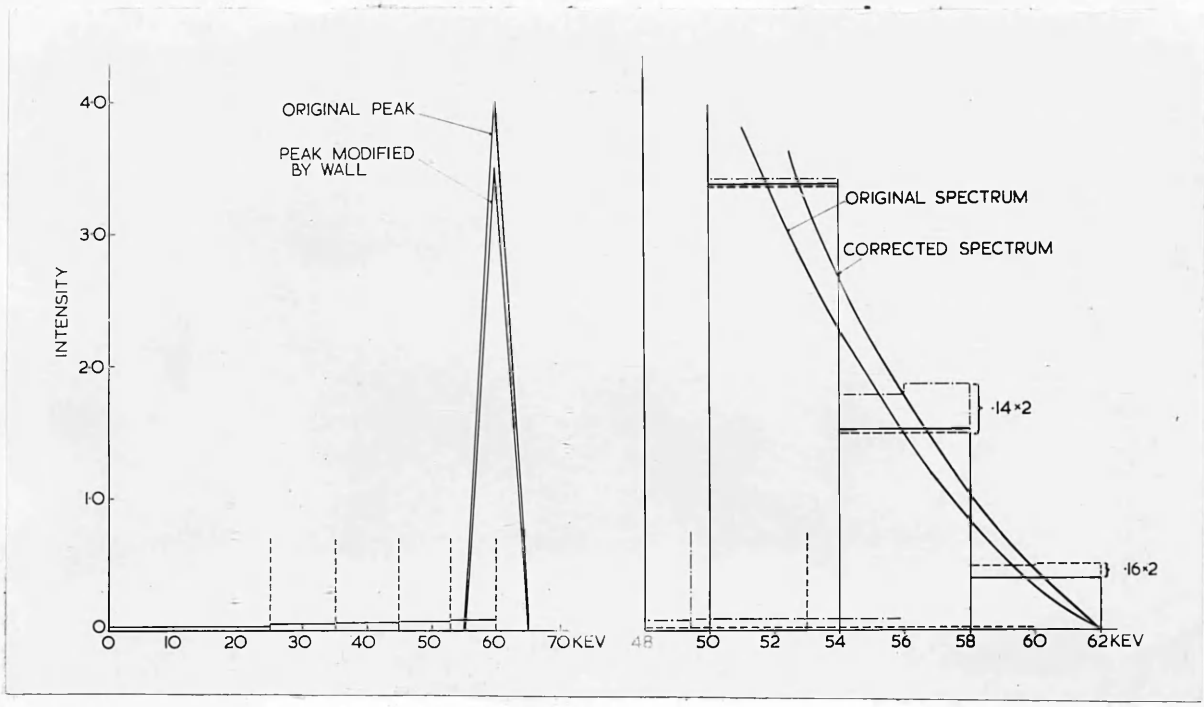


Fig. 7. (a) Distortion of an electron peak due to the wall effect. (b) Correction of a spectrum distorted due to the wall effect.

of Ni^{63}, to energies of 60 KeV, 41 KeV and 28 KeV respectively. Since most of the beta-particles have energies less than the maximum, the figure shows that the effect will affect the shape of the spectrum to a negligible extent. This will be demonstrated in a more convincing manner in the following section.

9. The Distortion of an Electron Peak due to the Wall Effect.

The effect of the wall on the shape of an electron peak will now be demonstrated. An approximately monokinetic group of electrons having a range $R = \frac{a}{2}$ (the worst case of fig.6) is shown in fig. 7(a). The corresponding energy, 60 KeV, is that calculated for the case of the counter used for the experiments on nickel, operating at two atmospheres pressure of argon, plus a little methane. For ease of calculation the line shape is assumed to be triangular. At $\Phi = 0$, (fig. 6), it is seen that one quarter of the electrons hit the wall. From the symmetry of fig. 6 about $\Phi = 45^{\circ}$, we see, that, averaging over all values of Φ, one eighth of all the electrons hit the wall. Therefore the number of unaffected electrons is represented by the lower triangle whose area is seven eighths of the original triangle. The difference in area corresponds to electrons which have given up less than 60 KeV in the gas and which go to form the low energy "tail" of the spectrum as shown in the figure. Hence the shape of the line, as modified by the wall consists of the smaller triangle and the tail. It is now obvious that, even for the highest energy involved in the case of Ni^{63}, the effect of the wall was very small.

Strictly speaking, to the left side of the triangle should be added the particles in that portion of the tail which lies between 55 and 66 KeV. However it is impossible to reproduce in a small photograph the very small additional correction that results.

10. The Correction of a Spectrum Distorted due to the Wall Effect.

Instead of starting with an unmodified peak as in the previous section, one can divide the spectrum of e.g. Ni^{63}, fig. 7(b), into a series of electron peaks, now of rectangular shape, and consider each as a modified peak. In the case shown the high energy end of the spectrum is divided into 4 KeV energy intervals. It is not the intention to discuss in any detail the calculations involved in the adjustment of the spectrum but merely to sketch the method. In constructing the tail of fig. 7(a) the energy intervals into which the tail was divided corresponded to equal angular intervals of θ greater than the critical value at which a 60 KeV electron just reaches the wall, i.e. for a given value of θ, the energy is that given up by a 60 KeV electron, travelling at an angle θ, before hitting the wall. Returning to fig. 7(b), consider the electron group of mean energy 60 KeV. If it had not been for the wall it would have been higher by a factor 0.16. For the sake of clarity, the magnitude of the correction has been multiplied by two. However the pulses going into the 60 KeV group must be subtracted from the rest of the spectrum. A "tail" for the 60 KeV group is calculated as in the case of the single peak of fig. 7(a) and its area is made equal to the area which has been added to the 60 KeV group. This tail is now subtracted from all the lower energy groups and is indicated by a dashed line in the 56 KeV and 52 KeV energy groups in the figure. Now the 56 KeV peak is modified in an exactly similar manner, the height being increased by 0.14 (multiplied by two to exaggerate the effect) and the tail is subtracted from the rest of the spectrum. The changes due to the correction of the second peak are indicated by dot-dashed lines. Unfortunately in the figure the tail has, by error, been added to the 52 KeV peak instead of being subtracted. By proceeding in this way the

whole spectrum can be corrected for the wall effect to any desired degree of accuracy.

11. Calibration.

To carry out accurate beta-ray spectroscopy with the proportional counter it is essential to have an accurate and unambiguous means of calibration. At first it was suggested that X-rays reflected by a crystal at a particular angle might be used. Using the experimental set-up shown in fig. 8 the writer carried out some experiments using a Geiger counter as a detector. As an example of the resolution obtainable by this simple apparatus, fig. 9 shows the L_γ peak emitted from the tungsten target of the X-ray tube. This showed the method to be quite practicable for calibration purposes. If used for this purpose, the crystal would be set at the angle corresponding to the energy desired. Since the relatively insensitive Geiger counter gave 300-400 c.p.m. in this region ($\sim$ 11 KeV) it is evident that there would be adequate intensity for the proportional counter which has, as normally used, a considerably higher efficiency for the detection of X-rays, relative to the Geiger counter.

However shortly after this itwas found that by holding a thin metal foil opposite the window of the proportional counter, at an angle of 45° to the X-ray beam and to the perpendicular to the counter at the window, the X-rays reflected into the counter consisted almost entirely of the X-rays characteristic of the metal used. The most prominent line is usually the K_α (and L_α if the L X-rays are examined). The resolution obtainable ~~may be obtainable~~ may be observed in fig. 10, which shows the X-rays of silver. This very simple method has remained the one most used ever since. Most commonly, copper foil (K_α = 8.04 KeV) or silver foil (K_α = 22.1 KeV) has been used but chlorine (in the form of chloride),

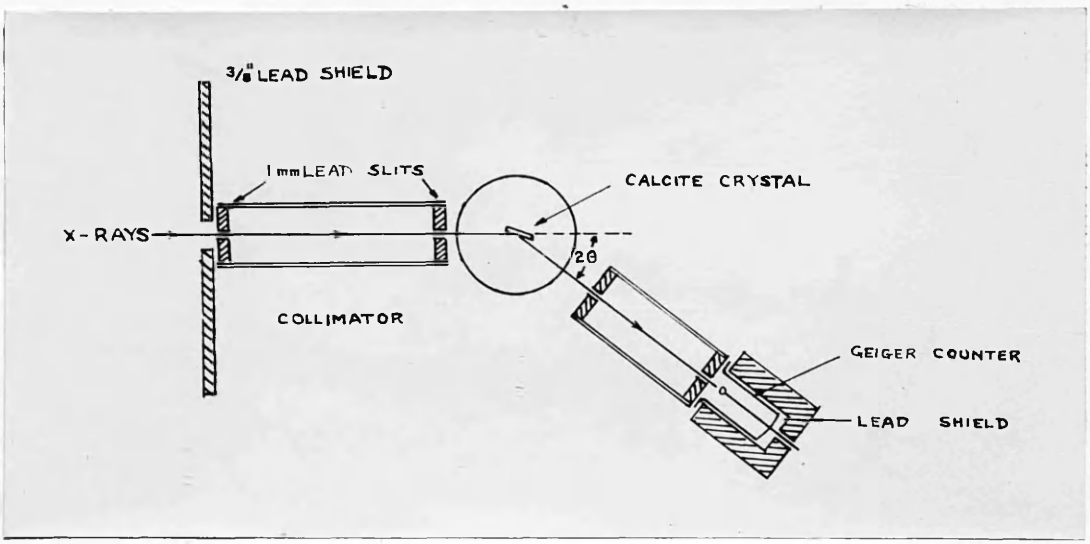


Fig. 8. X-ray monochromator experimental arrangement.

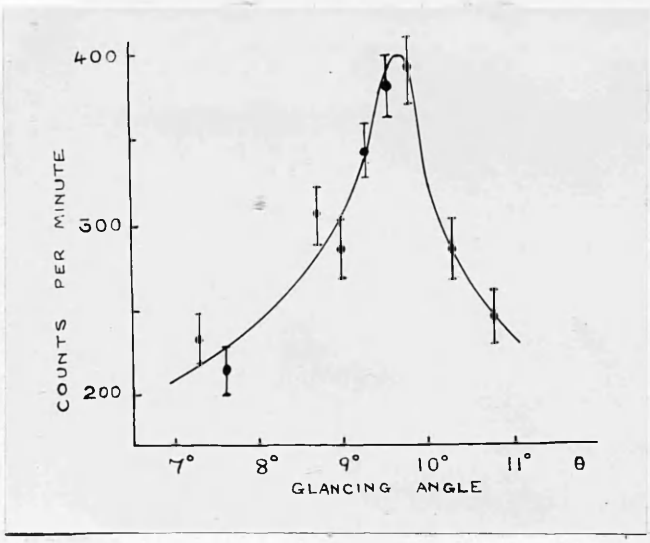


Fig. 9. L γ X-ray peak of tungsten.

calcium, manganese, molybdenum, tin, barium, samarium and platinum have also been used. Some of these elements were used in the metallic form, others as chemical compounds.

For calibration in the higher energy regions the characteristic tungsten radiations from the tube itself have been used. In this case the X-rays were fired through the copper wall of the counter. Initially the ratio of the intensities of the K_β to the K_α radiations is 0.36:1. After passing through the $\frac{1}{4}$" copper wall used in most of the counters, the ratio becomes about 12:1. Thus the peak observed consists mainly of the K_β X-rays.

Another method of calibrating is to use sources emitting monoenergetic X-rays or electrons. (Insch, 1950), Rothwell and West, 1950). Particularly suitable are the K-capture sources such as Cr^{51}, Fe^{55}, Zn^{65}, Ge^{71}, Se^{75}, Pd^{103}, Sn^{113} and Yb^{169} which emit the characteristic X-rays of V, Mn, Cu, Ga, As, Rh, In and Tm respectively. Also the isomeric transition of In^{114} gives rise to a 190 KeV gamma-ray which internally converts producing In X-rays. Insch also showed that, if X-rays from a isotope of given Z were passed through or reflected by a thin layer or foil of material of lower Z, the X-rays of the lower Z material were strongly excited. In the reflection case the thickness of the reflector did not matter but in the transmission case the thickness was chosen to give a high ratio of modified to unmodified X-rays. Rothwell and West tried to excite characteristic X-rays by electron bombardment of metals but were unsuccessful.

12. The Experimental Technique.

The signal from the counter passed to a head amplifier described by Curran, Angus and Cockroft (1949a). It was then amplified by a push-pull amplifier designed and built by A.L. Cockroft and described by Curran and

Craggs (1949, p.129). Usually the pulse was then displayed on the screen of a cathode ray oscilloscope. The deflection of the beam was proportional to the energy of the event giving rise to the pulse. The pulses were recorded on a moving 35 mm film which, after processing, was analyzed by projecting the pulses on a grid of equally spaced lines. The pulse height was measured in arbitrary units and compared with a standard pulse size arising from the use of calibration X-rays. At other times the pulses were fed into a one channel kick-sorter. The first method was felt to be more dependable since all the data could be accumulated in a few minutes of operation as it was easy to record up to 50,000 events per minute - provided the source was of sufficient strength to give such a counting rate. The main disadvantage of the photo graphic method was the length of time necessary to analyze the results. When using the one channel kick-sorter method, runs of many hours duration were often necessary and the requirements of voltage stability became much more stringent as a result. The use of multi-channel kick-sorters would reduce the time of experiment considerably. Work is proceeding on the development of such kick-sorters.

Before commencing an experiment with the proportional counter, it was customary to fill it with a fresh gas mixture. The counter was then left with the (approximately) correct voltages applied to the cathode and end correction tubes for at least half an hour before filming or making other observations. In the case of the glass walled counters used in the early experiments, it was not so essential to refill them before use since the counter action did not deteriorate for some weeks at least, but the normal metal counter usually showed evidence of deterioration in a period of a few days if it was not refilled. A few counters deteriorated more rapidly

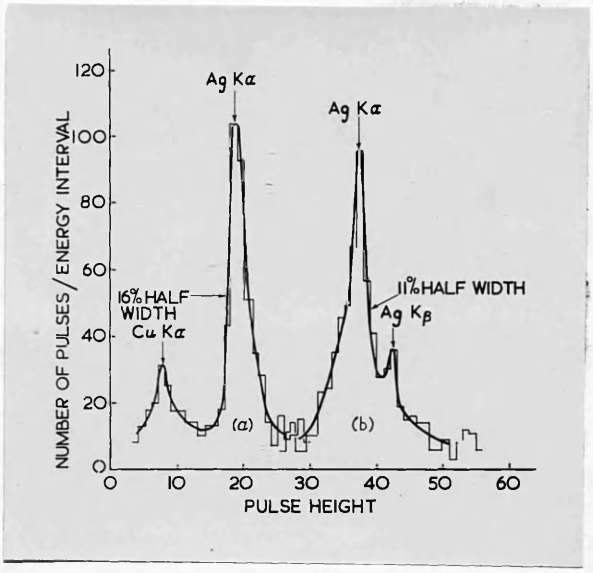


Fig. 10. Calibration X-rays (a) just after filling; (b) after 32 days.

than this for no obvious reason. On the other hand a counter used by the writer to examine the beta-rays from long-lived samarium actually improved with time. In fig. 10(a) a histogram of the silver K X-ray made when the counter was just filled is compared with the histogram obtained 32 days later, fig. 10 (b). It will be noticed that the line width is reduced from 16% to 11% and that the K_β line is resolved in the second case. 103 days after filling the silver group was again examined and even after this period the line width appeared to be as narrow as before. The counter did not receive any special treatment in filling. It is believed that the presence of the radioactive source inside the counter during this period may have had some purifying action on the gas. The copper peak shown in fig. 10(a) is due to the silver X-rays striking the copper window support.

13. The Purpose of this Research.

As has been mentioned earlier, two of the great advantages of the proportional counter technique are its abilities (a) to measure low energies accurately and (b) to detect and accurately measure the energies of very weak radioactivities. Whereas the first property was the more important one in the earliest studies with this technique (on tritium), the principal object of this research was to exploit the second. Weak radioactivities which could not otherwise have been examined and which were of particular interest were therefore investigated. In many cases a weak radioactivity infers a long half life and this is often associated with low energy radiations, since, for a fixed ft value, if t is large, f is small (relatively speaking) and therefore the maximum energy released tends to be small. For this reason the first property mentioned above was generally also of considerable value in this work.

The research commenced with the study of the long lived radioactivity of nickel, which, because of low activity, had been investigated previously only by absorption. The particular reasons which made this activity one of considerable interest are given at the beginning of part 2, which follows. The study of the radioactivity of Hg^{203} was undertaken because the integrating properties of the proportional counter were of great value in this case for reasons which will be made clear in the introduction to part 3. The activities of Sm^{151} and Eu^{155} had not been reliably investigated when the research described in this thesis began and knowledge of the decay schemes was very imperfect. Since the energies involved were low and the activities were rather weak, it appeared that utilisation of the properties of the proportional counter would afford much help in arriving at a clearer understanding of the decays.

It was evident that the sensitivity of the counter would be of most value in the study of the weak natural radioactivities which occur in the region of atomic number $Z < 80$. In this region a number of natural radioactivities have been reported in recent years, as has been mentioned earlier e.g. Nd^{150}, Lu^{176}, Re^{187}, Sm^{147}, Gd^{148} etc. Rubidium and potassium were very early shown to be radioactive (in 1905, by Sir J.J. Thomson). In spite of the fact that considerable work had been carried out on these activities, it is only recently that any accurate results and understanding of the decay schemes involved have been attained and this, up till the time of this research, in the case of K^{40} only. When the research on Rb^{87} was commenced, the end-point of the beta-spectrum was reported variously as 10 KeV, 130 KeV, 144 KeV, 250 KeV, 560 KeV, 1.1 MeV. This uncertainty was mainly due to the difficulty of measuring such a weak activity with any accuracy. However the source strength available was sufficient to allow

an accurate investigation to be made by the proportional counter technique. The only technique rivalling the counter in this field is the scintillation spectrometer. However for accuracy in the low energy region of the beta-spectrum the proportional counter is to be preferred although the scintillation counter is unequalled in its ability to measure weak gamma-radiations above, say, 50 KeV accurately. The data on the radioactivity of Nd^{150} was also felt to be worthy of confirmation and preliminary work was carried out on this activity, which incidentally did not agree at all with the previous findings. In the introduction to part 5, which deals with the investigations into the radioactivities of Rb^{87} and Nd^{150}, there is a fuller discussion of the natural radioactive decays, and of the role of the proportional and scintillation counters in unravelling these decays.

Further work on these weak activities is being carried out in Glasgow and the writer is at present investigating the possibilities of a new method (electron sensitive emulsions) in this field. There are at least half a dozen activities of the nature described which ought to be investigated.

The possibility of using the proportional counter method in the investigation of possible double beta-decay should not be overlooked. The half life for such a process depends markedly on whether or not two neutrinos are emitted. In either case the half life expected is extremely long and only the most sensitive methods hold any promise of providing an answer.

PART 2. RADIOACTIVITY OF NICKEL.

1. Introductory.

When this research was undertaken early in 1948, the only long lived isotope of nickel reported was Ni^{59}. According to several publications (Science 1946, Rosenfeld, 1948) Ni^{59} was believed to be a positron emitter of upper energy limit 50 KeV and of half life 15 years the decay being presumably

$$Ni^{59} \longrightarrow Co^{59} + \beta^+$$

If the facts were correct, this was a very interesting source since no positron emitter of comparable low energy limit exists. A search of the nuclear tables shows that the lowest energy emitter, apart from Ni^{59}, is Zn^{65}, which has a positron energy limit of 320 KeV and a half life of 250 days. Also this is the longest half life associated with any other positron emitter. The long half life attributed to Ni^{59} is to be expected because of the low energy limit, and corresponds to an allowed transition.

If the energy limit were correct, the average energy of the beta-rays would be of the same order as the K-shell energy of the daughter nucleus. Careful examinations of the spectrum shape might have been expected to give evidence of, and information about, extra-nuclear effects due to the atomic electron shells.

For these reasons it was thought that the source was well worthwhile investigating and it seemed that the proportional counter technique was admirably suited to it, because of its low energy and long life, and therefore, presumably, its low activity. It was hoped that it would be possible to use the nickel source in the form of nickel carbonyl vapour, $Ni(CO)_4$, so that no source thickness difficulties would present themselves.

This was the technique used in the examination of H^3, C^{14} and S^{35}.

The first sources were prepared from several grams of metallic nickel irradiated for three months at a neutron flux of 2×10^8 neutrons per cm² per second in the early pile, "GLEEP", at A.E.R.E., Harwell. Later irradiations were carried out in the pile "BEPO" at fluxes of 10^{10}-10^{12} neutrons per cm² per second. Iron-and cobalt-free nickel oxide was used in the later irradiations. The active nickel was always allowed to stand for several months to allow short lived activities to decay.

2. Chemical Purification and Source Preparation.

I. Plated Sources: Although for counter examination it was desired to use the nickel as a vapour, nickel carbonyl, for preliminary examination a solid source seemed preferable. The source as received from Harwell appeared to be quite strongly radioactive and emitted penetrating radiations attributed mainly to Co^{60}. It was evident that careful chemical purification was necessary. The plated sources were prepared by dissolving the nickel in hot concentrated nitric acid, neutralising and precipitating the nickel by adding dimethylglyoxime solution which precipitates nickel in the presence of its chemically very similar neighbours, iron and cobalt. The details of this process will be given later. The precipitate was filtered, dried, oxidised rapidly and the residue dissolved in sulphuric acid. The solution was neutralised by adding ammonium hydroxide and then used as an electrolyte in the formation of nickel sources by electroplating. The cathode material was generally copper foil.

Sources prepared in this way showed a beta-activity of low energy accompanied by a little high energy beta- and gamma- activity which was

ascribed to small traces of radioactive impurities still present. The chemical purification technique about to be described completely eliminated these impurities. Their presence, however, did not materially affect the results of the preliminary experiments.

II. Nickel Carbonyl Preparation: The process of preparing nickel carbonyl was found to be neither easy nor straightforward. Nickel is prepared in a very finely-divided metallic state and carbon monoxide gas is passed over it at a temperature of less than 100°C. Up to 100° increase of temperature increases the rate of reaction. The process given by Mellor (1932) was first tried. Nickel oxide was reduced to nickel by passing a stream of hydrogen over it at a temperature of between 300° and 400°. Then carbon monoxide was passed over the nickel at 30° - 50°. However no nickel carbonyl was formed.

After a considerable amount of effort, a modified form of the process described by Fernelius (1946) and due to Gilliland and Blanchard was found to give satisfactory results. The whole chemical procedure can be considered in two parts (a) the preparation of pure nickel hydroxide for the manufacture of nickel carbonyl and (b) the preparation of the carbonyl. Details of these preparations are given for two reasons. Firstly, work has been published on the radioactivity of nickel which is not at all in accord with experiments carried out, with considerable care, by the author, nor with more recent work in the United States (which substantially confirms the work described in this thesis). It is evident that the chemical purification of the source was not carried out sufficiently carefully or completely. Secondly, no process of preparing nickel

carbonyl given in the literature which came under the writer's attention was successful as written. It seems wise, therefore, to put the following information on record. The quantities used are based on starting with one gram of nickel.

(i). Dissolve the nickel (or nickel oxide) in hot concentrated nitric acid. Evaporate gently to dryness. Add concentrated hydrochloric acid and again evaporate to dryness. Repeat with hydrochloric acid. This converts the nickel nitrate to nickel chloride.

(ii) Precipitation of cobalt by α- nitroso -β- naphthol. Dilute with water and add as carrier 0.05 gm cobalt, as chloride, per 100 cc solution. Heat to boiling and add hot fresh α- nitroso -β- naphthol solution at the rate of 50 cc per 0.05 gm of cobalt. The Method of preparing the α-nitroso -β- naphthol solution is given by Mellor and Thomson (1938). Heat for a few minutes, stirring well, allow to stand for two hours and filter. The precipitate always showed some activity, even when pure cobalt- and iron - free nickel oxide was irradiated in the pile, the activity being weakest in a sample of pure nickel oxide which was irradiated in the thermal column of the pile.

(iii). Precipitation of nickel by dimethylglyoxime. Boil off the acetic acid left after last stage. Dilute the solution to 2700 cc. Add ammonia solution till just alkaline, heat to 60° and add, while stirring, 1100 cc of a 1% solution of dimethylglyoxime in alcohol. Stand for half an hour and filter.

(iv). Dry the precipitate in 115° oven and convert to oxide by heating in a silica crucible. As the precipitate is somewhat volatile this must be carried out rapidly and in a well ventilated fume cupboard.

(v). Dissolve the residue in concentrated nitric acid, evaporate gently

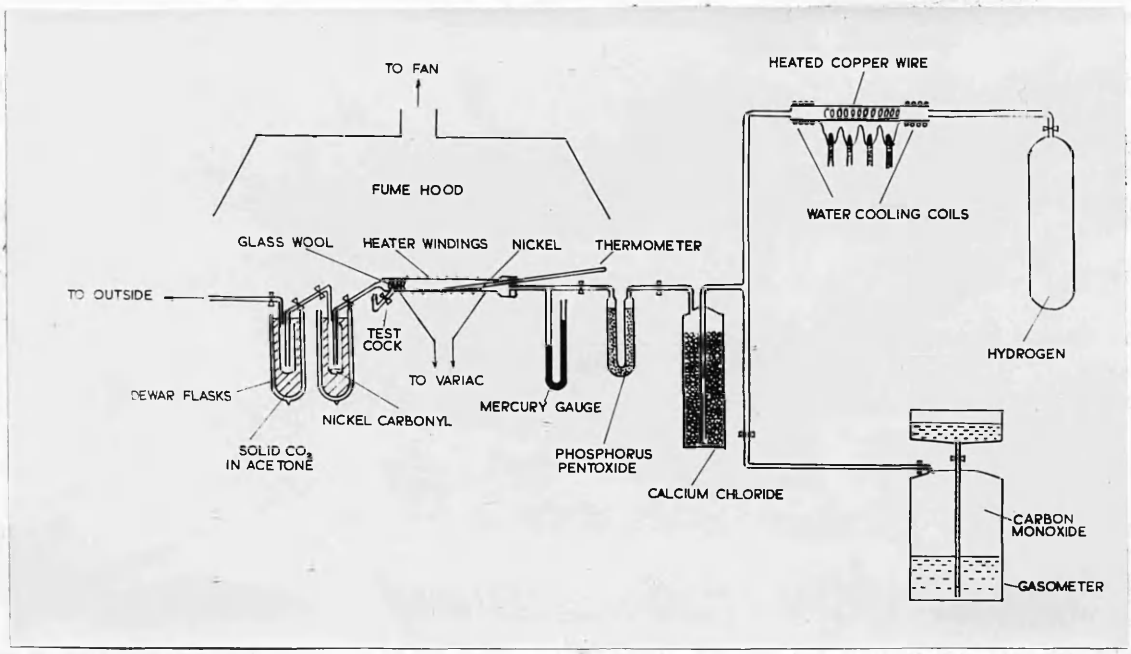


Fig. 11. Apparatus used in the preparation of nickel carbonyl.

and dissolve in water. Precipitate nickel as hydroxide by adding ammonium hydroxide. This precipitate dissolves in excess ammonia but can be re-precipitated by boiling. Filter. Precipitation of nickel by the obvious method of adding sodium or potassium hydroxide was found to give a form of nickel hydroxide from which it was impossible to prepare nickel carbonyl. Dry the hydroxide precipitate very gently, at a temperature not exceeding 100°C.

(vi). The hydroxide is now mixed with 1% of its weight of mercuric oxide. This acts as a catalyst in the subsequent process.

The remaining part of the process was carried out using the apparatus shown in fig. 11. Hydrogen from a cylinder was passed through a tube containing red-hot copper wire to remove all traces of oxygen which appears to act as a poison. The ends of the tube were water-cooled. The hydrogen was dried by passing over calcium chloride followed by phosphorus pentoxide. It was then passed over the nickel hydroxide (mixed with mercuric oxide) in a specially constructed Pyrex combustion tube at a temperature not exceeding 250°C. The tube was heated by a Nichrome resistance wire winding, the current through which was controlled by a "Variac" transformer. When the hydroxide was completely reduced to nickel (indicated by the cessation of production of water) the tube was allowed to cool with hydrogen still passing over it. The nickel was now very finely divided indeed. (On opening the tube to the atomsphere after a week's run, the remaining nickel would catch fire spontaneously).

Carbon monoxide, prepared by adding concentrated sulphuric acid to sodium formate, was contained in a gasometer of 50 litres capacity. The gas, dried by calcium chloride and phosphorus pentoxide, was passed very

slowly over the active nickel forming nickel carbonyl. The carbonyl, which boils at 43°C. at atmospheric pressure and has a vapour pressure of 300 mm of mercury at 18°C. passed as vapour into a series of two source bottles which were immersed in a solid carbon dioxide-acetone mixture, or liquid oxygen, contained in Dewar flasks. It was possible to test whether or not the reaction was proceeding by examining the colour of the flame with which the carbon monoxide burned. In the presence of even slight traces of nickel carbonyl, the flame was bright yellow instead of the faint blue flame of carbon monoxide burning alone. This test had to be carried out under a well ventilated hood. Quite apart from the radioactivity of the nickel, nickel carbonyl is a very poisonous gas.

This process was carried on for about a week, the temperature never being allowed to exceed 80° C. Not only does the rate of decomposition of the carbonyl increase if this temperature is exceeded but also the nickel appears to become less active. After a period of about a week, the reaction slowed down so much that the nickel had to be reactivated. However enough carbonyl for the experiments on hand was produced in that time. Nickel carbonyl does not keep well. It had always to be kept surrounded by a solid carbon dioxide-acetone mixture.

Unfortunately it was found that nickel carbonyl vapour had a poisoning effect on a proportional counter even in very small amounts so that, after all, solid sources had to be used. Nevertheless, this process had two great advantages. (a) It is an excellent method of purifying nickel, since only nickel, of all metals, forms a volatile carbonyl. Thus, even if any impurity did remain in the active nickel, only nickel would have been transferred from the combustion tube to the source bottle. (b). It

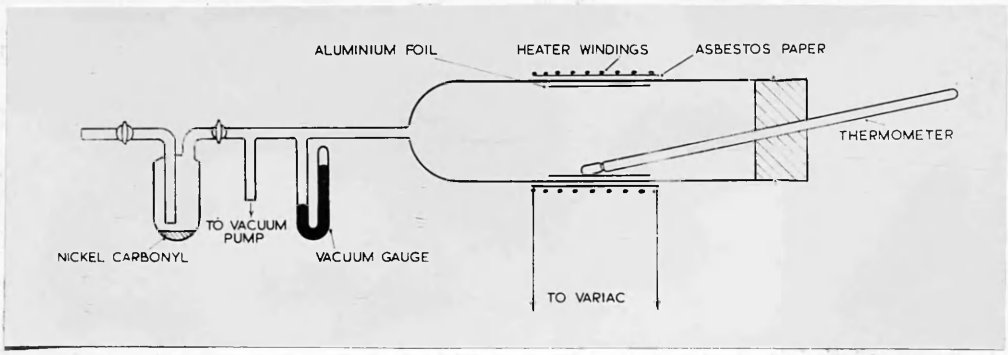


Fig. 12. Apparatus used in depositing nickel from nickel carbonyl.

is possible to form extremely thin and perfectly uniform sources over large areas by decomposing the carbonyl on a hot surface. The uniformity of source deposition is equalled only by evaporation techniques (i.e. evaporation of solids in vacuum.) The method of depositing the nickel is described in the next section.

III. Preparation of Nickel Sources from Nickel Carbonyl.

The apparatus employed is shown in fig. 12. A bottle containing nickel carbonyl was attached to a large Pyrex tube on the inside wall of which was rolled a sheet of aluminium of thickness one or two thousandths of an inch. The foil was heated by passing a current from a "Variac" through an external coil of Nichrome wire wound on asbestos paper wrapped round the tube. The whole apparatus was evacuated as thoroughly as possible with a "Hyvac" pump and the temperature raised to 200°C. The pumping tube stop-cock was then closed and the carbonylallowed to warm up to room temperature. Then the vapour was allowed to enter the evacuated tube. As soon as it came in contact with the hot aluminium surface, it decomposed and formed a black deposit on the foil. In early experiments the apparatus was left at this stage for a considerable time. The current was then switched off and the bottle immersed in liquid air to trap all the carbonyl vapour remaining in the apparatus. In the last experiment (a search for X-rays emitted by the source) the process was repeated several times, the large tube being evacuated each time between cooling and heating of the source bottle.

3. Absorption Experiment.

Preliminary experiments on the deflection by a magnetic field of the particles entering a Geiger counter seemed to indicate that the particles were negative instead of positive. This seemed to contradict

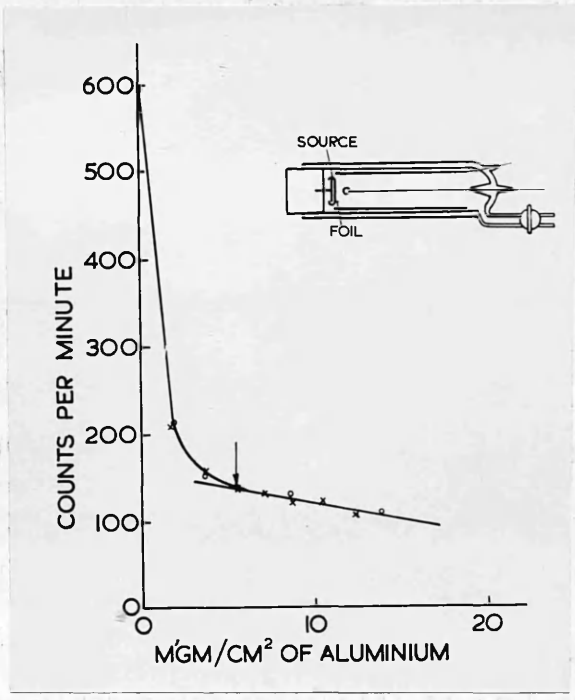


Fig. 13. Absorption curve of the beta-particles emitted by radioactive nickel.

the information published in the literature. In this experiment the source was mounted inside the same glass envelope as the Geiger counter in order to eliminate window losses. If this had not been done one could not have been sure that the higher energy particles, which only would have been transmitted through the window, were not due to the presence (in small quantity) of another activity in the source. As it was necessary to be sure that we were investigating the same radiation as that attributed to Ni^{59}, it was thought wise to carry out an absorption experiment to determine the energy of the beta-rays approximately. The results of two experiments are shown by circles (first experiment) and crosses (second experiment) in fig. 13. An inset figure shows the nature of the Geiger counter used to measure the counting rate of the particles. The source, which consisted of radio-nickel plated on to a copper foil, was attached to a brass support which in turn was rigidly fixed to a plug in the end of the counter. This plug could be removed and as absorbing foil placed over the sourc, which was situated as near the end of the counting region as possible to avoid absorption in the counter gas. This arrangement avoided the window difficulties which arise when the experiment is carried out in the usual way. The plug was replaced very accurately each time another absorbing foil was added and, on re-filling, the gas mixture was made as nearly identical to the original as possible. A good end-point was obtained in this manner at 5.5 mgm/cm^2 of aluminium. Allowing for the gas between the source and the counting volume, this corresponds to an energy of ~60 KeV. (Glendenin 1948). This was in reasonable agreement with the figure of 50 KeV quoted earlier for the long life activity of nickel and is in excellent agreement with the end-point of 63 KeV determined by the proportional counter

method (to be described later). This gave confidence that the activity investigated by previous workers and the present activity were identical.

It was then considered necessary to determine the charge carried by the particles more definitely, as this was of fundamental importance.

4. The Charge of the Beta-Rays.

The more obvious methods of determining the charge of the particles could not be employed since the counting rates of the sources available were far too small. The negative beta-spectrometer is out of the question even now with the stronger sources now available (due to higher neutron flux densities in the pile). The use of an electroscope or electrometer was also out of the question as a simple calculation will show. Taking the capacity of an electroscope of the type available as, say, 10$\mu\mu$F and the change in voltage reasonably easily detectable as 10 volts, then a charge of $10 \times 10 \times 10^{-12} = 10^{-10}$ coulombs would be necessary. Since the charge on an electron is 1.6×10^{-19} coulombs, about 6×10^{8} electronic charges would be required, or 3×10^{8} decays if use was made of the charge left on the nucleus. As the strongest sources then available gave about 10,000 counts/minute (over large areas), 3×10^{4} minutes, or over 8 hours, would be necessary to observe the required change in voltage - during which time the leakage losses would have been much larger.

Although the magnetic spectrometer was out of the question it was decided to use the sign discrimination properties of a magnetic field in conjunction with the integrating properties of a photographic plate. In the arrangement employed, the condition of high resolving power, normally important in a spectrometer, was relaxed. By having no energy discrimination a high solid angle was obtainable (probably about 1/6th. of 2π) and because of this, and the fact that electrons impinged on the

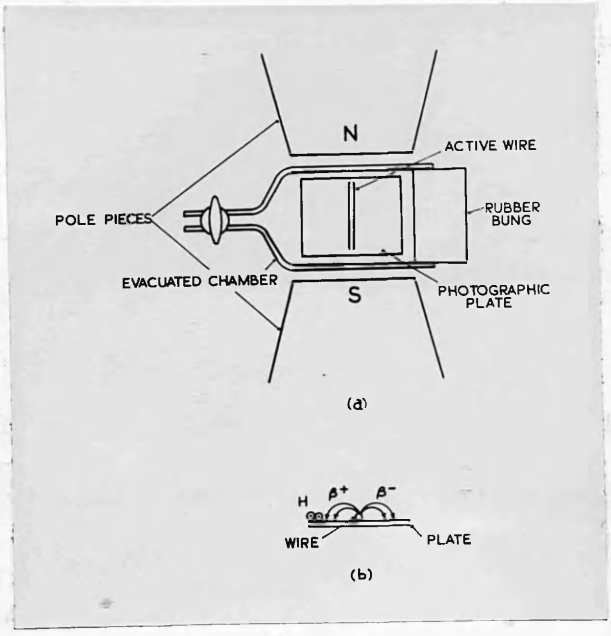


Fig. 14. Experiment to determine the charge of the beta-particles.

plate over a fairly small area, the experiment was possible. The arrangement is shown in fig.14. A platinum wire, on which radioactive nickel was electrolytically deposited was placed on a photographic plate which was inserted into a tube which could be evacuated. The tube was painted black and was covered with black tape to make it light-tight. The arrangement was inserted in a permanent magnetic field of about 2500 gauss, with the wire parallel to the lines of force. The apparatus was evacuated and put into a light-tight box where it remained for 30 hours. Now 50 KeV electrons would have a radius of curvature of a little less than 0.5 cm in such a field. Fig.14 (b) shows the effect of the field on positive and negative electrons. If positive they would be deflected to the left; if negative, to the right. The blackening on the plate showed that the particles were for the most part negative and its extent corresponded roughly to that expected for 50 KeV electrons.

5. The Search for Annihilation Radiation.

To confirm this finding, i.e. that the particles were negative, a search for annihilation radiation was made since such radiation would certainly be present if there was any appreciable positron activity. A special Geiger counter with a lead cathode (to increase the efficiency of hard gamma-ray detection) was constructed. 0.015" lead sheet, thoroughly cleaned with alcohol, was used for the cathode. A foil giving 50,000 counts per minute of beta-particles, prepared by deposition from nickel carbonyl, was wrapped round the outside of the counter. The length of foil, parallel to the axis of the counter, was $4\frac{1}{2}$". The counting rate of the counter without the foil (i.e. the background counting rate of the counter) was

153.2 ± 2.1 counts per minute.

With the foil wrapped round it, the counting rate was

148.8 ± 2.1 counts per minute.

If we assume the particles to be all positive, two quantum annihilation of the particles would have given rise to an additional 500 counts per minute, assuming the solid angle to be approximately 2π and the efficiency of detection of the counter for such radiations to be ~1%. The result implies that less than 1% of the particles could, in fact, be positrons.

Examination of a nuclear chart shows that the only likely mode of decay of Ni^{59} is by positron emission or by electron capture to form Co^{59}. No positrons were detected and it was evident that very few K X-rays of cobalt or Auger electrons could be present, experiments at this stage failing to reveal such radiations. Therefore the amount of Ni^{59} activity present in the source must have been very small. (It may be well to state, at this point, that a very careful search carried out more than a year later did reveal the presence of the K X-rays of cobalt, thus indicating the existance of Ni^{59}. However the intensity was found to be only about 1 part in 1200 of the negative particle intensity).

The question then arose as to which isotope of nickel the negative electron activity was to be ascribed. The existence of a 2.6 hour beta-activity had been known for some time and work by Swartout et al. (1946) and Conn et al. (1946) indictated that this activity belonged to Ni^{65}. This left Ni^{63} as the only possible isotope responsible for the soft beta-activity under investigation. This could be formed by an (n, γ) reaction on the relatively rare (3.8%) isotope, Ni^{62}, and the resulting nucleus, Ni^{63}, would decay by beta-emission thus

$$Ni^{63} \longrightarrow Cu^{63} + \beta^-$$

It is evident that the interesting soft positron activity ascribed to

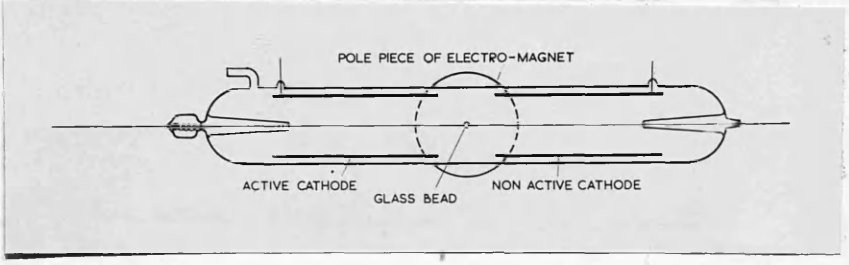


Fig. 15. Apparatus to detect coincidences between beta-particles and soft electromagnetic radiations or electrons.

Ni^{59} is in fact the negatron activity of Ni^{63}. It was considered well worth while to investigate the spectrum of this activity which had previously been measured only by absorption. Before discussing this work, an experiment which was carried out to detect the presence of coincidences between the beta-particles and X-rays, gamma-rays or other electrons, will be described.

6. Coincidence Experiment.

The apparatus employed is shown in fig.15. Two Geiger counters were enclosed co-axially in the same envelope. The cathode of the left hand counter consisted of a copper cylinder on the inside of which was electrolytically deposited a layer of radioactive nickel. The other cathode, also of copper foil, was inactive. An electromagnet was positioned so that a deflecting field could be applied between the counters if desired.

In the first experiment the single counting rate of the non-active cathode counter was measured without and then with the field on. The counting rate dropped considerably suggesting that particles which formerly came from the active cathode to the non-active counter were now being cut out by the magnetic field. The results are as follows:-

Field off : 119.2 ± 3.1 counts per minute.

Field on : 78.9 ± 2.6 counts per minute.

If the above explanation is correct this leaves 40.3 ± 4.1 counts per minute due to electrons coming from the active to the non-active counter. To confirm this view, the coincidence counting rate between the two counters was then measured, firstly, with the field off, and then with the field on. This gave:-

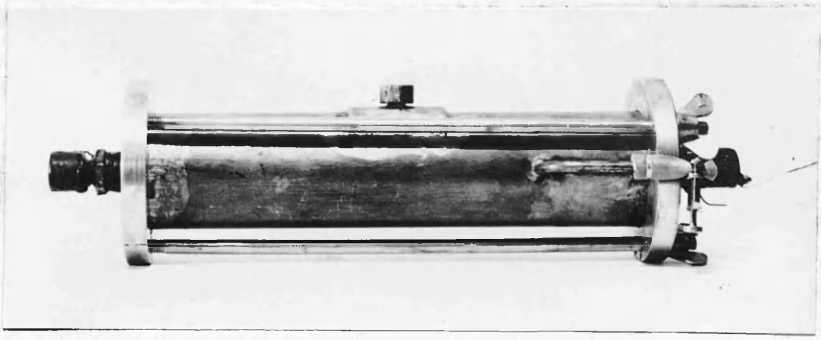


Fig. 16. Photograph of the proportional counter used in the investigation of Ni^{63} and Ni^{59}.

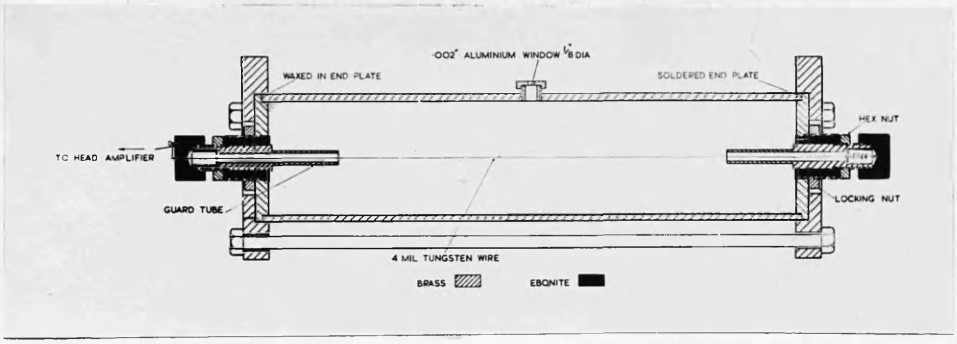


Fig. 17. Drawing of the proportional counter shown in fig.16.

Field off : 38.6 ± 2.2 coincidences per minute.

Field on : 2.3 ± 0.5 coincidences per minute.

Thus the coincidences counting rate fell by 36.3 ± 2.3 counts per minute which is in agreement with the drop in counting rate of the non-active counter. The coincidence counting rate observed, therefore, is not due to true coincidences between beta-rays and electromagnetic or other radiations. Indeed, since the activity of the foil was ~18,000 counts per minute, the low coincidence counting rate with the field on shows that a negligable number of soft gamma- or X-rays are in coincidence with the beta-rays emitted by Ni^{63}. It is unlikely that the coincidence rate observe with the field off is due to true beta-e^- coincidences, since no beta-X-ray coincidences are observed. It is almost certainly due to single beta-particles making a count in each counter in course of flight.

Since the experiment made to try to detect annihilation radiation, as well as other experiments to detect gamma-rays, showed that the hard gamma-ray intensity of the source was negligible and since the experiment just described shows that very few coincidences exist between electrons and softer electromagnetic radiations, we may conclude that the spectrum is simple. The unsuccessful search for X-rays, mentioned earlier, confirms this view.

7. The Beta-Spectrum of Ni^{63}.

It was now decided to investigate the spectrum of Ni^{63} with the aid of the proportional counter. A photograph of the counter used is shown in fig.16 while a drawing showing its construction is shown in fig.17. The tube was $10\frac{1}{2}$" in length and $2\frac{1}{2}$" in diameter. The active

length of the counter was about 7" and the wire was of 4 mil tungsten. The first attempts to observe the spectrum were made using nickel in the form of nickel carbonyl vapour. The nickel carbonyl was transferred from the source bottles of fig.11 to a larger container of capacity 315 cc. The vapour pressure of the carbonyl vapour was 7.7 cm of mercury. (Saturation vapour pressure at room temperature is about 28.5 cm). A small 15 cc container was then filled with vapour from the first container and this was shared with the proportional counter which had a volume of 450 cc and a background counting rate of ~350 per minute. The pressure of the nickel carbonyl in the counterwas about 2.5 mm of mercury. Argon and methane were added in the usual proportions but the counter did not operate as a counter with this amount of nickel carbonyl. There was no plateau, no proper pulses visible on the oscilloscope screen, and the counting rate as observed with a sealer was very variable but of the order of the background value. By a sharing technique, the pressure of the carbonyl in the counter was reduced to about one tenth, i.e. to ~0.25 mm of mercury. The counter behaviour was but little better than previously. The carbonyl pressure was again reduced by a factor of ten, to ~0.025 mm. Pulses were now apparent but the counting rate was too low, ~280 per minute. This is actually less than the normal background rate, showing clearly that the carbonyl vapour was acting as a poisoning agent. Also when K X-rays of silver and copper, and the X-radiations emitted by RaD and Zn^{65} sources were allowed to enter the counter, no homogeneous groups of pulses could be observed. The pressure of carbonyl vapour was again reduced by a factor of ten, giving a pressure of ~0.0025 mm of mercury. The counting rate now rose to the background value, but still

no uniform pulse size grouping was apparent when homogeneous X-rays were fired into the counter. It is extremely surprising that the effect of the vapour should be so marked at this low pressure.

The counter was now pumped for about five minutes and filled with the usual gas mixture, but with no nickel carboryl. It now operated perfectly and gave the usual background value of counting rate. This showed that the abnormal behaviour of the counter was due to the carbonyl vapour and further that the amount of vapour retained or dissociated by the wall was negligible.

Another experiment was carried out to determine the specific activity of the source. The 15 cc bottle was filled with nickel carbonyl vapour to a pressure of 6.7 mm of mercury. This was deposited on an aluminium foil with the apparatus of fig.12. This gave a counting rate in excess of background of 3800 counts per minute with a solid angle was about 2π. If the counter contained carbonyl to a pressure of 0.0025 mm of mercury (the lowest pressure used) about 4.5 counts per minute would be expected. This counting rate is much too low to be of any use and, even at the very low pressure corresponding to this counting rate, we have seen that the counter did not operate satisfactorily. Furthermore it was obvious that the higher pile neutron fluxes expected when the larger pile came into operation still would not yield a high enough specific activity of Ni^{63} to make the use of a vapour source possible. It was evident that the vapour must capture electrons very strongly indeed.

For comparison, an aluminium-glass counter of the type shown in fig.1 was filled with gas mixtures which contained various pressures of the halogens which are well known to have a poisoning effect. Both iodine and bromine vapours were tried. A change of partial pressure of iodine

from 0.003 mm to 0.2 mm of mercury was found to have no appreciable effect on the behaviour of the counter. However there was some reason to believe that the rate of diffusion of the iodine into the counter from the vessel in which it was contained was rather slow and the experiment was repeated using bromine, taking steps to ensure that the partial pressures reported were actually attained. When the counter was filled with the usual gas mixture plus bromine to a partial pressure of 31.5 mm, no homogeneous grouping of pulses was observed when the counter was irradiated with monoenergetic X-rays. By the sharing technique, the pressure of the bromine was reduced to 3 mm. It still did not operate as a proportional counter. The pressure was further reduced to 0.3 mm. There was now slight evidence of a monoenergetic group of pulses and the behaviour was considerably better than with nickel carbonyl at 0.0025 mm pressure. On reducing the pressure to 0.03 mm the grouping was much better though not perfect. However it was evident that nickel carbonyl had a much more harmful effect on the operation of the counter than had bromine. The poisoning effect of the carbonyl is presumed to be due to the high capture cross-section of the carbonyl molecule for electrons in which it resembles oxygen, water, the halogens and other substances in effect though by no means in degree. Since Professor H.S.W. Massey has done much work in this field, his opinion was sought on the above explanation of the phenomenon. He agreed that nickel carbonyl was very likely to have a high electron affinity but could not say so definitely.

It was now decided to investigate the spectrum with nickel deposited on an aluminium foil, using the apparatus shown in fig.12.
As has been stated, this gave a thin, finely divided, uniform and tenacious

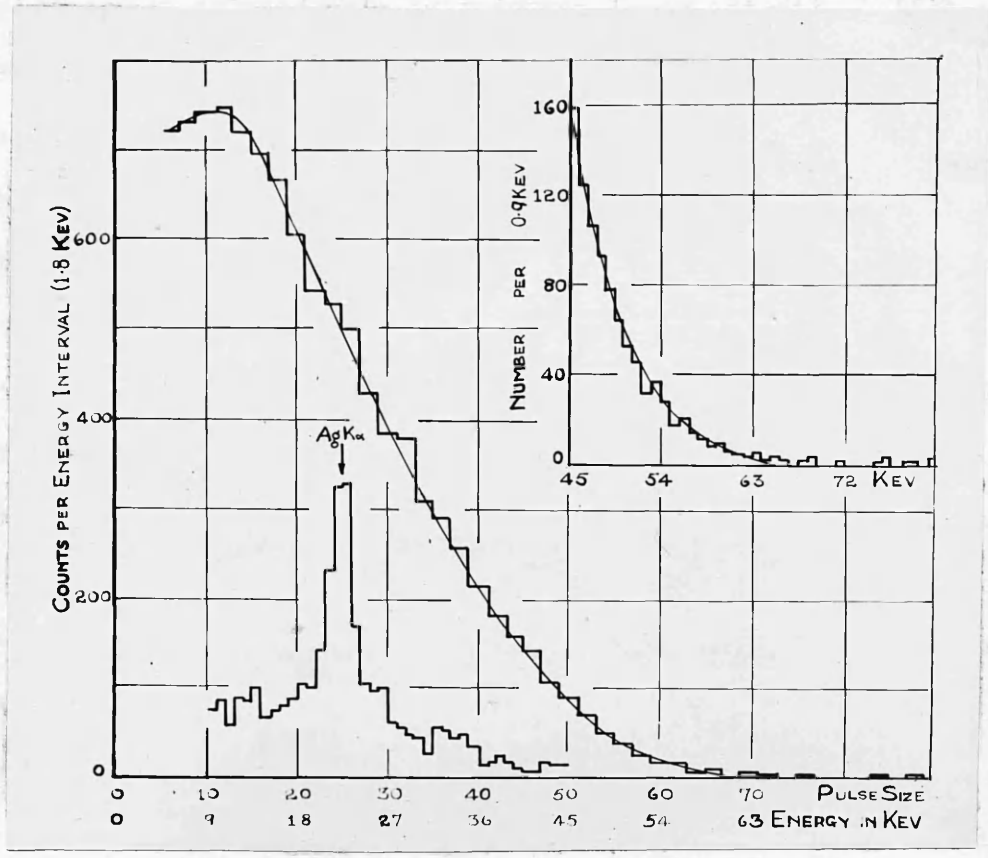


Fig. 18. Beta-spectrum of Ni^{63} with calibration and end-point (inset).

film of metallic nickel. The source, which covered an area of about 6" x 3", had an activity of ~50,000 disintegrations per minute. This corresponded to a source thickness of about 0.57 mgm/cm^2. The foil was inserted into the centre of the proportional tube with its shorter length parallel to the wire. The counter was filled with 15 cm of methane and argon to a total pressure of two atmospheres. The K X-radiations of silver were used for calibration (K_α = 22.07 KeV for silver) and the pulses were recorded on a moving photographic film in the usual way. The energy spectrum obtained is shown in fig.18. The calibration histogram is underneath the spectrum and the inset figure gives the endpoint of the spectrum with better statistics. A good end point was obtained and was estimated to be

$$E_{max} = 63 \pm 2 \text{ KeV.}$$

The histogram of the spectrum is composite. The low energy region of the spectrum was examined at higher gain and is fitted to the main spectrum The maximum intensity occurs about 10 KeV. A Fermi plot of the spectrum is shown in fig.19. N is the intensity per energy interval, p the momentum W the total energy (including rest energy) and G(W,Z) the correction factor for the Coulomb field. G is given by

$$G(W,Z) = \text{const.} \times p^{2s-2} \xi / (e^{-\xi} - 1)$$

where $s = (1-\alpha^2 Z^2)^{\frac{1}{2}}$, $\alpha = 1/137$ and $\xi = 2\pi Z/137$. The plot is straight over most of its length though it drops at lower energies (below 15 KeV). This might suggest a first forbidden spectrum but in view of the source support thickness this conclusion is very tentative. With the energy obtained here and the half life of 61 years obtained by the author (as will be described later) a log ft value of 6.40 is obtained. In a paper

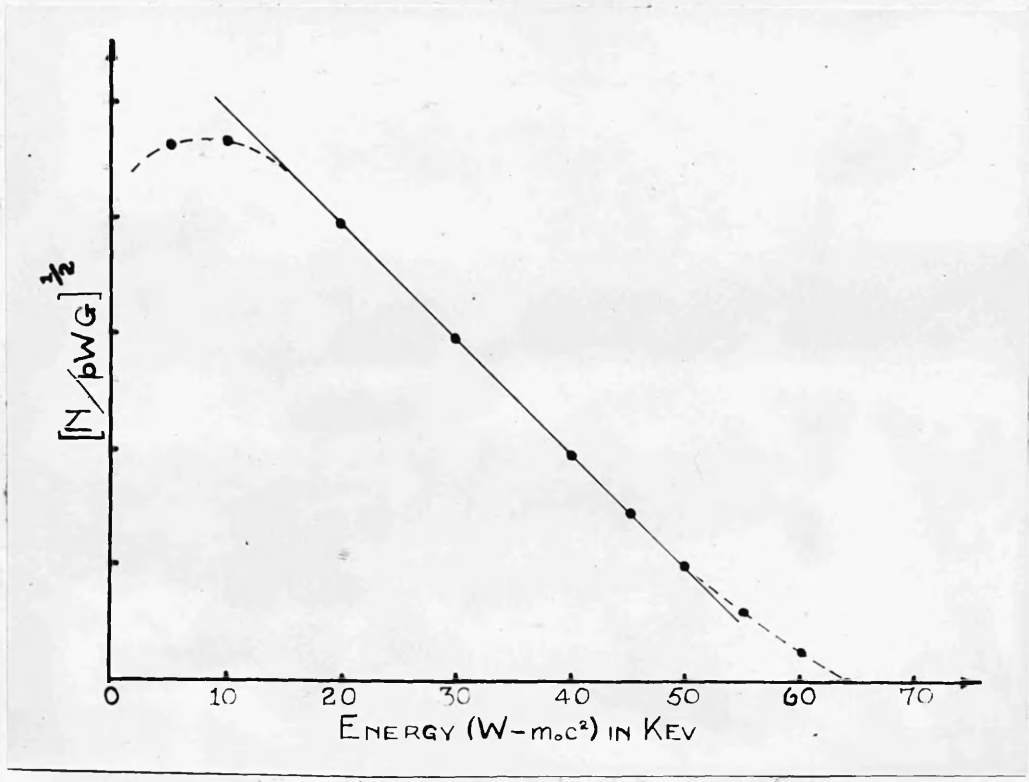


Fig. 19. Fermi plot of spectrum.

of Feenberg and Trigg (1950), the remark is made in reference to fig.10, a histogram of log ft values, that the peak at log ft = 6.2 is "predominantly first forbidden". The value of log ft quoted here indicates that the spectrum could therefore be either first forbidden or allowed with the evidence slightly in favour of the former classification.

8. Possible Sources of Distortion of the Spectrum Shape.

It might be well to mention the possible sources of distortion in the spectrum shape obtained. The one usually of most importance, "end effect", does not concern us here, since the source was confined to the central region of the counter. The "wall effect", investigated in the introduction to the thesis, was shown there to have a very small effect in this case. A more serious effect might have been caused by the source support which was of aluminium. This metal was chosen in view of the smaller amount of electron reflection caused by a low Z material. For the energies involved it is likely that not more than ~10% of the particles going into the support will be reflected. At any rate this is more likely to give rise to an increase in the number of low energy particle whereas a decrease was observed. The source thickness was very small and should not have affected the spectrum shape above, say, 10 KeV. It would cause a loss in the number of low energy particles. This work was published in the Philosophical Magazine (Wilson and Curran, 1949).

9. The Radioactivity of Ni^{59}.

About a year after the above experiments were completed, it was decided to carry out a very careful search for soft electromagnetic radiations. These might arise from complexity of the Ni^{63} spectrum, or from K-capture in Ni^{59}. Ni^{59} must certainly be present in a source

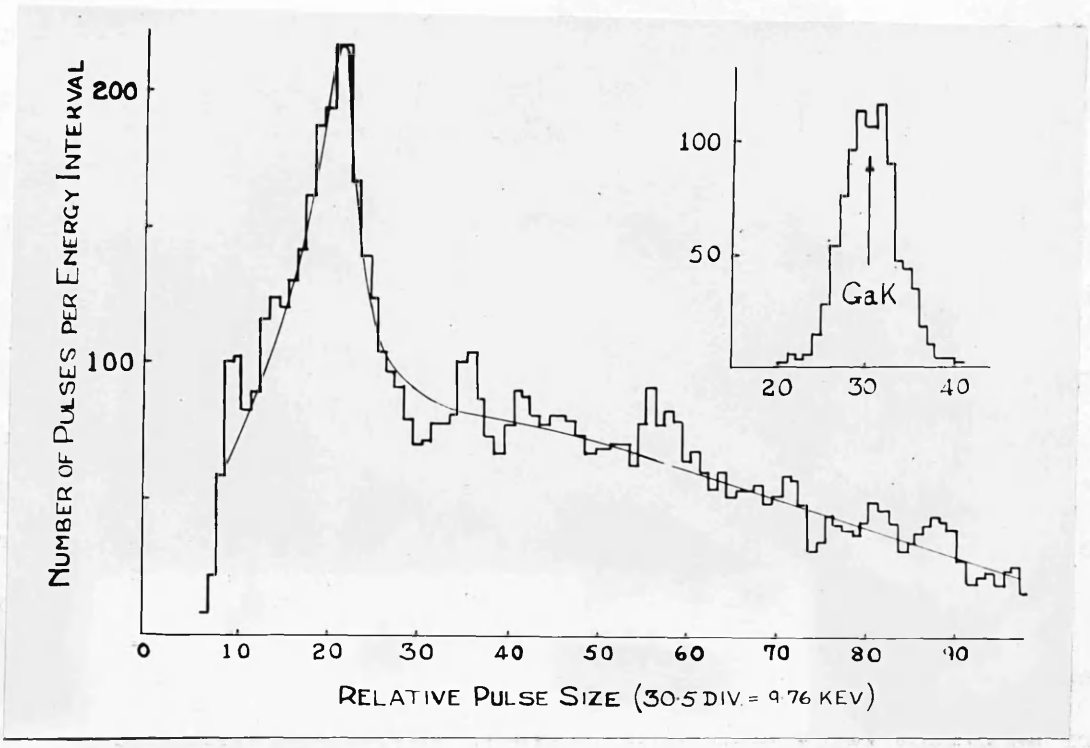


Fig. 20. Spectrum of radiations up to ~30 KeV.

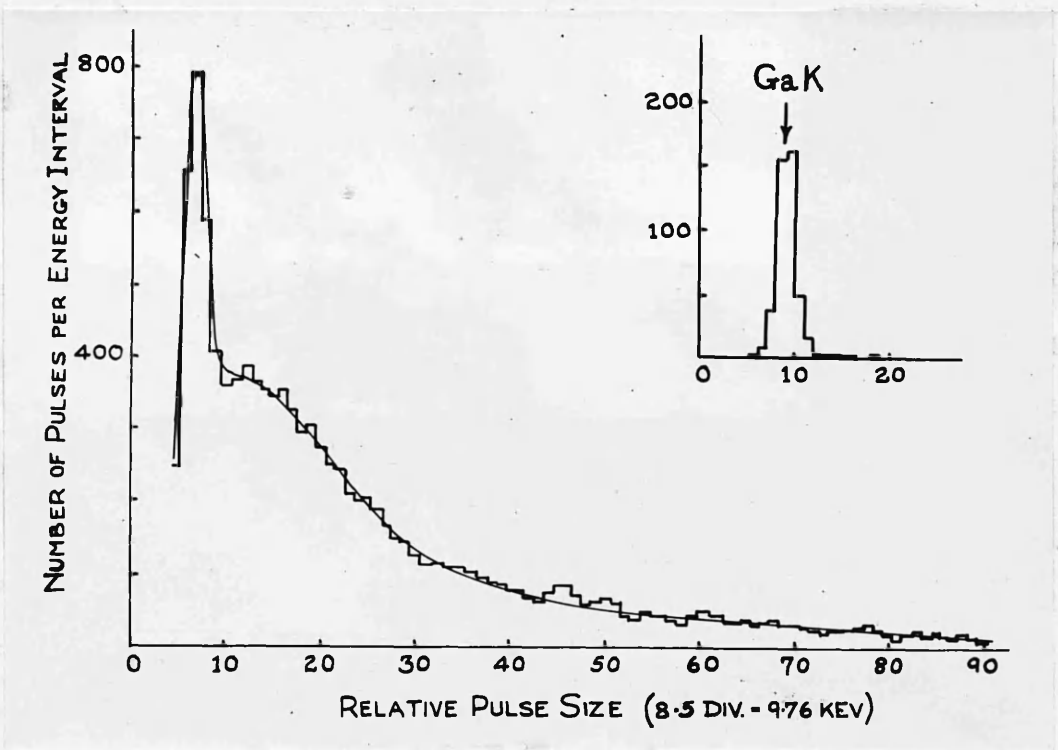


Fig. 21. Spectrum of radiations up to ~100 KeV.

irradiated by slow neutrons, since there exists a stable isotope, Ni^{58}, which is present in ordinary nickel to the extent of 67.67% and which has a reasonably high cross-section for thermal neutron capture. According to Ross and Story (1948-1949), the absorption cross-section of nickel for thermal neutrons is between 4.2 and 4.6 barns and is mainly due to Ni^{58}. As had been mentioned before, Ni^{59} would be expected to decay by positron emission or by K-capture, or both, to give $Co.^{59}$. The search for annihilation radiation gave a negative result, so it was more likely to decay by K-capture. However the low intensity of X-radiation found previously showed that the half life of Ni^{59}, if it existed, must be very long indeed.

The counter used in the examination of Ni^{63} was also used for this experiment. A source giving a counting rate, into 2π, of 34,000 per minute was inserted and covered with an aluminuim foil of thickness 1 mil. This was sufficient to stop all the beta-radiations of Ni^{63}. The counter was filled to a total pressure of two atmospheres. With the absorber foil the counting rate was reduced to the background rate of the counter, within statistical fluctuations, but nevertheless it was thought desirable to examine the pulse distribution carefully. The results for two energy ranges are shown in figs. 20 and 21. Fig.20 was taken at high gain (extending to ~30 KeV) and fig. 21 at low gain (to ~100 KeV). A peak is clearly seen in both histograms, superposed on the spectrum of the counter background. The K-capture radiation of Ge^{71} (X-rays of Ga^{71}) was used for calibration in each case (inset figures). The Ga^{71} X-rays passed through Perspex of such a thickness that the K_α and the K_β intensities were approximately equal. The K_α and the K_β peaks are shown slightly resolved in fig.20. Taking the mean energy of these radiations as 9.76 KeV, the

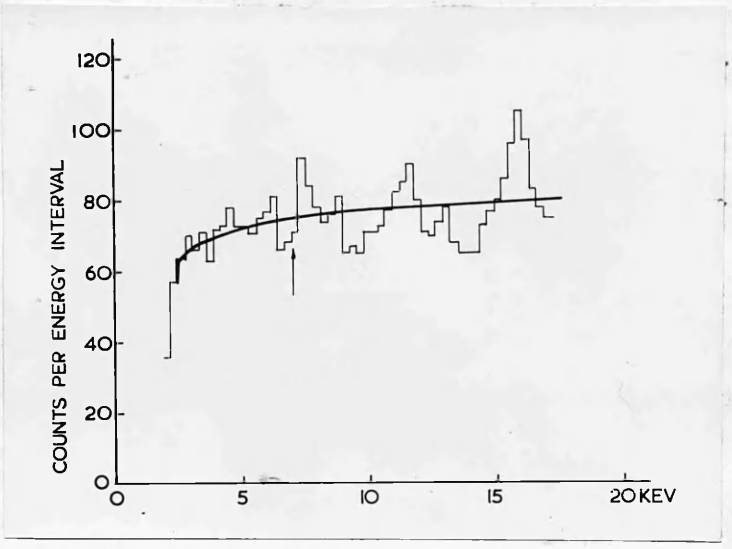


Fig. 22. Spectrum of counter background.

energy corresponding to the peak observed on the spectra is 6.9 ± 0.3 KeV. The K_α X-ray energy of cobalt is 6.93 KeV. Because of the nearly perfect agreement between these values, it is almost certain that the radiations observed are due to Ni^{59} decaying by K-capture to Co^{59}, and emitting the X-rays of cobalt.

To show that the peak was due to the radioactive nickel, the source was removed from the counter and replaced by a thickness of aluminium equivalent to the source mount and absorbing foil. This gave the true background spectrum of the counter which was analysed and is shown in fig.22 The spectrum shows no evidence whatsoever of a peak at the energy corresponding to the peak observed in the previous case.

The radiation is of very low intensity indeed. By counting the number of pulses under the peak of fig.20, and subtracting the background, the counting rate was estimated to be ~22 per minute. Allowing for the efficiency of the counter and its solid angle of 2π, this corresponds to 49 disintegrations per minute in a source weighing approximately 17.3 mgm. This very low activity explains why it has escaped detection hitherto. The proportional counter technique is possibly unique in its ability to reveal and give accurately the energy of a radiation of such low intensity and energy. The half life was estimated at a later date (see later) to be 7.5×10^5 years. This work was published by the author (Wilson, 1950).

About this time Thomas and Kurbatov (1950) described a search which they had made for photons emitted by long lived nickel isotopes by the absorption method and, by cloud chamber, for the continuous spectrum of Ni^{63}. They found evidence of the former but not of the latter activity. However, the experiments just described showed that a relatively intense

beta-emission of Ni^{63} is accompanied by a weak K-capture emission of Ni^{59}. The ratio of the activities is about 1200:1. It is evident that, if they detected photons from nickel, they should also have observed the relatively much stronger activity of Ni^{63}. It seems very probable that the radiations which they detected were spurious and due to impurities in the source. The work done in Glasgow showed that very careful chemical purification of the source is required and the results of Thomas and Kurbatov fully justify the preparation of nickel carbonyl as an intermediate step in spite of the difficulty involved and thus, although the attempt to use the carbonyl vapour itself as a source was unsuccessful, its manufacture proved well worth while.

10 Search for other Electromagnetic Radiations.

The values obtained by Thomas and Kurbatov by absorption for the energies of the photons observed were as follows : 7.5 ± 1 KeV, 15 ± 2 KeV, 38 ± 3 KeV and 80 ± 5 KeV in the intensity ratios 6:1:1:1 and possibly one of greater than 500 KeV. As it was possible, though not likely, that radiations corresponding to these values might exist in our source, but of such an intensity that they could not have been observed in the spectra obtained, it was decided to use a much stronger source. Accordingly a source was prepared from nickel irradiated at a much higher neutron flux density, $\sim 10^{10}$ neutrons per cm^2 per second. Also the technique of depositing the nickel on the foil was modified as described in section 2 so that a much larger mass was obtained. With this foil in the counter, the beta-counting rate was too high to measure with the scalers in use. It was believed to be in excess of 500,000 per minute and had an end-point corresponding to the Ni^{63} spectrum. The source was covered with an absorber to

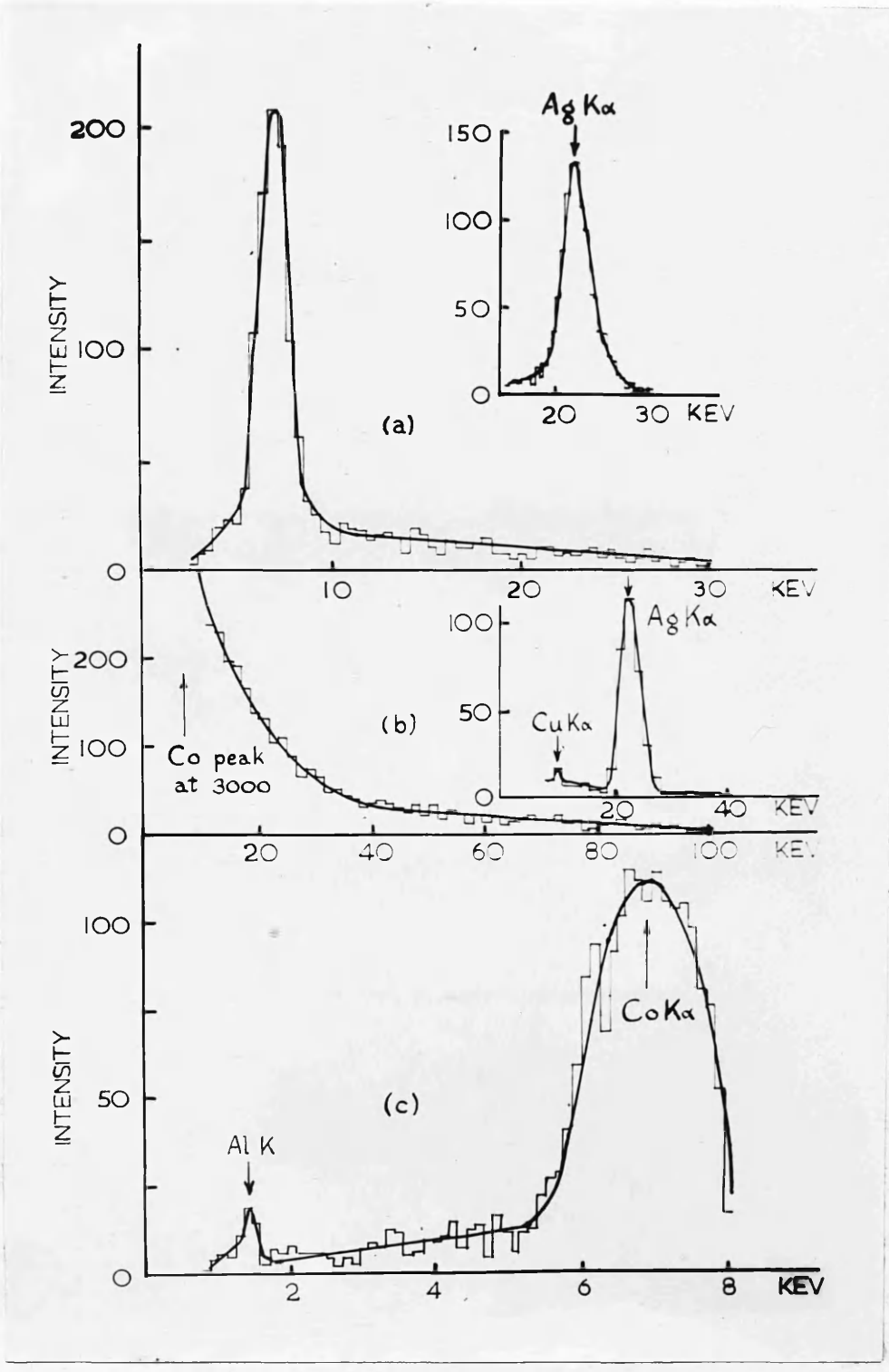


Fig. 23. Histograms of radiations of long lived nickel.

cut out the beta-rays and a monoenergetic grouping of pulses corresponding to the X-rays of cobalt was observed. The ratio of the intensities of the present and former sources was measured to be 21:1. The radiations were analysed, using the K X-rays of copper for calibration, and the results are shown in fig.23. Graph (a) covers the region 0-30 KeV; graph (b), 0-100 KeV and graph (c), under 8 KeV. A very strong peak due to K-capture in nickel (X-rays of cobalt) was observed as before and the energy of this peak (6.9± 0.1 KeV) was in perfect agreement with the KX-ray energy of cobalt. The only other radiation observed was taken to be the K X-rays of aluminium excited by the cobalt X-rays (graph (c)). No evidence of the gamma-rays of Thomas and Kurbatov could be detected. Allowing for the efficiency of the counter, the 15 KeV and 38 KeV gamma-rays could have been detected if their intensities had been more than one-twentieth and one-tenth of the intensity of the cobalt X-rays, respectively. The 80 KeV radiation would have been detected only if its intensity was of the same order as that of the X-rays.

The results (Wilson, 1951) confirm the existence of the K-capture radiation of Ni^{59} but indicate strongly that the gamma-rays detected by Thomas and Kurbatov arise from the presence of some contaminant in their source.

11. The Half Lives of Ni^{59} and Ni^{63}.

The half life of a radio-isotope prepared by neutron capture can be determined by the so-called "yield method", if one knows the mean neutron flux density which produced the isotope, F say, the isotopic abundance, K, the neutron capture cross-section of the parent nuclide, σ, the time of irradiation, t, and the specific activity of the source produced. If the

source is long lived (half life much greater than the time of irradiation) we may write

$$\tau_{1/2} = 0.693 \frac{N_0}{\Delta N}$$

where $\tau_{1/2}$ is the half life, ΔN is the number of disintegrations per second in a mass, M gm say, of source and No is the number of atoms of the radioactive isotope in the source which is given by

$$No = 6.02 \times 10^{23} \times FMK\sigma t/A$$

where No, F, K, σ and t are defined as above and A is the mass number of the parent nuclide. In the case of Ni^{58}, Ross and Story (1948-1949) gave the thermal neutron cross-section of nickel as 4.2-4.8 barns and stated that this absorption is due mainly to Ni^{58}. Later work (Senftle and Leavitt 1950, Pomerance 1949) gave the values of 4.2 and 4.17 ± 10% respectively. In the case of Ni^{62}, Segre (1948) quoted a value of 0.51 barns, on the basis, however, of a wrong mass assignment of the 2.6 hr. activity of nickel. More recent work (Senftle and Leavitt 1950, Pomerance 1949) gave 15 and 14.8 ± 10% respectively. Using these values, the half lives were calculated to be (Wilson, 1951)

Ni^{59} : 7.5×10^5 yr.
Ni^{63} : 61 yr.

These values disagreed somewhat with the half lives quoted by Seaborg and Perlman (1948), who gave 5×10^5 yr for Ni^{59} and 300 yr for Ni^{63}. Later work by Friedlander (1949) and Brosi and Griess (1949), in unpublished reports, give the half life of Ni^{59} as $2\text{-}3 \times 10^5$ yr and 1.5×10^5 yr respectively, while Brosi and Griess give 85 yr for the half life of Ni^{63}.

The estimation of the activity was made as follows:- About 3 mgm of very pure radioactive nickel was weighed accurately, converted to nitrate

and deposited evenly on a sheet of 0.002" aluminium over an area 6cm x 8 cm by the insulin technique, described in the introduction. The foil was inserted into the centre of the proportional counter and the counting rate obtained at fairly high gain. The counting rate, less background, when corrected for solid angle and reflection at the source mount, gave ΔN. The value of F was obtained from A.E.R.E., Harwell and, since it was known, the half life could be calculated.

Since this work was published, there appeared a paper by Brosi, Borkowski, Conn and Griess (1951) on the long lived activities of nickel. Their findings confirm in the main, the work carried out in Glasgow. They used, for the analysis of the beta-rays, a 10 $\mu gm/cm^2$ source on a platinum foil. In spite of the high Z source mount used, a linear Fermi plot was obtained which gave a value of Eo of 67 $\pm$ 2 Kev. The half life was estimated to be 85 yr. This is probably the same determination as that due to Brosi and Griess, mentioned earlier.

This concludes the work on the radio-activity of nickel. To summarise, the charge, mass assignment, upper energy limit, spectrum shape and half life of the long lived nickel beta-activity have been found and it has been shown that no X-rays or gamma-rays accompany the decay. The existence of Ni^{59} has been demonstrated and its half life measured.

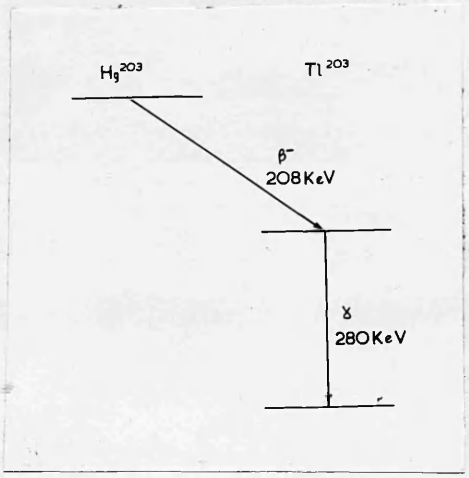


Fig.24. Decay scheme of Hg^{203}.

PART 3. RADIOACTIVITY OF MERCURY 203.

1. Introductory.

As has been mentioned in part 1 of this thesis, the integrating property of the proportional counter is one which can be of very considerable advantage in the elucidation of spectrum shapes, decay schemes and other features of radioactive decays. This property depends on the fact that, if two transitions, which are in coincidence, occurring within say $< 10^{-6}$ seconds, are both detected by the counter, the output pulse energy is equal to the sum of the pulse energies of the individual transitions. This feature can at times be a disadvantage, but in the case of a decay such as Hg^{203}, RaD and many others, it is a very useful property. The first use made of the property was in the examination of the decay of Hg^{203}, a decay which was of interest in itself and also one well suited to the application of the integrating method. In the following section the results of previous work are outlined and the reasons which made it desirable to investigate the activity for its own sake, while the third section will give the theory of the integrating action in more detail.

2. The radioactivity of Hg^{203}.

The radioactivity of Hg^{203} has been studied by a number of workers whose results will now be reviewed briefly. The essential features of the decay scheme are represented in fig.24. A beta-transition of energy $\sim$ 208 KeV is followed by a gamma-ray of $\sim$280 KeV which is strongly internally converted. No direct transition to the ground state of Tl^{203} has been observed. The first investigation of this activity was by Friedlander and Wu (1943) who obtained a value for E_o of 460 KeV by absorption in aluminium. They also reported a half life of 51.5 days. Millar and Curtiss (1945) measured the maximum beta-energy by spectrometer and found it to be less than

300 KeV. They also found a gamma-ray of ~280 KeV accompanying the decay. Saxon (1948) carried out a fairly careful magnetic spectrometer study of the radiations using a source of thickness ~2.4 mgm/cm^2 on a nylon backing of 0.54 mgm/cm^2 and counter window thicknesses of 0.05 and 0.2 mgm/cm^2. It was impossible to observe the end-point of the beta-spectrum due to the fact that the strong K conversion line of the 280 KeV gamma-ray is superposed on the high energy tail of the spectrum. However Saxon concluded the spectrum to have an allowed shape over the limited range from ~40 KeV to ~140 KeV and by extrapolation of the Fermi plot obtained an energy limit of ~205 KeV. The gamma-ray energy was found to be ~286 KeV. The internal conversion coefficients were estimated to be $\alpha_K = 18\%$; $\alpha_L = 6\%$; $\alpha_M < 0.5\%$. Hence $\alpha_K/\alpha_L = 3$; $\alpha_L/\alpha_M > 12$. He obtained a value of 43.5 days for the half life. Slätis and Siegbahn (1949) eliminated the conversion line by measuring only those beta-particles which were in coincidence with gamma-rays. However a small peak remained due to K photoelectrons which were in coincidence with their own X-rays. Nevertheless these workers were able to show that the spectrum was of allowed shape over a larger range than had been possible before and concluded that E_0 was about 208 KeV. The gamma-ray energy was estimated to be 279 KeV. and the internal conversion coefficient ratios were $\alpha_K/\alpha_L \sim 3$ and $\alpha_L/\alpha_M \sim 2$. The latter figure is an estimate of the writer's on the basis of the published graphs.

It is clear from the above that a certain amount of uncertainty existed in some of the data. For example it is difficult to obtain a precise value for the upper energy limit of the beta-spectrum, due to the conversion lines. One cannot compare the shape of the spectrum with theory at energies near the end-point. Also the low energy region of the spectrum had not been investigated. A considerable variation existed in the quoted

values of the half life (43.5 - 51.5 days). The internal conversion coefficients were not very well known and their ratios, as quoted by Saxon and Slätis and Siegbahn, differ somewhat. It was therefore of importance to examine the decay in more detail with the means at our disposal which were able to resolve many of these questions.

Also relatively soft beta-emitters of high Z-value such as Hg^{203} had not been examined in detail in the past and it was believed possible that they could exhibit rather unusual features in this region. Thus one might expect rather large effects from the atomic electrons. Also radiative transitions of the type discussed by Ivanenko and Lebedev (1950) might be prominent. They suggested that sometimes beta-particles might be captured into unoccupied levels of the outer electron shell, with the emission of a photon. This could reduce the probability of escape of beta-particles by several tenths for very soft spectra of heavy atoms and it is claimed that it could account quantitatively for experimental results of work on the radioactivities of MsTh1 and MsTh2. Since Hg^{203} fulfils both the conditions of high atomic number and low maximum beta-ray energy, it is evident that the spectrum shape might be appreciably affected due to this cause.

Schwartz (1952) has very recently discussed the possibility that a negative beta-spectrum may be affected at the lower energy end by the energy liberated as a result of the readjustment of the atomic electron system due to the change in nuclear charge. In the case of Hg^{203} the energy release is $\sim$15 KeV. If all, or most, of this energy were carried off by the beta-particles, the spectrum would possess a "hole" between zero energy and $\sim$15 KeV. However, although earlier writers considered the energy would be taken up in this way, Schwartz does not believe this likely. A study of the shape of the Hg^{203} spectrum at low energies would help to

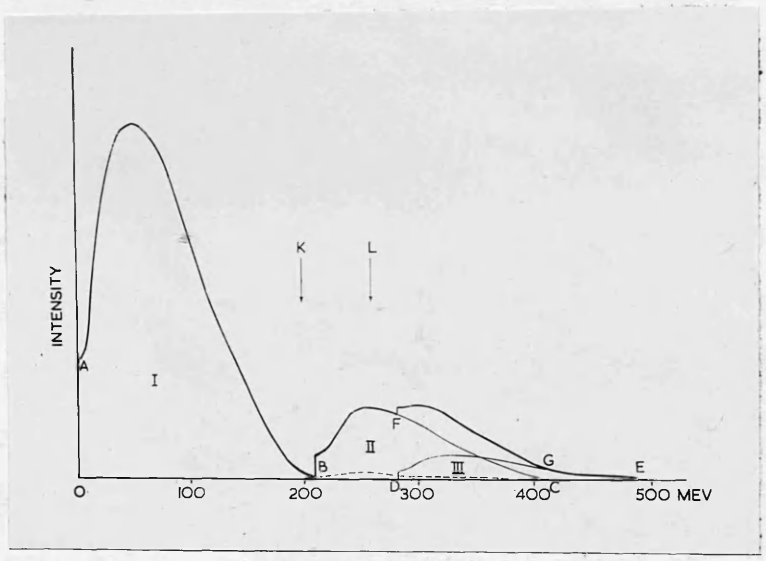


Fig. 25. Modified spectrum formed by integrating action of the proportional counter. A hypothetical primary spectrum shape (group I) was assumed.

answer this question.

3. The Integrating Properties of a Proportional Counter.

Consider the decay scheme of fig.24. If the source is situated within a proportional counter (solid angle of 4π), three distinct possibilities arise which are as follows.

(i) The unconverted gamma-rays largely escape from the detector since the efficiency of detection of such radiations is $<1\%$, while all the beta-rays of the primary spectrum are recorded. This gives a normal spectrum distribution extending to about 208 KeV.

(ii) Some of the gamma-rays convert in the K shell and photoelectrons of energy $\sim$200 KeV are emitted in coincidence with the primary beta-particles. This gives rise to a spectrum of the same shape as the primary spectrum but displaced along the energy axis by 200 KeV. However since the conversion of the gamma-rays in the K shell normally gives rise to L, M, N, ... X-radiations together with K X-radiations and since the former are detected with almost 100% efficiency, the actual displacement of ~~group (i) and~~ group (ii) will usually be $h\nu_{\gamma} - h\nu_{K\alpha}$, i.e. 208 KeV.

(iii) The gamma-rays will also convert in the L,M,N.....shells giving rise to the corresponding photoelectrons and X-rays. This gives rise to a spectrum due to the beta-rays in coincidence with these radiations and since all the photoelectrons and practically all the L, M, N... X-rays will be captured, the spectrum will be displaced by the full energy of the gamma-ray, viz. 280 KeV. Since the efficiency of detection of the K X-rays of thallium is about 2% in the counter and experimental arrangement used, occasionally processes of group (ii) will be displaced into group (iii)

Fig.25 shows diagramatically the kind of resultant spectrum shape one might expect. The area marked I corresponds to group (i) and therefore to

the primary spectrum. For this purpose, a hypothetical spectrum shape has been assumed. The area marked II corresponds to the group (ii) pulses and is displaced along the energy axis by 208 KeV (to a point coincident, by chance, with the end-point of the primary spectrum). Area III corresponds to the third group which is displaced by 280 KeV. Naturally the spectrum due to this group must be added to that due to the second group so that the final shape of the histogram obtained would be given by the uppermost line, viz. ABFGE, provided the detector has perfect resolution at all energies.

The advantages of the method are immediately obvious. (a) The group (i) pulses give the shape of the spectrum except near the upper and lower energy limits. (b) The beginning of the group (ii) pulse distribution gives the shape of the spectrum at low energies in detail, free from many of the difficulties of making measurements in this region. (Ofcourse, the source must still be thin and on thin backing). (c) The portion GE of the composite spectrum gives the true shape of the end-point region of the spectrum free from confusion due to electron lines. (d) The upper energy limit of the spectrum can be estimated from the point E, provided the gamma-ray energy is known. It was re-measured in the course of this work yielding a result in agreement (within experimental error) with that of Slätis and Siegbahn. Thus the whole spectrum can be examined in detail and compared with theory. (e) The relative intensities of the three groups can be estimated by comparison of the three areas and so one can determine the K and L, M,.... conversion coefficients rather precisely. To carry out this operation, the line FC is drawn in the spectrum by comparison with the group (i) pulse distribution, or by calculation from the Fermi plot.

In the following series of experiments the spectrum shape was examined in detail and compared with theory from 20 KeV to the end-point, whose

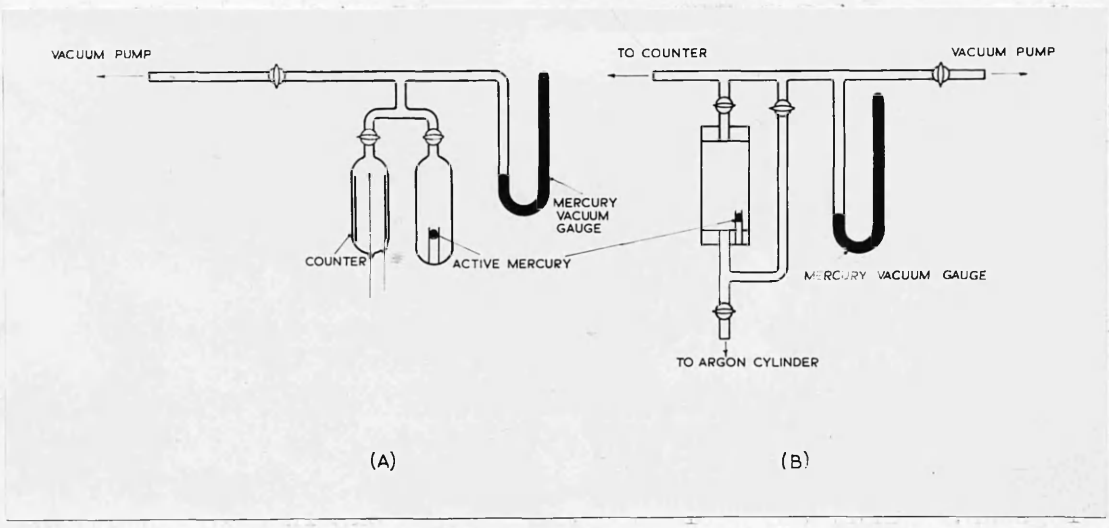


Fig. 26. Experimental arrangements used to measure the beta-activity of the radioactive mercury vapour.

energy was found both by Fermi plot and also directly. The energy of the gamma-ray was measured using a magnetic spectrometer and its internal conversion coefficients evaluated as just described. The half life was measured with considerable accuracy. A discussion of the decay scheme with reference to spin and parity changes is given towards the end of this part, which is concluded by pointing out further possible uses of the technique, including its extension to scintillation counters, which also possess the integrating property.

4. Source Preparation.

The sources used in the following studies were prepared by slow neutron irradiation of, firstly, pure mercuric oxide and later, pure distilled mercury. This was carried out in the pile at A.E.R.E., Harwell. In the earliest experiments the source consisted usually of a thin layer of mercuric oxide spread over a perspex support to which the oxide was caused to adhere by spreading a thin layer of amyl acetate cement over the support. In another of these experiments, the source was deposited on a thin aluminium wire which was then used as the central wire of a proportional counter. Aluminium wire was used to provide a low-Z support. However not much success attended these early experiments and it was found to be largely due to certain faults in the counter.

It was then believed that it might be possible to use the vapour of metallic mercury as a gaseous source. Tests to determine the source strength available were made and, since these indicated that the method was feasible, a high pressure, steel-walled counter was designed for this purpose. The tests were carried out using the experimental arrangements sketched in fig. 26. In the first case, fig.26 (a) a small globule of

radioactive mercury was contained in a short tube in a vessel, which, together with a Geiger counter attached to the system, was evacuated. The mercury vessel, which had a volume equal to that of the counter, was then filled with argon to twice the pressure normally used in the Geiger counter and the gas (including mercury vapour) in the mercury vessel was shared with the counter. The specific activity of the mercury vapour was then estimated since its vapour pressure at room temperature was known. However the rate was not nearly as much as previous rough calculation had suggested and therefore a new technique was tried. The apparatus employed is shown in fig. 26(b). In this case the vessel containing the mercury was thoroughly evacuated and then left for $2\frac{1}{2}$ hours. It was hoped that, by this time, the vapour pressure of the mercury would be at equilibrium throughout the volume at its saturation value. Then the Geiger counter, already filled to 1.5 cm of alcohol, was filled to its usual pressure of argon (10 cm), the argon being passed through the mercury vessel thus sweeping out the mercury vapour with it. The counting rate of the Geiger counter, in excess of the background rate, was found and the specific activity calculated. This procedure was repeated several times as described except that the mercury vessel was left evacuated for increasing periods, to ensure that equilibrium was reached. From the results it was calculated that a counter 6' long and 4" in diameter, if filled with mercury vapour at the saturation vapour pressure at $18^{\circ}C$, i.e. 0.001009mm, would give a counting rate of ~40,000 counts per minute. This was evidently a practicable method of carrying out the spectrum analysis from the point of view of intensity. Since these experiments were carried out a value for the slow neutron capture cross-section of Hg^{202} has been published and a calculation of the counting rate expected after a four week bombardment at a neutron flux

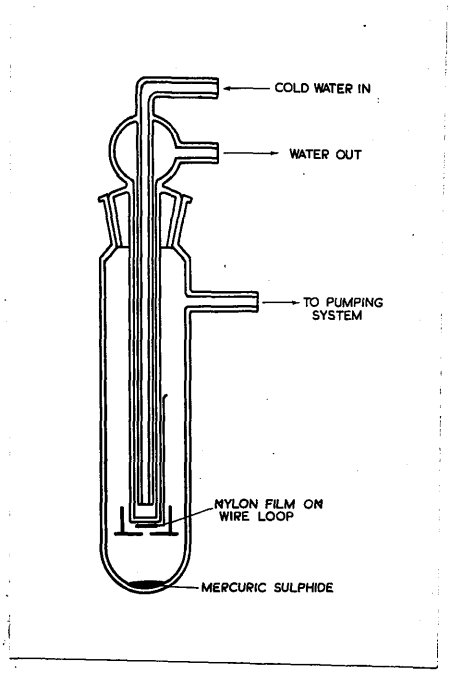


Fig. 27. Apparatus used for the preparation and evaporation of mercuric sulphide.

of 10^{11} per cm^2 per second, gives ~32,000 per minute. The flux was usually 1-3 times the value quoted so that the calculated value agrees reasonably well with that obtained experimentally.

However the main analysis was carried out using a solid source in a counter which was placed in a magnetic field, as shown later in fig. 31. The source was prepared by the evaporation technique which, as we have seen, gives a very uniform source thickness. In the first attempts mercuric chloride (corrosive sublimate) was used since its sublimation temperature (302°C) was fairly low. However it had a harmful effect on the source mounting materials used, viz. aluminium foil and nylon film, causing them to break. Finally mercuric sulphide, which evaporates at normal pressure at 583°C, was found to be satisfactory, in that it had no bad effects on the source mounts. The apparatus used is sketched in fig. 27. The metallic mercury was dissolved in concentrated nitric acid in the outer tube and evaporated very gently to dryness. The residue was dissolved in water and again evaporated to dryness to get rid of any acid. The mercuric nitrate was again dissolved in water, a little hydrochloric acid added and hydrogen sulphide bubbled through till the mercury was completely precipitated as sulphide. The supernatant liquid was decanted off and the source dried. The inner tube was then inserted. This consisted of a water cooling tube against which was held the source mount, which consisted, in one experiment, of a nylon foil supported by a wire loop and, in another experiment, of 0.16 mgm/cm^2 aluminium foil supported in a similar manner. To localise the source on the central zone of the foil, away from the wire loop, a mask with a small circular hole was held over the source loop. Cold water was passed through the cooling tube and the outer tube was evacuated by normal high vacuum methods. The source was now heated till

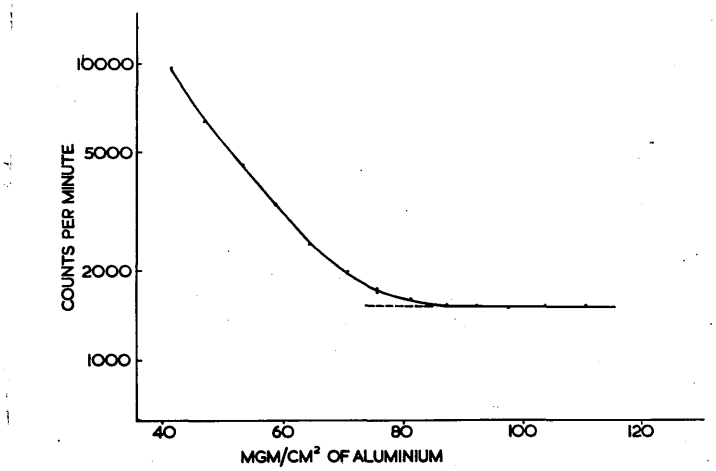
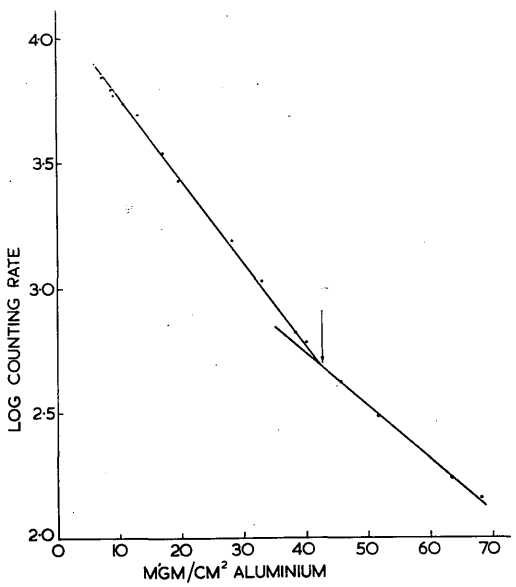


Fig. 28. Absorption of beta-rays in aluminium.



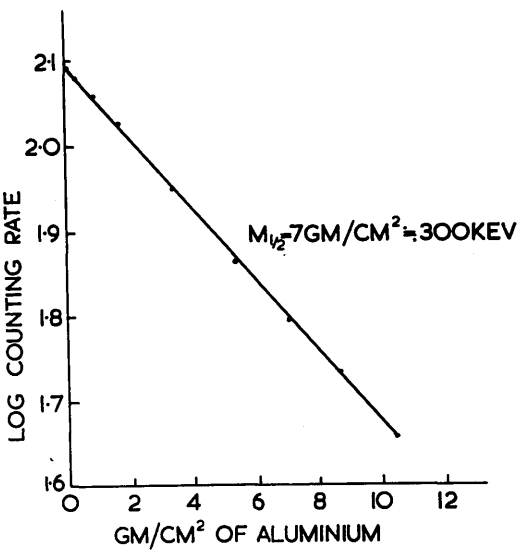


Fig. 29. Fig. 30.

Fig. 29. Absorption of beta-rays in aluminium.

Fig. 30. Absorption of gamma-rays in aluminium.

sublimation took place. This gave, in each case, an almost invisible source, which in the case of the aluminium foil source mount, had a mass of about 3 μgm/cm^2 and a total activity of ~12,000 disintegrations per minute. The nylon film source had an activity approximately one third less.

5. Preliminary Experiments.

A series of absorption experiments were carried out in the course of a preliminary examination of the activity. In the first instance these experiments were carried out to confirm previous findings and also to ensure that the activity observed in the source was due largely to Hg^{203}. In addition to this, however, further information was elicited from the curves obtained, with regard to the possible existence of a higher energy ground state-to-ground state transition. Fig.28 shows an absorption curve giving the detail at its end-point. The absorber mass scale has been corrected for counter window thickness and air gap (total ~6 mgm/cm^2). A good end-point is apparent and according to Glendenin (1948), the range corresponds to an energy of 280 KeV. This confirms the gamma-ray energy measurement which was made later since there are photoelectrons present having practically all the quantum energy. Fig. 29 shows a distinct break about 42 mgm/cm^2 which corresponds to an energy of ~200 KeV, i.e. the limiting energy of the beta-rays. The counting rate beyond this value is mainly due to photoelectrons. Fig.30 is an aluminium absorption curve of the gamma-rays emitted by the source. This yields a good straight line on a logarithmic plot and the gradient corresponds to a gamma-ray energy of ~ 300 KeV which is in good agreement with previous and present work. From these results it appears that the activity is almost entirely due to Hg^{203}, that the results obtained are in agreement with those previously

quoted, and that there is no evidence of a ground-to-ground transition which would have an upper energy limit of $\sim$490 KeV, or $\sim$160 mgm/cm^2. Confirmation of the last comes from other experiments, especially the magnetic spectrometer results which showed no beta-activity beyond the highest energy photoelectron peak at $\sim$280 KeV. Indeed it is believed that the transition to the excited state is more probable than the transition to the ground state by a factor >100.

A number of investigations on the beta-spectrum shape were made using a brass-walled, aluminium-lined counter of active length 10.5" and diameter 5.5". At first the source was in the form of a thin layer of mercuric oxide on a dystrene source holder which was inserted through a window situated in the middle of the counter wall. Since the spectrum shape obtained was not very satisfactory, a new method of mounting the source was tried. A thin layer of source was deposited on a perspex backing which was held by a steel rod in the middle of the counter, 0.875" from, and accurately parallel to, the wire. The correct voltage, corresponding to its position in the field was applied to it. Again the spectrum shape obtained was unsatisfactory and the source coated aluminium wire described in section 4 was inserted into the counter as the counting wire. Finally in a fourth attempt, active mercury was electroplated on to the central wire of the counter. Much labour was expended in analysing the results of these experiments but, although the energy distributions obtained possessed a certain similarity to fig.25, the energy resolution was quite inadequate. This was confirmed by the broadness of the calibration peaks. It was evident that the counter was responsible for the poor results obtained and repeated efforts were made to improve the situation by thoroughly cleaning the counter, using carefully purified gases and so on

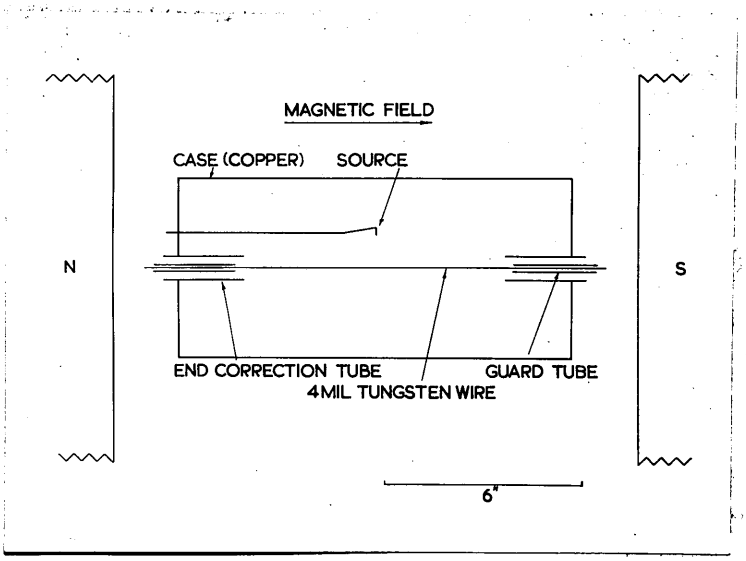


Fig. 31. Experimental arrangement of proportional counter in magnetic field.

without success. Finally the counter was completely rebuilt, only the cylindrical case and end-plates being retained. Advantage was taken to incorporate end-correction tubes. After this the counter operated perfectly and was used by the writer in studies on the long lived activities of samarium and europium. Meanwhile, however, the activity was investigated using another counter specially built by A.L. Cockroft for use in magnetic fields. This work is described in the following section.

6. The Beta-Ray Spectrum of Hg^{203}.

The experimental arrangement for the examination of the spectrum was as shown in fig. 31. The counter, which was end-corrected and had an active length of 8" and a diameter of 5.5" was placed within a uniform magnetic field of 1400 gauss, parallel to the wire. The source was mounted on a separate probe terminating in a circular loop $\frac{1}{4}$" in diameter. The plane of the loop was parallel to the end plates of the counter. As was mentioned earlier, the loop supported a thin foil of aluminium (0.16 mgm/cm^2) on which was sublimed a 3 μgm/cm^2 thickness of mercuric sulphide having a total activity of ~12,000 counts per minute. The probe was at a distance of 1.1" from the wire and the voltage applied to it was adjusted to the correct value corresponding to its position in the counter. The source was confined to the centre of the counter and the recorded rays were emitted into a solid angle of 4π, half the number, of course, passing through the thin supporting film. The counter was filled with methane to a partial pressure of 20 cm of mercury and argon to a total pressure of 3.75 atmospheres. The pressure and magnetic field were such that no particles emitted by the source could reach the cylindrical wall or escape into the non-counting volumes at the ends of the counter.

The counter was calibrated by use of the K_β radiations of tungsten,

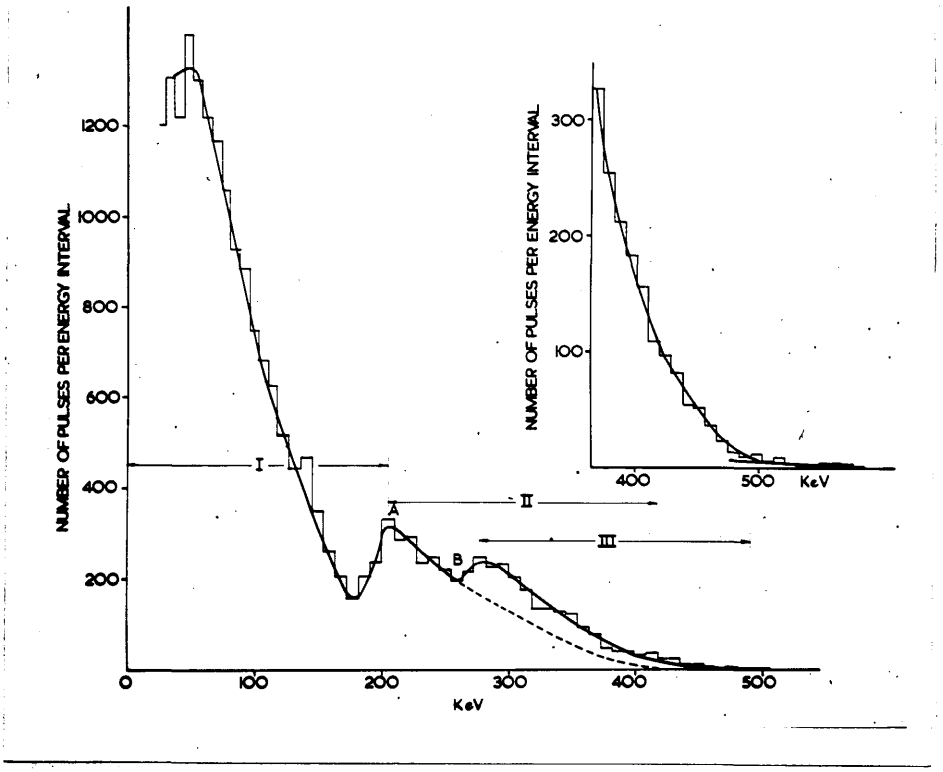


Fig. 32. Complete spectrum of pulses observed in proportional counter. Detail near end point shown in inset figure.

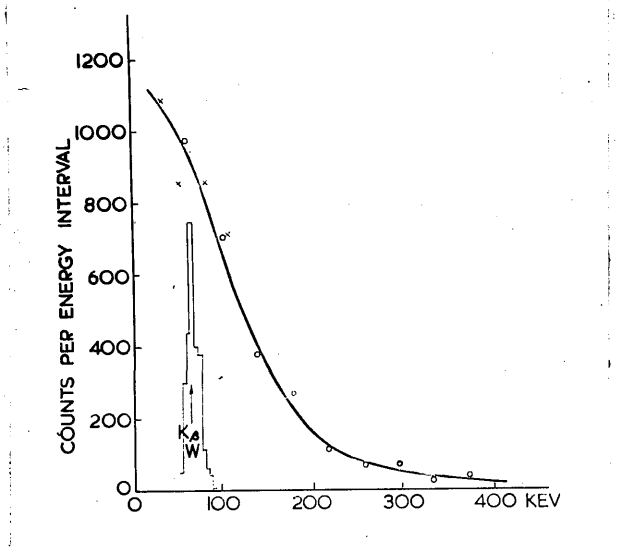


Fig. 33. Spectrum of background pulses in proportional counter.

obtained by firing the X-rays from the X-ray tube directly at the wall of the counter as described in part 1B, section 11. The pulses were recorded in two ways, viz. (i) by photographing on a moving film the pulses displayed on a cathode ray oscilloscope and (ii) by passing the pulses into a single channel kicksorter. Both methods gave very similar results and the form of the spectrum obtained is shown in fig. 32. It is evident that the spectrum is repeated as expected (see fig.25); groups I, II and III correspond to the groups similarly marked in fig.25. The end-point of the spectrum is shown in more detail in the inset figure. No serious effort was made to carry the analysis of group I particles to very low energies since the beginning of group II gives the shape in the very low energy region. Nevertheless the spectrum was examined at higher gain to give the shape of the low energy spectrum more precisely and the spectrum shape below 130 KeV in fig. 32 is taken from this examination.

The spectrum shape includes the background particle spectrum (~ 700 counts per minute). This was deducted in making the calculations for the Fermi plot, fig. 34, and in calculating the internal conversion coefficients. The shape of the background spectrum is shown in fig. 33. The calibration radiation, $K\beta$ X-rays of tungsten, is included. The points marked "x" were obtained at four times the gain at which the points marked "o" were observed.

7. Fermi Plots.

The Fermi plots are shown in fig.34. This figure appeared in an account of this work published in the Philosophical Magazine (Wilson and Curran, 1951) and the fig. 3 referred to in the figure is fig. 32 of this thesis. The Fermi plots were made as follows: (i) The middle range, marked with circles, is taken from the group I pulses. It will be noticed

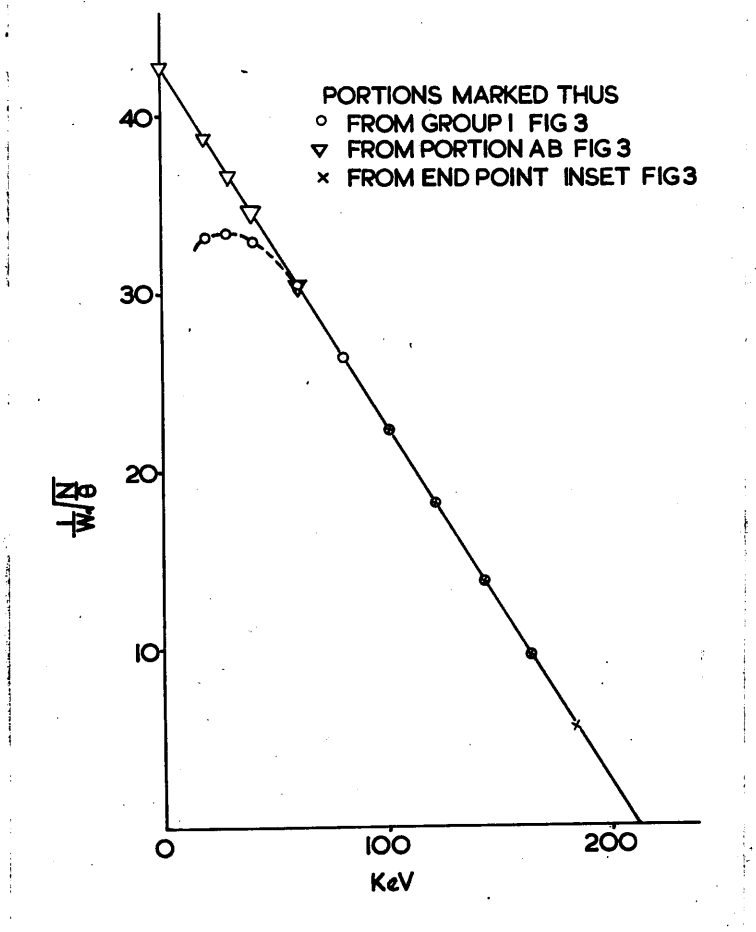


Fig. 34. Composite Fermi plot for Hg^{203}.

that the line obtained deviates from straightness at low energies due to the difficulties of analysis at low energies. (ii) The end-point region, marked with crosses, is taken from the high energy region of fig. 32 (inset figure). This region lies on the same straight line as the main portion and the freedom from distortion due to photoelectron lines, observed in magnetic spectrometer studies, will be noticed. (iii) The low energy region, indicated by triangles, was obtained from the portion AB of fig. 32, i.e. the start of the second spectrum. The Fermi plot was calculated by the Bleuler and Zünti method as described in Part I. The fitting together of the three regions results in an excellent straight line with an end-point at 210 ± 5 KeV. The end-point energy is given more precisely by the limiting energy of the spectrum, provided the energy of the gamma-radiation is known. This was found to be 278 KeV, as described later, and the end-point was again calculated to be 210 ± 5 KeV. This agrees with the value given by Slätis and Siegbahn. The Fermi plot is shown as linear to zero energy, the point at 0 KeV being taken from the corresponding point on AB, fig. 32. However in view of the source support thickness, this linearity to such a low energy must be regarded as coincidental.

However, there is no evidence of any fall-off in intensity at the lower energy end of the spectrum. This supports the view of Schwartz (see section 2) that the atomic binding energy liberated is not carried off, mainly, by the electrons. The neutrino appears to carry off most of the energy. It is quite probable, in fact, that the energy is shared between the electron and the neutrino in much the same way as the nuclear transformation energy is shared.

8. The Internal Conversion Coefficients.

As has been mentioned it is easy to calculate the internal conversion coefficients using this technique. In fig. 32 the second spectrum is continued, as a dotted line, by comparison with the main spectrum. By comparing areas under the second and third spectra, (corresponding to the areas BFCB and FGECF of fig. 25) with the area under the main spectrum the internal conversion coefficients (defined as the number of conversion electrons divided by the total number of gamma-ray disintegrations) were found to be

$\alpha_K = 15.6\%$; $\alpha_{L,M..} = 4.2\%$; $\alpha = 19.8\%$.

The ratio $\alpha_K / \alpha_{L,M..}$ is 3.71 . Saxon gives the value of α as 0.24 . If we define α in the more usual way as the ratio of the number of conversion electrons to the number of gamma-rays emitted, our value of α rises to 0.247, in good agreement with Saxon. The work of Slätis and Siegbahn yields a ratio of 3.0 for $\alpha_K / \alpha_{L,M}...$.

Taylor and Mott (1932) predicted a value for α of $\sim 14\%$ for the internal conversion of electric quadrupole radiation of energy $mc^2/h\upsilon = 1.83$ and $Z = 83$. Since in this case $Z = 81$, these results strongly suggest that the gamma-ray transition is electric quadrupole in type. This result is of importance in a discussion of spin assignments which will be made later.

9. Energy of the Gamma-Radiation.

The gamma-transition energy was measured using the semi-circular magnetic spectrometer designed by Dr. E.R. Rae. The resolution of this instrument is 2%. The detector consists of two Geiger counters, situated one behind the other, operating in coincidence, which results in a very low background counting rate. This was desirable since the original total

source strength available was rather small, viz. ~30 microcuries. Two sets of measurements were made. In the first, the mercury source, which was contained in a small glass tube, was surrounded by a 50 mgm/cm^2 lead foil. In the second, this was replaced by a much thinner platinum radiator, having a thickness of 4 mgm/cm^2. The electron spectrum obtained using the platinum radiator is shown in fig. 35. From the strong K and L conversion electron peaks which were observed, the gamma-ray energy was estimated to be 278 ± 3 KeV. This is in good agreement with the result of Slätis and Siegbahn. Saxon's result (286 KeV) appears to be a little high.

10. The Half Life of Hg^{203}.

In view of the considerable variation in the value of the half life as determined by different workers (43.5 - 51.5 days), a careful measurement was made. The apparatus used is sketched diagramatically in fig. 36. Two similar Geiger counters were enclosed in a single envelope in such a fashion that each could be individually irradiated. One was exposed to a source of Hg^{203} (as mercuric oxide) and the other to a source of C^{14} (as barium carbonate). The latter, which possesses a long half life, ~6500 years, was intended as a constant control. Actually a very slight variation of counting rate with temperature was observed in the case of C^{14}, which did not show in the decay curve for Hg^{203}. This variation was allowed for by finding experimentally the law relating the C^{14} counting rate to temperature and reducing all C^{14} readings to a standard temperature of 17°C. The apparatus was so designed that the chance of a particle from one source being scattered into the other counter was extremely small. The lead shield was used to keep the gamma-rays of Hg^{203} from affecting the standard C^{14} counter. The plateaux of the two counters are shown in fig. 37.

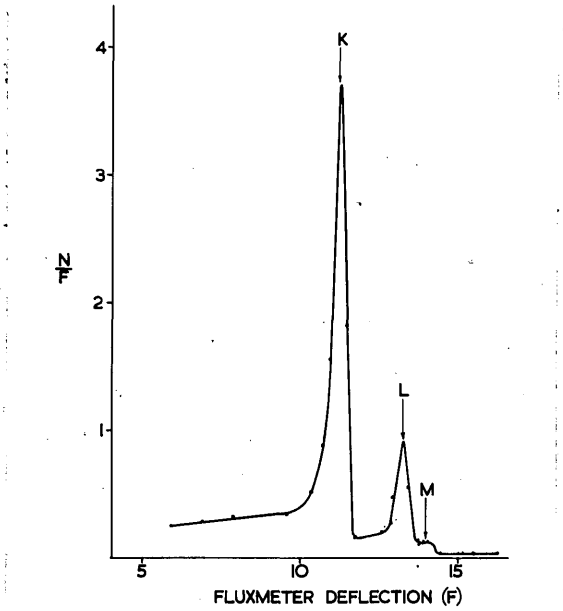


Fig. 35. Photoelectric spectrum of the gamma-ray of Hg^{203}, with 4 mgm/cm^2 platinum radiator.

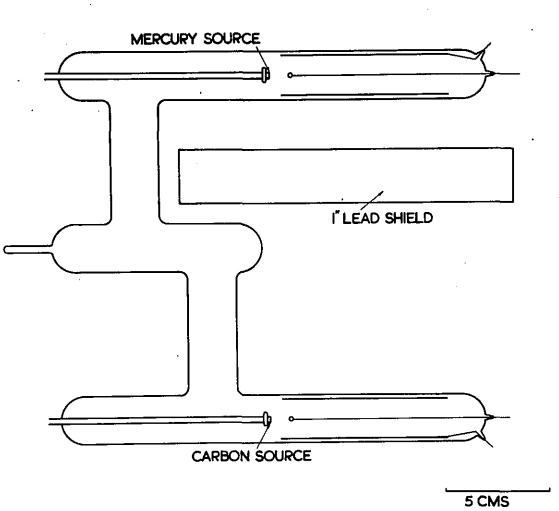


Fig. 36. Controlled Geiger tube apparatus used for half life determination.

The working voltages are indicated by arrows. It was found that the starting and working voltages remained the same throughout the duration of the experiment which gave confidence in the apparatus. Before starting counting, the apparatus was switched on for 15 minutes to allow it to settle down and then the total number of counts from each counter in the same interval of time was recorded. Counting was continued until more than 400,000 counts were obtained from the counter possessing the lower counting rate. In view of the fairly high counting rates, counter dead time corrections had to be made to the observed rates. The experiment was continued for 62.3 days, corresponding to 1.36 half lives. The half life graphs are shown in fig. 38. Graph (a) is the logarithm of the mercury counting rate (after correction for dead time losses) plotted with respect to time as abscissa, while (b) is the logarithm of the ratio of the mercury counting rate to the standard carbon counting rate. (a) gave a value of 45.84 days and (b), a value of 45.95 days for the half life. The final value of the half life was taken as

$$= 45.9 \pm 0.5 \text{ days.}$$

which lies within the range of previously measured values.

11. The Decay Scheme of Hg^{203}.

The decay scheme of Hg^{203} will now be discussed in more detail, with reference to the spin changes involved. It will be well to review the facts which give some indication as to the correct spin assignment.

(a) The ft Value. The value of "f" was calculated from the curves of Feenberg and Trigg (1950) to be 0.6. Combining this value with the half life, t = 46 days, we obtain an ft value of 2.4×10^6, or log ft = 6.38. It is not certain from this whether the beta-transition is allowed or first forbidden although Feenberg and Trigg remark, with regard to fig.10 of

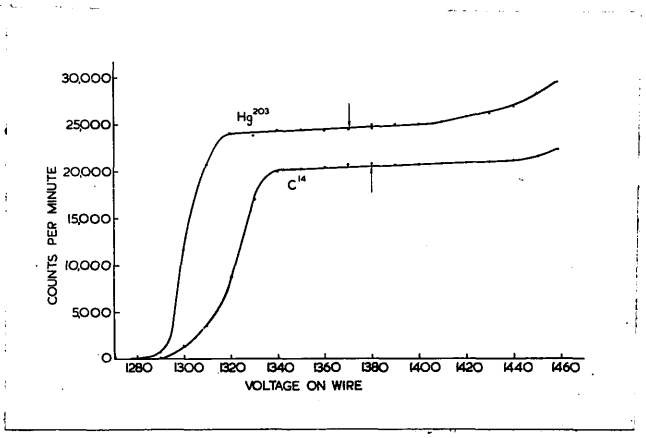


Fig. 37. Plateaux of half life Geiger counters.

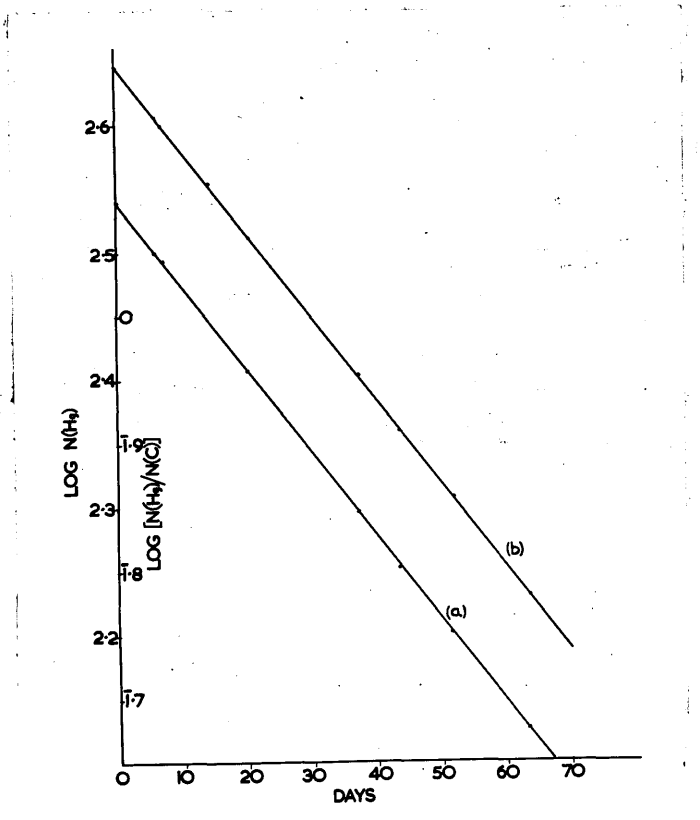


Fig. 38. Graph of half life results.

their paper, that the peak at log ft = 6.2 is "predominantly first forbidden". The log ft value for Hg^{203} lies a little above this value.

(b) Sargent Diagrams. Feather and Richardson (1948) have published Sargent diagrams for the limited region of atomic number, $80 \leq Z \leq 84$. If we fit the data to these diagrams we find that a first forbidden transition with $\Delta I=-1$ and a change of parity is suggested for the 210 KeV beta-transition. Both (a) and (b) would lead one to expect a spectrum having an allowed shape.

(c) Internal Conversion Coefficients. As has been mentioned earlier, the value of $\alpha_K(15.6\%)$ strongly suggests that the gamma-transition is electric quadrupole in type. This corresponds to a spin change of one or two with no parity change.

(d) Half Life of the Excited State. The experiments of Deutsch and Wright (1950) showed that the half life on the 0.28 MeV excited state is $< 0.3 \times 10^{-8}$ sec., and independently Binder (1950) showed it to be $< 2 \times 10^{-8}$ sec. This suggests that the gamma-ray transition is likely to be not more forbidden than electric quadrupole or magnetic dipole. (Moon, fig. 20, 1949).

(e) Ground State to Ground State Transition. According to our experiments, this transition, of energy 490 KeV, is less probable than the 210 KeV transition by a factor $\gtrsim 100$. Also it has never been observed by other workers. According to calculations of Feenberg and Trigg in the paper referred to previously, the energy releases favour the higher energy transition by a factor of about 17. Hence the ft value for this mode of decay must exceed that for the lower energy decay by a factor of $\gtrsim 1700$.

(f) Shell Model. The predictions of the shell model (Haxel, Jensen and Suess, 1950) do not seem sufficiently precise in the region of atomic

number about 80 to assist materially in the assignment of spin values. However it appears reasonable to suppose that Hg^{203} has a spin of 1/2 or 3/2 and it is much less likely to have a still higher spin value. Also it is significant to note that almost all nuclei in the neighbourhood of mercury have spin values in the range $0 \leq I \leq 3/2$. The ground state of Tl^{203} has been measured and has the value 1/2. The spin value of Hg^{201} in the ground state is experimentally found to be 3/2 and it is very reasonable to suppose that the addition of two neutrons to form Hg^{203} would leave the spin unchanged.

In view of the shell model and spin arguments just made, one may tentatively construct the decay scheme of fig. 39(a) . The gamma-ray transition is obviously in line with the experimental facts, (c) and (d). Both beta-transitions are allowed according to Gamow-Teller rules, although experimentally the (-1,no) transition is apparently forbidden relative to the (0,no). This is evidently the weak link in the chain. Also the ft value and Sargent diagram considerations, (a) and (b), suggest that the lower energy transition is first forbidden which also is in disagreement with the decay scheme.

It appears to the writer that the decay scheme represented by fig.39(b) is in better accordance with the facts. A spin value of 5/2 is assigned to Hg^{203} andit is now possible to assign a spin change of -1 with parity change to the observed beta-transition . This brings it into agreement with Feather and Richardson's Sargent diagram and also satisfies the ft value conclusions. The gamma-ray transition is left unchanged. The ground state to ground state beta-transition is now assigned a spin change of -2 with parity change. Although this is also classified by Gamow-Teller rules as first forbidden, there is considerable evidence that transitions

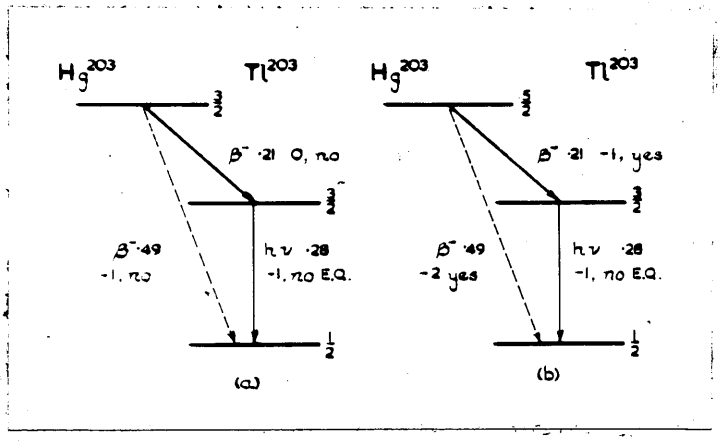


Fig. 39. Possible spin and parity changes in decay of Hg^{203}.

of the type ($\Delta I=2$, yes) have exceptionally large $(E_o^2 - 1)$ ft values, which are about 10^{10} and are remarkably consistent (Shull and Feenberg, 1949, Wu, 1950). Since the ft value of the lower energy transition is 2.4×10^6, this is in agreement with the argument (e) which concludes that the ratio of the ft values has to be $\gtrsim 1700$. On the whole, therefore, the scheme of fig. 39(b) is probably the most satisfactory from the point of view of both experiment and theory. Its main difficulty lies in its inconsistency with the shell model.

An interesting method of classification of beta-emitters due to Dr. K.M. Guggenheimer suggests that the 210 KeV transition is allowed. His calculations indicate that, if the upper energy transition is second forbidden, and the other allowed, the former will be less probable than the latter by a factor ~ 224. These conclusions could be satisfied by a scheme in which the beta-transitions of fig. 39(b) are changed from ($\Delta I=-2$, yes) and ($\Delta I=-1$, yes) to ($\Delta I=-2$, no) and ($\Delta I=-1$, no). This introduces, of course, disagreement with the method of classification of Feather and Richardson.

Much of the uncertainty as to the decay scheme would be removed by the experimental measurement of the spin value of Hg^{203}. This would resolve between fig. 39(a) and fig. 39(b) (and eliminate other less likely schemes which are not considered here) but would not distinguish between fig. 39(b) and its modified form mentioned above. If, also, the ratio of the probabilities of each mode of decay and the spectrum shape of the upper energy transition were measured accurately, assuming it were possible to do so, all doubt as to the correct form of the decay scheme would almost certainly be removed.

12. Conclusion.

It is evident that the use of the integrating method has been very successful and has confirmed the original predictions regarding it. The spectrum of Hg^{203} has been examined and a Fermi plot drawn, from the end-point to 20 KeV, free from the difficulties of superposed conversion electrons in the upper energy region and free from many of the difficulties usually associated with measurements in the low energy region. It has been possible to measure the internal conversion coefficients with what appears to be considerable accuracy. If future work is done on this activity two experimental modifications should be made. Firstly, a source on a thinner backing should be employed (probably nylon film) and, secondly, the gas should be continuously purified to improve the resolution of the instrument, thus making the experimentally obtained spectrum resemble even more closely the spectrum drawn in fig. 25 (apart, of course, from the arbitrary spectrum shape chosen when drawing the latter).

13. Future Use of the Technique.

The lines along which future research on the activity of Hg^{203} should run have been indicated above (Section 11). The purpose of this concluding section is to discuss future possible applications of the integrating technique. In general, it may be said that the method is one which offers considerable advantages in those types of decay in which the beta-rays are followed by photo-electrons which are more energetic than the highest energy beta-rays. As has been seen, the details of the low energy part of the spectrum are readily investigated by examining the spectra carried out to higher energies by the photo-electrons. Also internal conversion coefficients can be accurately determined. In the case of decays followed by complex gamma-ray spectra, one may be able to obtain the shape of the beta-

spectrum which, by normal methods, would be swamped by the presence of photo-electron lines, especially with weak sources at low energies when the resolving power is usually low. Work on RaD is being carried out by this technique, together with other experiments using scintillation counters, which it is hoped will clear up the long sought form of the decay scheme. Among other sources which might prove suitable for examination by this technique are Ir^{190}($E_o \sim 91$ KeV, $E_\gamma = 250$ KeV), Nb^{96} ($E_o = 67$ KeV, $E_\gamma = 1.03$ MeV) etc. It is not known with certainty in the case of Ir^{190} and Nb^{96}, however, whether or not the gamma-transition follows the beta-transition, or, if it does, whether it is delayed. If the gamma-rays are not appreciably converted, the scintillation counter, which has a much higher efficiency for the capture of gamma-rays, may be used in the same way as the proportional counter. The obvious method is to split a crystal into halves and to insert the source between the portions which would then be pressed together again. Such an arrangement constitutes a solid proportional counter identical in principle with the gas-filled proportional counter, and possessing a solid angle of 4π. If the efficiency of capture of the gamma-radiation was $\sim$100% a single spectrum would be observed which would be displaced along the energy axis by an amount equal to the energy of the gamma-ray. If the efficiency were less than 100% for the gamma-ray, but $\sim$100% for the X-rays of the daughter isotope, two groups would be observed corresponding to the primary spectrum and the spectrum displaced by the energy of the gamma-ray respectively. This method of employing the scintillation spectrometer was first applied to the radiations of Hg^{203} itself (Bannerman, Lewis and Curran, 1951). The source, which was that used in the proportional counter experiments, was clamped between two sodium iodide (thallium activated) crystals. The double crystal was

large enough to capture almost all the Tl X-rays but not all the unconverted gamma-rays. Thus group I, corresponding to transitions in which unconverted gamma-rays escaped, remained while group II was displaced to the right and coincided with group III. Fig.10 of their paper shows the results obtained. If a larger crystal were employed, one which would capture ~100% of the gamma-rays, group I would also coincide with group III thus yielding a single spectrum identical in shape to the primary spectrum but starting at ~278 KeV and having an end-point at ~488 KeV. Bannerman et al. point out that in the case of gamma-rays of high energy, possibly up to several MeV, liquid phosphors offer considerable promise, since the problem of making larger crystals of good optical quality is not an easy one.

This method also promised to provide a powerful tool to investigate the beta-spectrum of RaD. The hardest gamma-ray emitted by this source is 46.7 KeV. If a crystal big enough to stop all 46.7 KeV and softer electromagnetic radiations were employed one might expect to find the primary beta-spectrum free from conversion lines, displaced along the energy axis by 46.7 KeV, provided only one beta-transition was involved. The main disadvantage is the low resolving power which the crystal possesses in the region of the beta-spectrum energies (0- ~18 KeV). This study was undertaken and has recently been published (Bannerman and Curran, 1952). The spectrum was examined (a) by measuring the beta-rays in coincidence with higher energy gamma-rays and (b), by the integrating technique. The first method gave a spectrum, probably simple, with an end-point, corrected for resolution, of 18 KeV. The second method did not give the simple primary spectrum expected and it is surmised that one of the excited levels of the daughter nucleus is metastable, having a half life greater than 1 μsec.,

the resolving time of the circuit.

It should be emphasised at this point that, when using a scintillation spectrometer big enough to stop most of the gamma radiation, it is not essential that the gamma-ray energy should exceed the maximum beta-ray energy. This was necessary before in view of the fact that the two or three spectra obtain would overlap and cause confusion. However in the case of the large crystal, only one spectrum is obtained. Hence in addition to the sources mentioned previously as probably being suitable for investigation by the integrated spectrum technique one might add such sources as Na^{24}, Sc^{48}, Nb^{92}, Rh^{105}, Au^{198}, Au^{199} and others.

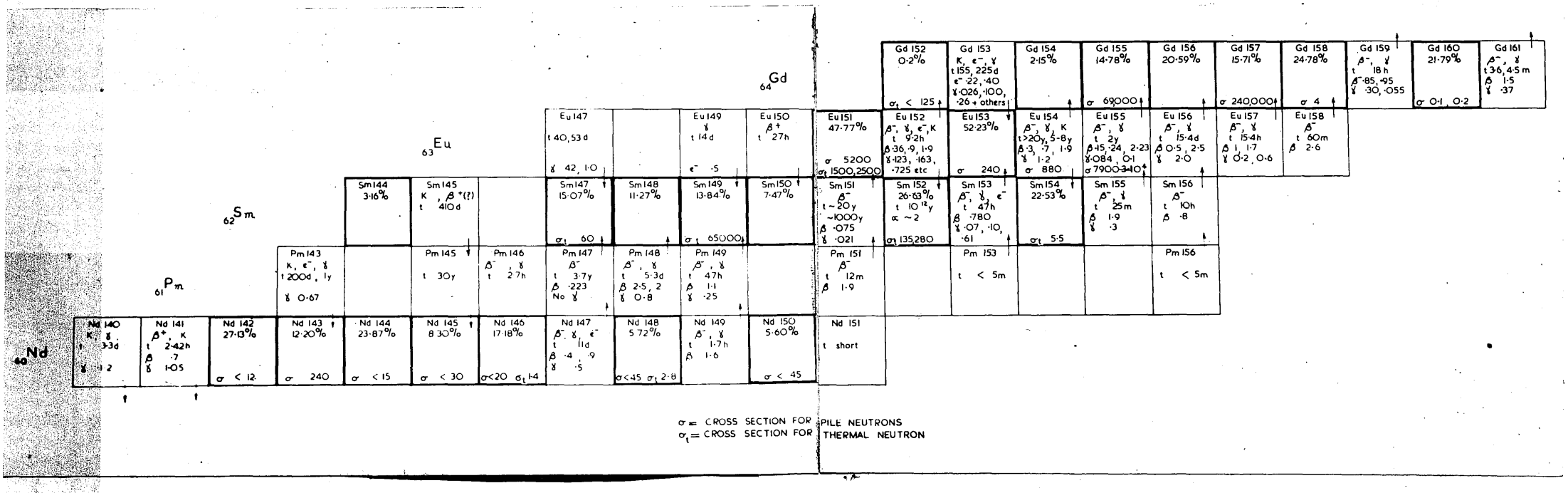


Fig. 40. Isotope chart in the region of samarium and europium. (More recent research indicates that the natural alpha activity of samarium should be identified with Sm^{147}).

PART 4. THE SAMARIUM 151 AND EUROPIUM 155 ACTIVITIES.

1. Introduction.

When the experiments about to be described were undertaken, it appeared that the proportional counter technique might be well suited to the examination of thelong life, low activity source, Sm^{151}. It had been shown, by absorption in aluminium, to have a limiting energy of ~60-65 KeV (Parker, Lantz, Ruch and Hebert, 1948, Parker and Lantz, 1948). Using a magnetic spectrometer, Ketelle and Parker (1949), obtained an end-point energy of 79 KeV, but due to counter window and source thickness difficulties, were unable to obtain any information as to the spectrum shape. It seemed desirable to obtain the spectrum shape of this source and as the energy and the specific activity obtainable were low it appeared, as mentioned above, that the proportional counter method was that best suited to carry out the investigation. Ketelle and Parker used a source obtained from pile products. Such a source was not available to the writer, but it was hoped that by neutron bombardment of pure samarium in the pile at Harwell, sufficient Sm^{151} would be formed to make the analysis possible. There were, however, other activities which could be formed by neutron bombardment but it was hoped, in view of experimental results of other workers, that Sm^{151} might predominate; for example, Inghram, Hayden and Hess (1947) were able to detect and measure the half life of Sm^{151} in a sample of samarium exposed to long pile irradiation. (It should be mentioned, however, that it is not quite clear from their letter whether the half life quoted was in fact their own measurement.)

To give a clearer idea of the activities with which one might be concerned , fig.40 has been prepared. This shows the isotopes in the region of samarium in the form of a chart. The boxes which are heavily

outlined indicate naturally occurring isotopes. The other isotopes are artificially radioactive. The presumed mode of beta-decay is indicated by arrows. The chart is brought up to date in the light of information published up to the summer of 1951, and it must therefore be realised that it does not show the state of affairs at the commencement of this study. Now, from the chart it will be seen that the following activities might be formed by (n, γ) reactions:-

(i) Sm^{145}. Since Inghram, Hess, Hayden and Parker (1947) were unable to detect this isotope in studies of radioactive samarium, it was hoped that this isotope would not interfere.

(ii) Sm^{151}. Already mentioned.

(iii) Sm^{153}. This is short-lived (47 hours) and would not interfere with the other activities, provided the short-lived activities were allowed to die before any work was carried out on the source.

(iv) Sm^{155}. This is also short-lived ($\sim$25 minutes) but it decays into Eu^{155} which also is radioactive and has a half life of $\sim$2 years.

In addition second order activities might take place as follows:-

(v) Eu^{154}. Since Eu^{153} is formed in considerable quantity in the pile and since Eu^{153}, to which Sm^{153} decays, has a high neutron capture cross-section, Eu^{154} might well be formed with an appreciable activity.

(vi) Sm^{156}. This could be formed by neutron bombardment of Sm^{155}. However since Sm^{156} decays with half life of 10 hours, and the product nucleus, Eu^{156}, also radioactive, decays with a half life of 15.4 days, this activity would not introduce complications.

(vii) Eu^{156}. This might arise from neutron bombardment of Eu^{155}. However, as mentioned above, Eu^{156} decays with a half life of 15.4 days, and again no difficulty would be caused by such a reaction.

It appeared, therefore, that Sm^{145}, Sm^{151}, Eu^{155} and Eu^{154} could be present. As has already been mentioned, Sm^{145} was not expected to cause trouble. However Eu^{155} might well have been formed with an activity comparable, with, or much exceeding, that of Sm^{151}. Since the slow neutron cross-section of Sm^{150} was not known (and still is not) it was not possible to calculate the relative activities expected. It was hoped, however, that either Sm^{151} or Eu^{155} would predominate. Very little was known about Eu^{155} at the time and it was evident that it also would well repay investigation. Finally the activity to be expected due to Eu^{154}, relative to that of Eu^{155}, was calculated. This could be done only approximately since the slow neutron cross-section of Sm^{152} was given variously as 135 and 280, while the cross-section of Eu^{153} was quoted at values ranging from 240 to 1000. The cross-section of Sm^{154} was taken as 5.5. Even assuming the most favourable conditions for the production of Eu^{154}, i,e, assuming the highest cross-section quoted, it was calculated that the ratio of the activity of Eu^{155} to that of Eu^{154} at the time of leaving the pile would be $\sim$935;1 and, after six months, $\sim$ 616:1. Thus Eu^{154} was not expected to introduce any complication, provided the original sample of samarium to be irradiated was sufficiently free from europium. The purest obtainable samarium oxide, provided by Nu-metals, Ltd., was therefore used. 0.050 gm of this was irradiated for eight weeks in the pile at Harwell at a neutron flux of 3.5×10^{11} per cm^2 per second.

To anticipate some of the results of the experiments to be described, it was found that the Sm^{151} and Eu^{155} activities predominated and that the ratio of the intensities was $\sim$1:3. The amount of Eu^{155} present was higher than was expected but, because of the different energies involved in this decay, it did not complicate matters very much. Since the intensities of

Sm^{151} and Eu^{155} were of the same order, it did not prove possible to examine the spectrum shape of either activity in detail. However the various experiments carried out, examination of the beta- and gamma-ray spectra by proportional counter and $\beta-\gamma$, $\gamma-\gamma$ and $\gamma-e^-$ coincidence absorption studies, led to considerable elucidation of the decay schemes of Sm^{151} and Eu^{155}. No attempt to construct a decay scheme of either had been made previously. Other information concerning the intensity of Sm^{145} formed in the pile and of the end point energies of the beta-spectra associated with the decay of Eu^{154}, was also obtained.

If the chemical separation of the europium and samarium had been carried out, it would have been possible to examine Sm^{151} and Eu^{155} separately. However in view of the great difficulty involved in carrying out such a separation and since, as far as could be ascertained, no one in the University of Glasgow possessed experience of rare earth separation techniques, it was not considered advisable to attempt it.

2. Brief Review of Other Investigations.

The results of other investigations on these activities will now be briefly mentioned. No distinction will be made between results published before the commencement of this study and results obtained subsequently.

Sm^{145}: This has been reported in earlier work to have a half life greater than 150 days, and to possess gamma-rays of energy 242 and 950 KeV. However recent work of Butement (1951) showed that there were no hard gamma-rays and that K X-rays characteristic of promethium (Pm) were present, thus indicating K capture as the principal mode of decay. He determined the half life to be 410 days.

Sm^{151}: The earlier work has already been described. Recently Agnew (1950), using a double magnetic lens spectrometer, obtained a beta-spectrum which

gave a linear Fermi plot indicating an end point of 75.5 KeV. Marinsky (1949), in work as yet unpublished, obtained an upper energy limit of 74 KeV. Both Agnew and Marinsky used Sm^{151} prepared from fission products. Parker et al. (1948) and Marinsky could detect no gamma-rays. However Scharff-Goldhaber, der Mateosian, McKeown and Sunyar, using an argon-filled proportional counter, detected gamma-rays of energy ~21 KeV, and also L X-rays (Scharff-Goldhaber et al., 1950). They used Sm^{151} separated from fission products by Marinsky and Glendenin. The half life value is not known with any certainty. Inghram, Hayden and Hess give a value of ~ 20 years, while private communications of Inghram and Marinsky to Ketelle and Parker (1949) give ~200 years and ~1000 ± 350 years, respectively.

Eu^{155}: Seaborg and Perlman (1948) suggest that there are two groups of beta-rays, with end points at 180 and 230 KeV, as determined by absorption methods. The Trilinear Chart (Sullivan, 1949) gives the end points as 160 and 240 KeV but does not give the source of information. Marinsky states that there are two beta-transitions. One, which occurs with 80% probability, has a maximum energy of 154 KeV, measured by spectrometer, while the other transition, occuring with 20% probability, has a maximum energy of 243 KeV. No experimental errors are quoted. With regard to gamma-radiation, Winsberg (1950) detected a gamma-ray of energy 84.4 KeV, as measured by the critical absorption technique. The Trilinear Chart gives three gamma-rays of energies, 84 KeV, 102 KeV and 1.2 MeV (weak). Marinsky, using a magnetic spectrometer, finds only two, of energies 85 KeV and 99 KeV, occuring with ~60% and ~40% probability respectively. It would appear that the 84, 84.4 and 85 KeV gamma-rays are identical as are the 99 and 102 KeV gamma-rays. Marinsky states that only the 154 KeV

spectrum is in coincidence with the gamma-radiation, that there are no gamma-gamma coincidences and that there are no delayed beta-gamma coincidences. The half life is given as 1.7 years by Hayden, Reynolds and Inghram (1949) and as 2 years by Winsberg.

Eu^{154}: According to Hayden et al. and Marinsky, this activity probably possesses three beta-transitions, measured by absorption, of 300 KeV, 700 KeV and 1.9 MeV with intensities in the ratios 5:4:1. A gamma-ray of 1.2 MeV (measured by absorption) is also reported. Marinsky states that the electrons giving rise to the 300 KeV and 700 KeV spectra are probably in coincidence with this gamma-ray. Hayden et al. state that if K capture takes place, it does so with an intensity relative to beta-emission of less than 0.05.

In addition to these radiations, other radiations have been observed and are given here although it is not known at present whether they belong to Eu^{154} or Eu^{152}, both of which are formed by neutron capture in europium and which have almost equal half lives. Certain gamma-rays have been observed in our studies which seem to correspond somewhat to these given below. This might suggest that these radiations belong to Eu^{154} rather than Eu^{152}. However there is reason to believe that a very small amount of europium may have been present in the original sample of samarium in sufficient quantity to make it doubtful. These radiations are as follows:- Beta-spectra possessing end points of 750 KeV and 1.57 MeV have been found by Shull (1948) and Cork, Schreffler and Fowler (1948), using magnetic spectrometers. In addition these workers have reported gamma-rays of the following energies: 0.040, 0.122, 0.247, 0.286, 0.343, 0.410, 0.442, 0.772, 0.959, 1.082 and 1.402 MeV, measured by spectrometer, while Cork et al. and Krisberg, Pool and Hibdon (1948) also reported gamma-rays of

energies 1.23 and 0.9 MeV by absorption. Finally, Hayden et al. report a half life of 5.3 years for Eu^{154}.

3. Preliminary Experiments.

The first experiments were carried out in an attempt to elucidate the main activity present in the source. The source proved to be more complex than had been hoped and it was evident that a number of different activities must have been present. Several separate absorption studies were made on the beta-radiations using mainly a thin window (1.7 mgm/cm^2) Geiger counter and aluminium absorbers. Then an attempt to measure the energy of the gamma-rays by absorption was made. This was done by deflecting the electrons away from the counter window by use of a strong magnetic field. However it was not possible to draw any conclusions from the gamma-ray absorption curves except that a considerable number of gamma-radiations of different energies were present. Fig. 41 shows a typical beta absorption curve in which the intensity scale is drawn both linearly (A) and logarithmically (B). From (A) it is evident that most of the particles were less than 250 KeV, and were therefore probably due to Eu^{155} and Sm^{151}. Fairly well defined end points on curve (B) can be observed at $\sim$ 75 KeV and $\sim$250 KeV. These values agree with the quoted upper energy limits of the Sm^{151} and Eu^{155} beta-spectra.

Since it was evident that the original hope to have a source consisting mainly of Sm^{151}, or Eu^{155}, was not realised, an examination of the beta-radiations using a magnetic spectrometer was attempted. Even using thick sources, it was not found possible to obtain a sufficiently high counting rate to examine the spectrum with any accuracy. From this experiment it appeared that, although the spectrum had not finished by 400 KeV, the highest energy examined, most of the particles had energies less than 300 KeV.

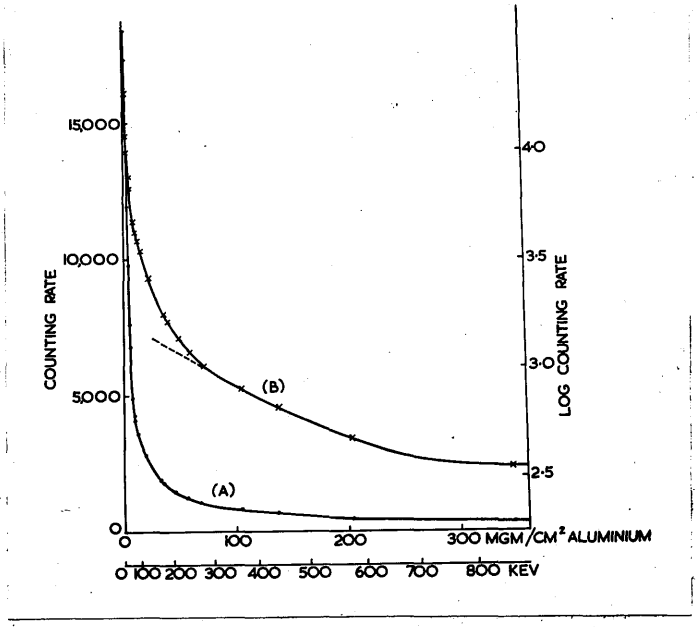


Fig. 41. Absorption of beta-rays in aluminium.

An attempt was then made to examine the gamma-radiations using a xenon-filled proportional counter which has a relatively high gamma-ray detection efficiency, compared to an argon-filled counter. It has the disadvantage that, since the Auger effect in xenon is not complete, a single gamma-ray gives rise to a number of electron peaks instead of only one, as with argon. Since it was not possible to resolve the separate peaks in this experiment it was evident that the number of gamma-rays present was too large to be isolated by this method. It was therefore decided to use an argon-filled counter to investigate the soft gamma-ray spectrum.

4. Gamma Radiation.

A proportional tube, diameter 5.5" and active length 8", filled with methane and argon to partial pressures of 15 cm and 60 cm of mercury, was used to examine the soft gamma-rays emitted by the source. The source holder consisted of a perspex block in a groove in which the radioactive samarium oxide was placed. The oxide was covered with $\frac{1}{2}$ mil aluminium foil. This was placed on the centre window (2 mil aluminium) of the counter. The resulting pulse distribution was analysed both by the single channel kicksorter method and by photographic recording. The results by each method agreed though those observed by the latter method were more precise and the histograms opposite and on succeeding pages were obtained by this method.

Fig.42 shows the existence of two gamma-rays of energies 15.2 KeV and 19.3 KeV. The peak at about 8 KeV is due to the excitation at the counter wall of the K X-rays of copper by the gamma-rays of the source as was proved by a separate experiment. The K X-radiation of gallium, due to K-capture in Ge^{71}, was used for calibration and the resulting histogram is shown in the inset figure. The two peaks at 15.2 and 19.3 KeV do not

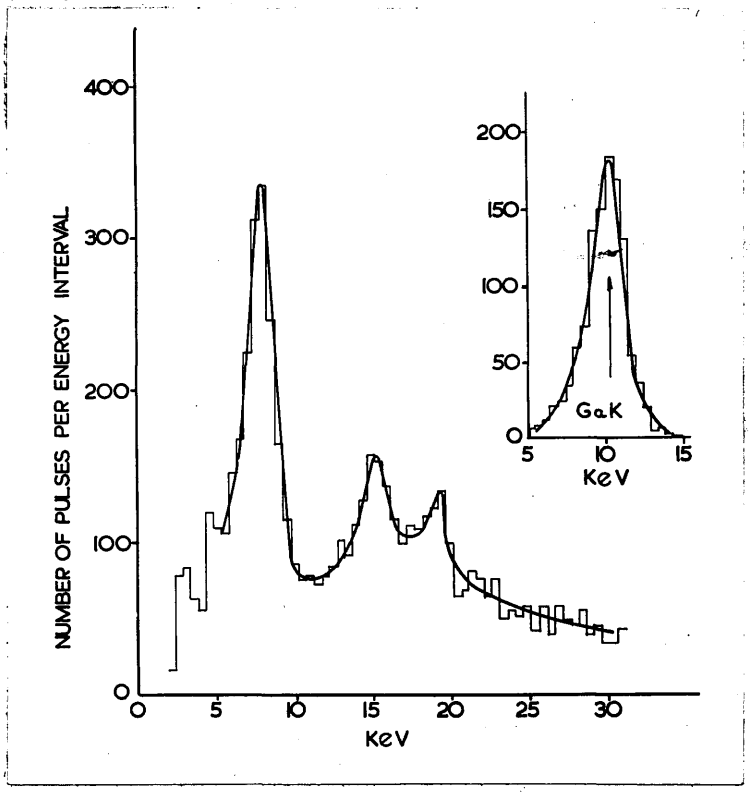


Fig. 42. Low energy gamma-ray spectrum.

correspond to any X-rays in the region of the atomic number of samarium and are therefore true nuclear gamma-rays. As has been mentioned previously, Scharff-Goldhaber et al. by the proportional counter method detected gamma-rays of energy 21 KeV emitted by Sm^{151}. The gamma-ray here estimated to have an energy of 19.3 KeV is presumably of the same origin. The source which they used was a fission product and was therefore expected to be relatively pure samarium. The fact that these workers did not detect a gamma-ray of energy 15.2 KeV, although of comparable intensity, suggests strongly that it is associated with the decay of Eu^{155}. (Butement found that X-rays, but no gamma-rays, are associated with the decay of Sm^{145} and the other possible activities are much too weak to give rise to a gamma-ray of such intensity). By measuring the area under the two peaks and making allowance for the X-ray capture efficiency of the counter it is estimated that the ratio of the intensity of the 15.2 KeV to that of the 19.3 KeV radiation is $\sim 0.7:1$.

Marinsky and others have found two gamma-radiations of Eu^{155} of energy 85 and 99 KeV. It was natural to relate the 15.2 KeV transition to the difference between 85 and 99 KeV, so that the 85 and 15.2 KeV gamma-rays would be emitted in cascade, as an alternative mode of decay to the 99 KeV transition. The 85 and 99 KeV transitions were measured by means of internal conversion electrons in a magnetic spectrometer and thus the very low energy radiation found here (15.2 KeV) would presumably escape detection.

To confirm that the 8 KeV peak of fig. 42 was due to copper K X-rays, an examination of the low energy gamma-spectrum was made using a glass walled, aluminium cathode counter having no copper or brass components at all. The spectrum obtained is shown in fig. 43. Calibration was effected by the use of copper X-rays. (inset figure). No peak corresponding to the

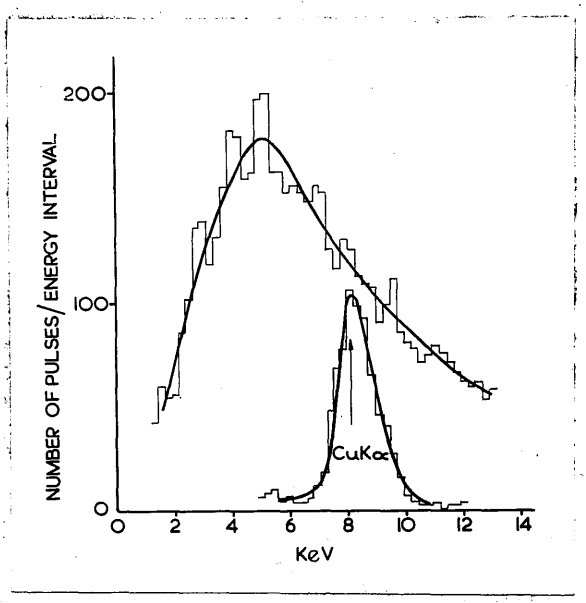


Fig. 43. Gamma-ray spectrum below 10 KeV.

copper K X-ray energy was found but the figure shows a broad peak about 5 KeV which can be ascribed to the L X-rays emitted by the source due to K- and L-capture and to internal conversion. The intensity of this radiation was evidently considerable since there must have been a large amount of absorption in the aluminium window and source cover.

The higher part of the gamma-ray spectrum is shown in fig. 44. There is an intense peak centred at ~40 KeV. The calibration peak on the same figure is due to the K_α and K_β flourescence X-radiations of samarium excited by allowing X-rays to fall on a thin layer of samarium oxide. The position and width of the source peak suggests that X-rays of energy higher and lower than the K_α X-rays of samarium are emitted by the radioactive material. It was thought that the peak was due mainly to K-capture in Sm^{145} and internal conversion of the gamma-rays of Eu^{155}, giving rise to the X-rays of promethium and gadolinium respectively. It would appear that the activity of Sm^{145} was roughly of the same intensity as that of Sm^{151} and Eu^{155}.

It is impossible by the proportional counter technique to resolve X-rays peaks as close in energy as the K X-rays in this region, so an attempt was made to resolve them using a motor driven X-ray crystal spectrometer which was borrowed from the Chemistry Department of the University. X-ray film was used as a detector and a calcite crystal for reflection of the X-rays. A preliminary run of ~24 hours, using all the available source, seemed to give a very slight indication of a first order reflection but on repeating the experiment no darkening could be observed indicating that the first result was probably due to a defect in the film. As it was necessary to go out to a much higher order to obtain adequate resolving power, and as the intensity falls off rapidly with increasing

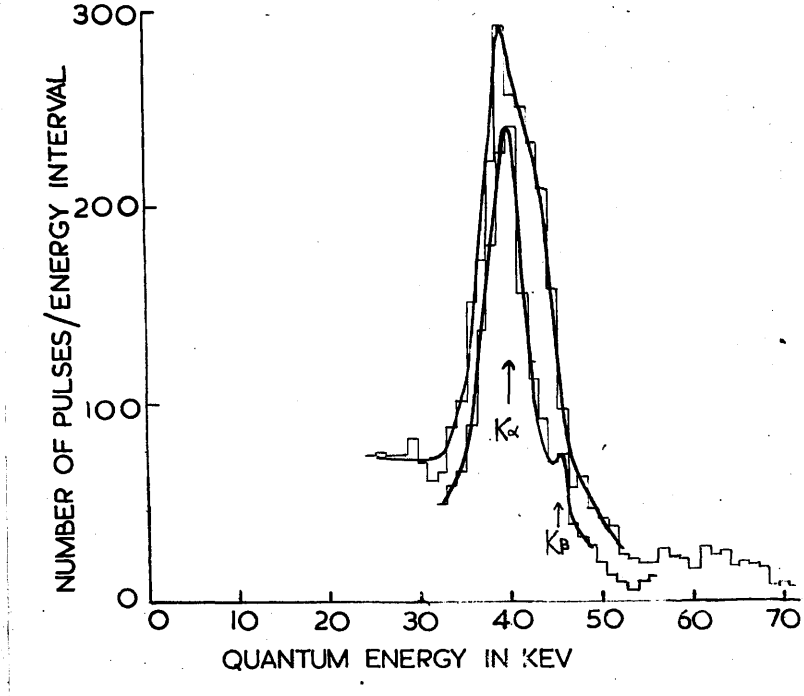


Fig. 44. Gamma-ray spectrum near 40 KeV.

order, it was evident that this method was not sufficiently sensitive. It is possible that the use of a curved crystal spectrometer might have proved successful but none was available. It might also have been possible to gain some idea as to the structure of the X-ray peak by the use of the critical absorption technique. However, in view of the fact that X-rays of this energy are to be expected in any case, the attempt to resolve the X-rays was not carried further.

Mr. R.C. Bannerman of this department carried out an investigation of the higher energy gamma-rays emitted by the source, using a scintillation spectrometer. The intensity of such rays was rather low so that it was not possible to use the double crystal technique as would have been desirable in view of the complexity of the gamma-ray spectrum. Nevertheless he obtained the following, somewhat tentative, values of energy and intensity of gamma-rays:-

Energy(MeV)	1.31	1.13	0.90	0.64	0.44	0.35	0.26	≈0.1
Intensity	1	1	1	0.3	weak	4	1.5	20(?)

By referring to the gamma-ray energies ascribed to $Eu^{152,154}$, mentioned earlier, it is evident these gamma-rays could have been due to the same activity or activities.

If Sm^{145} were to decay partly or wholly by positron emission, one would expect to observe annihilation gamma-radiation. In view of this and of earlier absorption and scintillation spectrometer results, which suggested the existence of gamma-rays of ≈500 KeV, an experiment to observe possible coincidences between the gamma-rays was carried out. The source was completely surrounded by 1/16th inch lead and the coincidence rate with two scintillation detectors noted (a) with an angle of 90°, and (b) with an angle of 180°, between the directions of the two detectors. If there had been present an appreciable intensity of annihilation radiation, one would

have expected an increase in the counting rate in the second case. Actually, using 1" crystals each at a distance of approximately 3", the coincidence rate remained constant within experimental error. The rates were (a) 13.5 ± 2.1. counts per minute and (b) 13.8 ± 2.1. counts per minute. These rates are "true" coincidence rates, i.e. the "random" coincidence rate has been deducted. These results show that Sm^{145}, if present in quantity (as was believed to be the case), decays almost entirely by K-capture.

5. Beta Spectra.

The beta-rays emitted by the source were examined using an end-corrected proportional counter of 8" operating length and 5.5" diameter placed in a magnetic field of 1400 gauss, parallel to the wire. The source was prepared as follows: 1 mgm of samarium oxide was dissolved in concentrated nitric acid, evaporated over a water bath to dryness, the residue dissolved in water and evaporated to dryness again. The samarium nitrate thus obtained, now free from acid, was dissolved in two drops of 10% insulin solution. This was transferred to a nylon film supported on a wire loop of diameter 0.35 cm. A thin layer of "Aquadag" was deposited on the other side of the film to render it conducting, though, as was mentioned in Part 1, this was probably not necessary. The source and holder were then attached to a thin rod, held parallel to the wire, so that the source was situated half-way along the length of the counter. The counter was filled with methane to a pressure of 10 cm and argon to a total pressure of 50 lbs/in^2.

Fig. 45 is representative of the spectrum analysis achieved using the single channel kick-sorter. The upper energy end of the spectrum is shown multiplied by a factor of ten for the sake of clarity. Discontinuities on the spectrum, indicated by arrows, appear at (i) 70-80 KeV, (ii) ~150

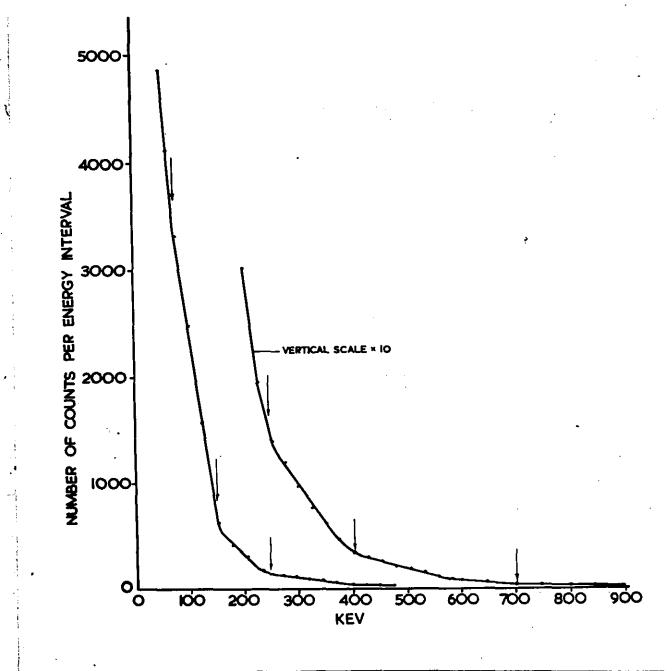


Fig. 45. Beta-spectrum obtained using proportional tube spectrometer.

KeV, (iii) ~250 KeV, (iv) ~400 KeV, (v) ~700 KeV. These various end-points are associated with Sm^{151} (70-80 KeV), Eu^{155} (150 and 250 KeV) and Eu^{154} (400 and 700 KeV). As has been mentioned earlier, previous work by the absorption technique indicated that Eu^{154} emitted beta-rays having spectrum end-points of 0.3 and 0.7 MeV.

Separate examination of the low energy region of the beta-spectra was carried out and the spectrum shape obtained is shown in fig. 46. The shape was considerably affected by the superposition of a number of peaks due to conversion electrons and X-rays. These peaks can be satisfactorily explained in terms of the conversion electrons of the various gamma-radiations known to be present, bearing in mind the integrating properties of the proportional counter described in part 3.

From the various spectra observed, and assuming an approximately allowed form for the beta-spectra, it was estimated that the ratio of the intensity of Eu^{155} to the intensity of Sm^{151} was approximately 3:1. Also the intensity of Eu^{154} is much greater than expected. The ratio of the intensity of Eu^{154} to that of Eu^{155} is roughly 1:5 which is much greater than the maximum calculated ratio (~1:600). This may be explained by the presence of a very small amount of europium in the original spectroscopically pure sample of samarium oxide (the (n,γ) cross-section for the production of Eu^{154} is very high). ~~or else reactions other than (n,γ) may have taken place to an appreciable extent, e.g. $Sm^{154}(n,p)Eu^{154}$.~~

6. Coincidence Studies.

In view of the complexity of the beta- and gamma-spectra indicated by the proportional counter and by other experiments it was decided to invoke the aid of the coincidence absorption technique which has been

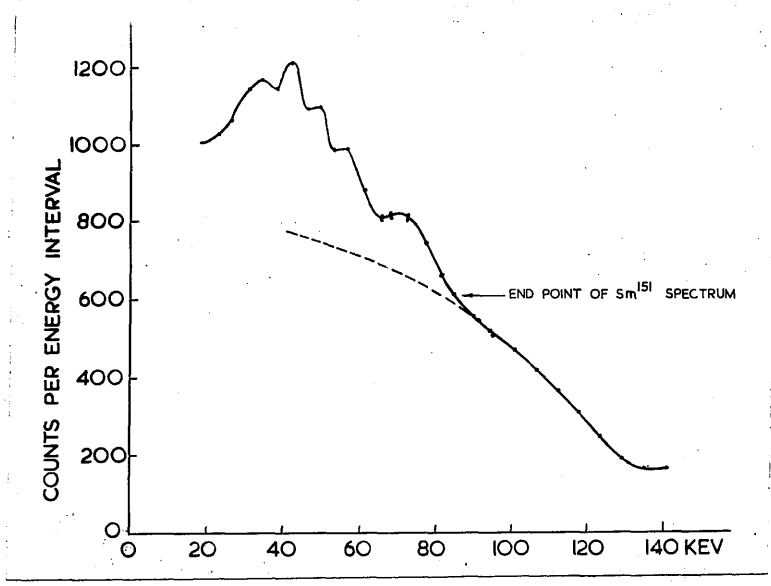


Fig. 46. Low energy region of beta-spectrum.

described in some detail in part 1. As a result of the experiments performed up to this point it was believed that the decay schemes associated with the decay of (a) Eu^{155} and (b) Sm^{151} were shown in fig. 47. In fig. 47 (A) the 15 KeV gamma-transition has been shown preceeding the 85 KeV transition. No clue is provided by the present experiments which allows any decision to be made regarding the correct sequence of these gamma-rays.

The first set of experiments to test the decay schemes was to investigate the way in which the number of beta-gamma coincidences relative to the total beta-counting rate varied as absorbers of increasing thickness were inserted in front of the beta-counter. In the initial experiment a thin window (1.7 mgm/cm^2) G.M. counter was used to detect the beta-rays while a lead-walled counter was used to detect the gamma-rays. (This counter was the one used in the search for the possible emission of annihilation gamma-rays in the decay of long-lived nickel, as described in part 2.) Due to the low counting rate it was difficult to attain good statistical accuracy, and the gamma counter was replaced by a scintillation detector to improve the efficiency for detecting gamma-radiation. This detector was shielded by 5/64" of copper to stop electrons and soft X-rays. It is believed that this may have been the first time that a scintillation counter and a Geiger counter had been used in coincidence. It was found fairly simple to operate them in this way and the set-up had a resolving time of $\sim 0.7 \mu$sec. This was measured by irradiating the two counters separately, giving rise to counting rates of N_1 and N_2 respectively, and measuring the random coincidence rate, N_r, obtained. The resolving time, τ, is given by the simple formula

$$\tau = N_r / 2 N_1 N_2$$

The results obtained using the two techniques were similar, except

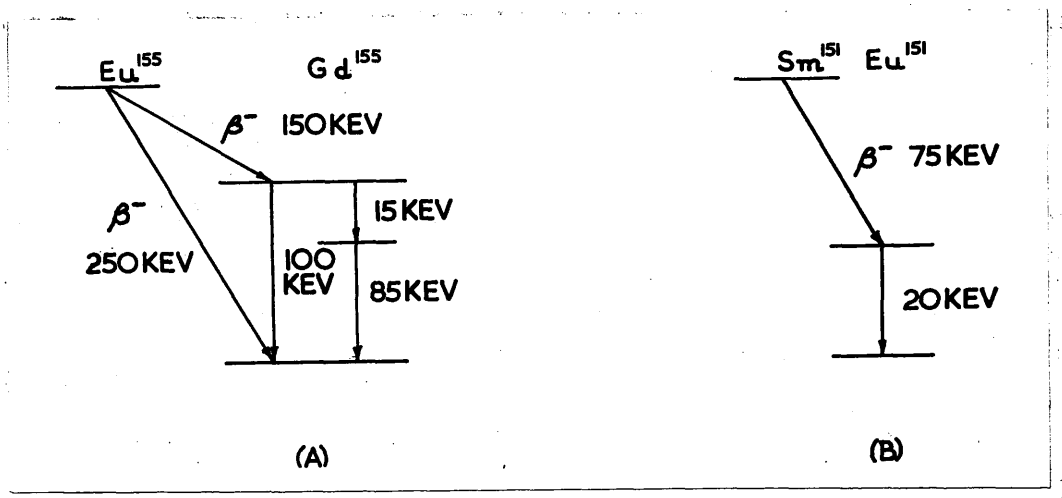


Fig. 47. Proposed decay schemes of Eu^{155} and Sm^{151}.

that smaller errors are associated with the second method and a typical set of results using the scintillation detector is shown in fig. 48, in which the beta-gamma coincidences per beta-particle are plotted in terms of the thickness of aluminium between the source and the beta-counter. The experimental arrangement used is shown diagrammatically in an inset figure. The coincidence absorption curve receives a ready interpretation in terms of the decay schemes of fig.47. The first steep rise of the curve to ~80 KeV is due to the elimination of the beta-rays of Sm^{151}. Since the 19.3 KeV gamma-ray which follows the decay is not detected by the scintillation counter, the beta-rays of Sm^{151} do not contribute to the coincidence rate but only to the single beta-counting rate. The less steep rise to ~150 KeV is presumably associated with the absorption of the beta-rays of higher energy (~250 KeV) of Eu^{155} since they do not contribute to the coincidence rate. The elimination of coincidences arising from the 150 KeV spectrum of Eu^{155} results in the break in the curve at ~150 KeV, assuming the proposed decay scheme is correct. The gradual falling off to lower values is mainly due to the small amount of Eu^{154} present which gives rise to beta-rays of energy as high as 0.7 MeV.

Some experiments on the variation of the electron-electron ($\beta - e^-$) coincidence rate with increasing absorber thickness were carried out. In the first experiment two thin window G.M. counters were placed opposite each other, windows facing inwards, the absorbers being inserted between the source and one of the counters. However as a large increase in the coincidence counting rate was caused by the particles making a count in one counter and then being reflected by the gas or the wall into the other counter giving another count within the resolving time of the circuit, a second arrangement was adopted in which the counters were situated side

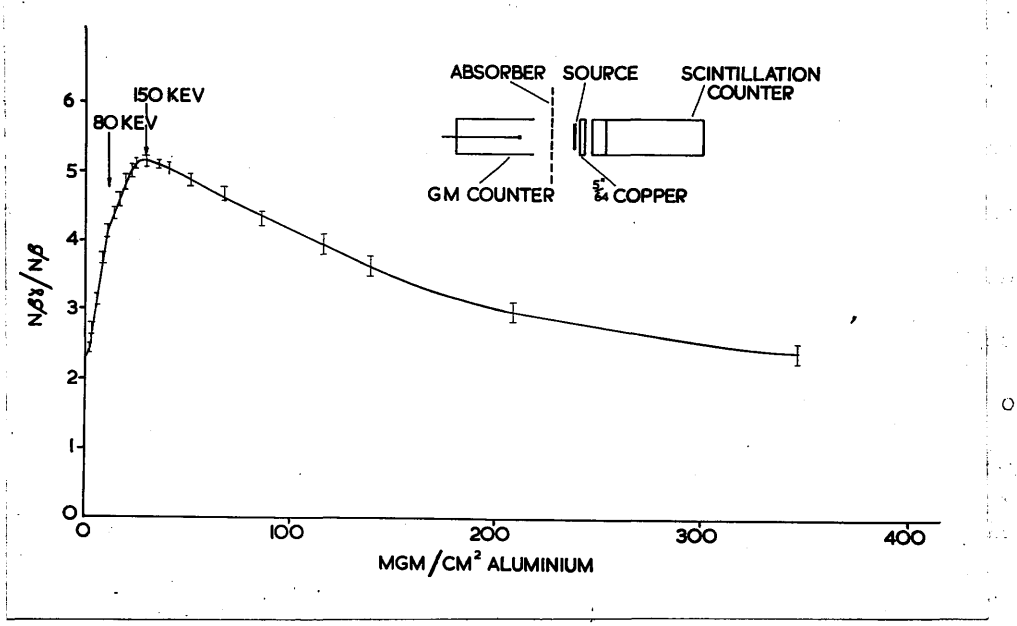


Fig. 48. Ratio of number of beta-gamma coincidences to the number of beta counts as a function of aluminium absorber thickness.

by side with the source about 1" in front of the windows, midway between the counter tube axes. These experiments gave results consistent with the interpretation of the beta-gamma coincidence experiments.

In the work of Marinsky on Eu^{155}, described earlier, he found that this isotope possessed two beta-spectra of upper energy limits 154 and 243 KeV, and gamma-rays of energy 85 and 99 KeV. He found that the lower energy beta-rays were coincident with gamma-radiation but he concluded that there were no gamma-gamma coincidences. The above work leads to the conclusion that the 99 KeV gamma-transition corresponds to the difference between the two beta-spectrum end-points and that the soft gamma-ray detected here (15.2 KeV) corresponds to the difference between the energies of the harder quanta. The writer's measurements of the beta-spectra are consistent with a difference of 99 KeV (see section 5). It is easy to understand Marinsky's failure to find gamma-gamma coincidences with Eu^{155}. The 15 KeV quanta are not detected efficiently by most normal G.M. counters and it is difficult to detect or measure gamma-rays of such low energy with the magnetic spectrometer. Moreover the presence of intense L-radiation and theoretical reasons suggest strongly that the quanta are very highly converted.

In spite of the high conversion of the 15 KeV quanta it was decided to make a direct test of the scheme of fig. 6 (A) by seeking to establish that the beta-rays belonging to the softer spectrum of Eu^{155} were followed by gamma-radiation of energy 15 KeV. A beta-ray counter was covered with sufficient material to stop all the beta-rays of Sm^{151} (75 KeV equivalent thickness). Opposite this counter was placed a special G.M. counter designed to detect 15 KeV quanta efficiently. It contained ethyl alcohol

to a partial pressure of 1.5 cm. of mercury and argon to a total pressure of one atmosphere. In spite of the high pressure employed, the counter operated in a stable manner and possessed a good plateau. This counter was covered with polythene of thickness 270 mgm/cm^2 which was adequate to stop all beta- particles from the source while transmitting quanta of 15 KeV fairly efficiently. Polythene contains only hydrogen and carbon and is therefore much more effieient in this respect than most other materials, even other plastics, which usually contain oxygen in addition to hydrogen and carbon. The source was placed between the two counters and the beta-gamma coincidence rate recorded as a function of the thickness of aluminium placed between the source and the gamma-ray counter. It was hoped that the coincidence rate might, initially, fall exponentially at a rate corresponding to 15 KeV and then, as the absorber thickness increased further, at a rate corresponding to the K X-ray energy of gadolinium, thus showing that the beta-rays of Eu^{155} were in coincidence with 15 KeV quanta (Sm^{151} beta-rays do not interfere since they are completely absorbed). The results are shown in fig.49 and indicate the prominence of two radiations. The lesser slope corresponds to the absorption coefficient in aluminium of quanta of energy 40 KeV which is approximately the energy of gadolinium K X-rays. The initial slope, after the subtraction of the harder radiation, corresponds to an energy of 6 KeV, which is approximately the L X-ray energy. Thus, while the test did not directly confirm the decay scheme of fig. 6 (A), this is the result one would expect if the 15 KeV quanta were strongly internally converted.

When the absorber in front of the beta-ray counter was removed, thus allowing the Sm^{151} beta-rays to enter, (total window thickness was then 1.7 mgm/cm^2), the number of beta-gamma coincidences per beta-particle

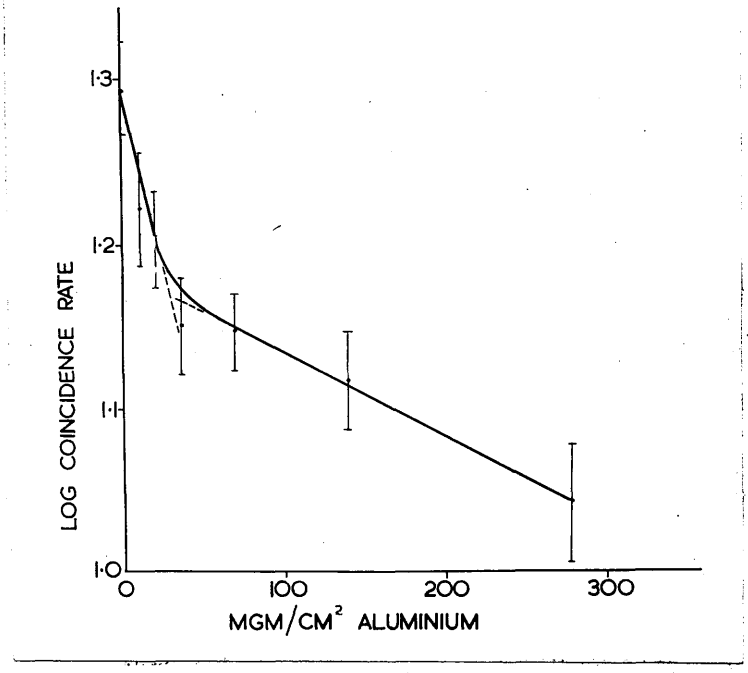


Fig. 49. The logarithm of the number of beta-soft gamma coincidences as a function of gamma absorber thickness.

remained almost constant (1.18×10^{-2} and 1.14×10^{-2} respectively) although the single beta counting rate had increased considerably. From this it is deduced that the Sm^{151} beta-particles are likewise in coincidence with soft gamma quanta (19.3 KeV) or with L X-rays arising from internal conversion. This result affords direct support to the scheme of fig. 6 (B).

At the same time another test was carried out in an endeavour to show that the 15 KeV quanta were in coincidence with 85 KeV quanta. Two sodium iodide crystals were used on opposite sides of the source with sufficient polythene in front of each to eliminate all beta-rays and the gamma-gamma coincidence rate was observed as an increasing thickness of absorber was placed in front of one of the counters. A straight line of slope corresponding to ~40 KeV was obtained, but no initial slope corresponding ~15 KeV or ~6 KeV. Again this is easily understood if the 15 KeV quanta are strongly internally converted, since the discriminator setting of the scintillation counter circuits made the counters insensitive to energies as low as 6 KeV.

In conclusion it may be said that, while it did not prove possible to fulfill the original purpose of the study, i.e. to investigate the spectrum shape of Sm^{151}, much useful information regarding the beta-and gamma-ray spectra of Sm^{151}, Eu^{155} and Eu^{154} was obtained and decay schemes, consistent with the findings, were presented for the first two for the first time. This work was published in the Proceedings of the Physical Society (Wilson and Lewis, 1952).

PART 5. THE NATURAL RADIOACTIVITY OF RUBIDIUM.

1. Introduction.

Since the principal purpose of the writer's researches was to investigate "radiations of low specific activity", it was natural (as has been mentioned in part 1) to turn one's attention to the natural radioactivities in the region of $Z < 80$. These activities are, without exception, very long-lived and hence of very low specific activity. Some of these activities, e.g. K^{40} and Rb^{87}, were discovered to be radioactive by actual observation of the radiations emitted. The activities of others, e.g. In^{113}, In^{115}, Re^{187}, etc., were deliberately searched for since the presence of a neighbouring naturally occurring isobar made it seem likely that one or other of the pair of isobars would prove radioactive.

The experimental results obtained for these activities have up till now been very unreliable indeed, with the exception, in recent years, of data on K^{40}. Using separated isotopes and the scintillation counter technique, it has proved possible to determine the end-point, beta-spectrum shape and decay scheme of K^{40} with some precision. In each of the other cases, if an upper beta-ray energy limit is quoted, this value is fairly low and well within the range of the proportional counter. The proportional counter is well-suited for other reasons which have been dealt with in Part 1, but will be briefly recapitulated here. These reasons are; (a) high solid angle, 2π for sources mounted on the wall, as will normally be the case for these activities; (b) large source area available; (c) since the background is spread over a wide energy range, the ratio of counting rate with the source present to the fraction of the background in the region of the spectrum is greater than would be the case if only the counting rates themselves were observed, as is the case with Geiger-Müller counters, for

example. A programme of work was therefore embarked upon in Glasgow, the purpose of which was to investigate some of these activities using, mainly, the proportional counter technique. A comprehensive investigation of the activity of Rb^{87} was made and will be described in the following pages, together with a brief account of preliminary results on Nd^{150}.

It may be worthwhile at this point to mention the method used by Libby to examine the natural radioactivities (Libby, 1934 a,b, Libby and Lee, 1939, Libby, 1939, etc.). The apparatus used by him is known as the "screen wall counter" and consists of a tube inside which is a coaxial wire mesh cylinder which acts as a Geiger counter cathode. An axial tungsten wire forms the anode. The counting volume is limited to one half of the cylinder length. The source, mounted on a half length cylinder which just fits inside the outer case, can be moved magnetically in and out of the counting volume. Hence the counting rate with and without the source can be determined. Ofcourse this method does not possess the background advantages of the proportional counter method, mentioned above. In an endeavour to estimate the energies of the radiations an electromagnetic field is applied axially and the smallest current necessary to keep the particles from entering the counter through the screen wall is noted. Calibration of current against limiting energy is carried out with known sources. This method of energy estimation has not proved very reliable however, and has tended to give low values as will be shown later. Also Libby sometimes used absorption methods to measure the particle energies. The technique has been used recently to measure the amount of C^{14} occurring in various materials with a view to estimating their ages. It has been necessary to cut down the background counting rate considerably and this has been done by surrounding the counter by an 8" thick steel cylinder and a cylind-

rical array of anticoincidence counters which reduce the background from 450 counts per minute to 4.5 counts per minute (Anderson, Arnold and Libby, 1951).

2. Previous Work on the Radioactivity of Rb^{87}.

This activity was first discovered by Sir J.J. Thomson as long ago as 1905 and has been the subject of many investigations since that time, in spite of which, this present research was the first to yield reliable information concerning the maximum beta-ray energy spectrum shape and the decay scheme. In this rather remarkable work of Thomson (1905), he investigated the emission of electrons from rubidium and from a sodium-potassium alloy, with and without incident light falling on the surface, using a high vacuum electroscope to detect the particles. He found that, even without light shining on these metals, ionizing particles were emitted which he showed to be negatively charged. Hence he concluded that rubidium and potassium or sodium (or both) were radioactive, decaying by beta emission. He showed that lead, silver and mercury gave no such effect. Two members of his research team, Campbell and Wood (1906), further investigated the radioactivities and showed, by absorption, that the radiation of rubidium was heterogeneous and less penetrating than that of potassium which they proved to be the active constituent in Thomson's alloy. They showed that sodium and lithium were not radioactive. More recent work is summarised in the following table (Table 1) which gives the source thickness in mgm/cm^2, the half life, τ, in 10^{10} years, the end-point energy, E_o, in MeV and the gamma-ray energies, $h\nu$, in MeV.

Table.1.

Author	mg/cm^2	τ	E_o	$h\nu$	Remarks
Mühlhoff,1930	1.5 to 3.0	12	1.1	-	Comparison of absorption with RaE
Klemperer 1935	"	-	0.25	-	Examination of Mühlhoff data
Libby and Lee 1939	?	-	0.132	-	Screen-wall counter
Ollano 1941	thick	-	~0.13	0.034 0.053 0.082 0.102 0.129	Gamma rays deduced from conversion photoelectrons
Saha 1946.	300	-	0.144	-	Modified Libby device
Eklund 1946	0.1	5.8	-	-	Counter with internal source
Fazzini and Franchetti 1948	?	-	0.56	-	Cloud chamber
Haxel and Houtermans 1948	~0.1	6.9	~0.01	~0.135	Coincidences in G.M. tubes
Haxel,Houtermans and Kemmerich 1948	~0.1.	6.0	~0.01	~0.135	Tentative allocation of decay β-rays and electrons
Kemmerich 1949	0.03	6.0	-	-	Study of effect of source thickness.

The value of τ is very important in connection with the question of the age of the earth and of mineral deposits and separate studies have been made with the evaluation of τ as a principal object. Hemmendinger and Smythe (1937) showed, by isotope separation, that the activity of rubidium was due to Rb^{87}. Hahn, Strassman and Walling (1937) showed that

the rubidium-containing ore, lepidolite (Canadian mica), contained a considerable quantity of strontium. (1012 gm of lepidolite contained 7.16 gm of Rb^{87} - allowing for the 27.2% abundance of this isotope - and 0.156 gm of strontium). Mattauch (1937) showed that only the mass 87 isotope of strontium was present ($> 99\%$ Sr^{87}) thus inferring that its presence was due to the decay of Rb^{87}. Using the above figures, Strassman and Walling (1938) estimated the half life of Rb^{87} by comparison with the uranium - lead ratio method of estimating the age of minerals and obtained a value of 6.3×10^{10} years. This disagreed with the value obtained by Mühlhoff but this could be partly explained by the fact that Mühlhoff was unable to allow for the abundance of the radioactive isotope. This would have reduced his value of 12×10^{10} years. The present agreement between the geological value of the half life and the values obtained directly by Kemmerich, 6.0×10^{10} years, and in this research, 6.15×10^{10} years, gives one confidence in the use of the Rb-Sr ratio method of estimating the age of minerals, as has been pointed out by Curran (1952) in his review on the age of the earth and of mineral deposits.

Table 1 indicates the great uncertainty and mutual disagreement that existed in the published values of E_o and the number and energy of gamma-rays emitted. It is an interesting fact that, after 45 years, so little should be known of the decay and that such a wide range of values of Eo should be quoted (~10 KeV - ~1.1 MeV). These data will be considered in more detail later when the results obtained in this research are discussed.

From the stand-point of beta-ray theory the transition $Rb^{87} \rightarrow Sr^{87} + \beta^-$ is of special interest. The spins of the ground states of both nuclei are known; the spin of Rb^{87} is 3/2 (Jackson, 1933, Kopfermann, 1933, Millman

and Fox, 1936, Millman and Kusch, 1940) and of Sr^{87} is 9/2 (Heyden and Kopfermann, 1938) giving a spin change $\Delta I=3$ in the ground state to ground state transition. The log ft value calculated by Feingold (1951) on the basis of E_o = 130 KeV and a half life of $6x10^{10}$ years, is 16.5, corresponding to a third forbidden transition. Both Konopinski (1943) and Greuling (1942) point out that the decay cannot be "second forbidden" since the half life would have to be shorter by a factor of $1-2x10^3$, while if "fourth forbidden", the half life would belonger by a factor $\sim 10^4$. According to Gamow-Teller rules a spin change of three corresponds to a second or third forbidden transition, while by Fermi rules, it is third or fourth forbidden. Wu (1950) has pointed out that this, probably unique, case in which the degree of forbiddenness (greater than unity) equals the spin change offers a seaching test of beta-ray theory. This fact, together with what has been mentioned previously, makes the investigation of the radioactivity of Rb^{87} of very great interest indeed.

3. The Beta Spectrum.

The experimental arrangement used was, firstly, an end-corrected proportional tube, diamter 5.5", active length 10", surrounded by a lead shield of thickness 2". Later, an array of Geiger tubes, 2" in diameter, operating in anticoincidence with the proportional counter, was added as shown in figure 50. The first arrangement reduced the background counting rate from ~1200 c.p.m. to ~400 c.p.m., and, in addition, the maximum of the background pulse spectrum moved to higher energies. The addition of the anticoincidence array reduced the background rate still further to ~100 c.p.m The spectrum was analysed both by the photographic and by the single channel "kicksorter" methods. The fluorescence X-rays of silver, used at high gain, were used to calibrate the spectrometer which was filled with methane to a

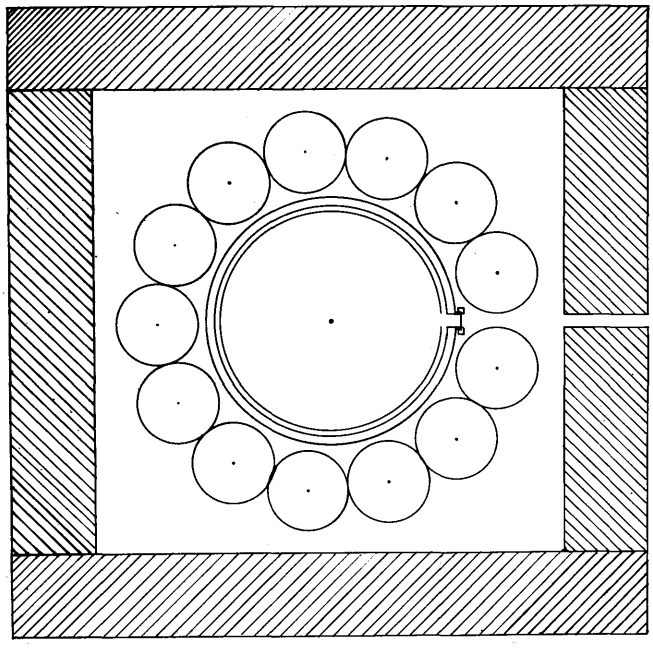


Fig. 50. Proportional counter with lead and anticoincidence shielding.

partial pressure of 20 cm. plus argon to a total pressure of five atmospheres.

The source was prepared by depositing spectroscopically pure rubidium chloride evenly on the inner surface of an aluminium cylinder which was inserted into the counter tube and which acted as the cathode. The surface of the aluminium was firstly thoroughly cleaned with hot concentrated sodium hydroxide solution. The rubidium chloride (~1 gm) was dissolved in hot water and laid down smoothly using a fine brush, over an area ~ 700 cm^2 giving a source thickness of ~1.5 mgm/cm^2. Later when a thinner source (0.128 mgm/cm^2) was used to investigate the low energy region and to determine the half life accurately, a slightly different technique was used to ensure that all of the carefully weighed chloride was deposited on the foil. In this case a specially prepared glass wiper was used to spread the source evenly. Both sources appeared to be uniform in thickness and care was taken to keep the sources sufficiently far from the non-counting regions so that no particles could enter these regions.

Using the thicker source at two different gain settings in the exact ratio of 4:1, the spectrum shape indicated by the open circles and dots in fig. 51 was obtained. These two sets of results were fitted by dividing the intensity values of the higher energy readings (dots) by four and multiplying the abscissae by four, since the time of running at each gain setting was the same. As can be seen from fig. 51, a good fit was obtained. The shape of the spectrum at high energies is shown in greater detail in the inset figure. From this graph, after subtracting the background spectrum, the upper energy limit, E_o, was calculated to be ~275 KeV, a result which was confirmed by the Fermi plot, shown later. The region of low energy, i.e. below 40 KeV, was examined separately using the thinner

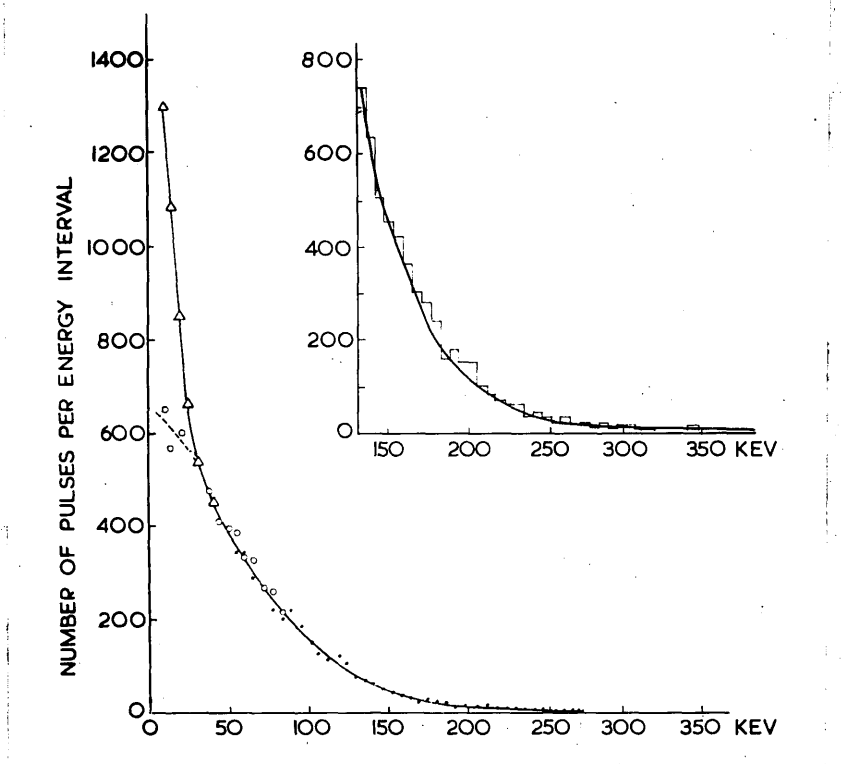


Fig. 51. Beta-spectrum of Rb^{87}.

source (0.128 mgm/cm^2). Fig. 52 shows the results obtained. The curve A is that obtained from the spectrometer, curve C is the separately determined background spectrum and B is the spectrum shape with background subtracted. This curve was fitted to be spectrum of fig. 51 (shown by triangles) and smooth coincidence over the range 30 - 40 KeV was obtained. Fig. 51 was obtained from photographic analysis results while fig. 52 was obtained from single channel "kick-sorter" data. It is evident that, below 30 KeV, serious absorption of the beta-particles took place in the thicker source (see dashed line). It is believed that the final form of the spectrum shape obtained is accurate down to ~10 KeV. No maximum appears on the curve which falls steeply at first with increasing energy and then exhibits an extremely long tail.

The "allowed" Fermi plot is shown in fig. 53. N is the number of particles per energy interval, p the momentum, W the energy in Mc^2 units and F is given by

$$F(Z,W) = \left[\frac{4}{[\Gamma(2s+1)]^2} R^{2(s-1)}\right]\left\{W^2(1+4Z^2\alpha^2)-1\right\}^{s-1} \times F_N(Z,W)$$

where $$F_N(Z,W) = \frac{2\pi Z\alpha W}{p} \frac{1}{1-e^{-2\pi Z\alpha W/p}}$$

$$s = \sqrt{1-\alpha^2Z^2},\ \alpha = \frac{1}{137},\ Z = \text{atomic no.},\ R = \text{nuclear radius in } \hbar/2\pi mc \text{ units.}$$

The factor contained in the square brackets is constant and was omitted. At this particular value of Z the simpler Bleuler and Zünti method of constructing Fermi plots cannot be employed. The curve departs very markedly from the straight line form given by an allowed transition. The shape of the curve is in qualitative agreement with that expected for a third forbidden transition with a spin change of three. The plot is linear in the region of the maximum energy, E_0, and extrapolation of this line gives E_0 = 275 KeV which is in agreement with direct observation. When this result is combined with the value of the half life obtained in this work,

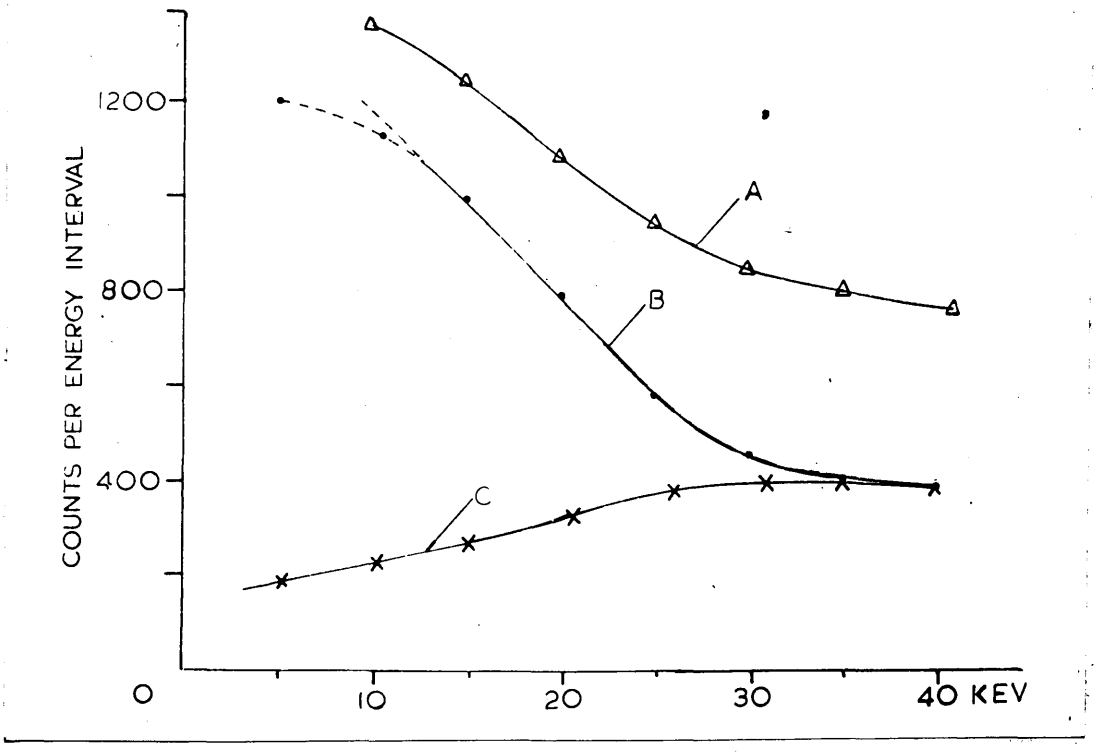


Fig. 52. Beta-spectrum of Rb^{87}, using 0.128 mgm/cm^2 source.

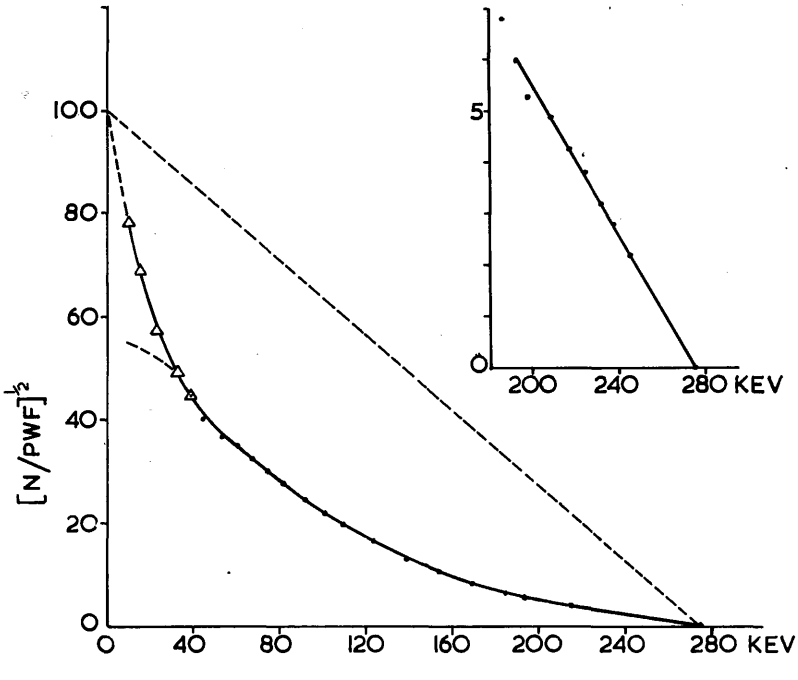


Fig. 53. Fermi plot of beta-spectrum of Rb^{87}.

the log ft value becomes 17.6, using the data supplied by Feenberg and Trigg (1949). The correction factor C_{3T} given by Greuling (1942) can be put in the following, slightly simplified form:-

$$C_{3T} = \left\{ \frac{1}{30}(W_0 - W)^4 \left[\left(\frac{\alpha Z}{2\rho}\right)^2 + \frac{1}{3}\left(\frac{\alpha Z}{\rho}\right)\frac{p^2}{W} + \frac{p^2}{9} \right] \right.$$
$$+ \frac{1}{36}(W_0 - W)^2 p^2 \left[\left(\frac{\alpha Z}{2\rho}\right)^2 + \frac{2}{5}\left(\frac{\alpha Z}{\rho}\right)\frac{p^2}{W} + \frac{4p^2}{25} \right]$$
$$+ \frac{1}{270} p^4 \left[\left(\frac{\alpha Z}{2\rho}\right)^2 + \frac{3}{7}\left(\frac{\alpha Z}{\rho}\right)\frac{p^2}{W} + \frac{9p^2}{49} \right]$$
$$+ \frac{1}{35}(W_0 - W)^5 \left[\left(\frac{\alpha Z}{2\rho}\right) + \frac{2p^2}{7W} \right] + \frac{1}{9}(W_0 - W)^3 p^2 \left[\left(\frac{\alpha Z}{2\rho}\right) + \frac{2}{5}\frac{p^2}{W} \right]$$
$$+ \frac{7}{135}(W_0 - W) p^4 \left[\left(\frac{\alpha Z}{2\rho}\right) + \frac{2p^2}{7W} \right] + \frac{1}{840}(W_0 - W)^6 + \frac{1}{54}(W_0 - W)^4 p^2$$
$$\left. + \frac{7}{1800}(W_0 - W)^2 p^4 + \frac{1}{1960} p^6 \right\} + y^2 \left\{ \frac{1}{30}(W_0 - W)^4 + \frac{1}{9}(W_0 - W)^2 p^2 + \frac{1}{30} p^4 \right\}$$
$$+ 2y \left\{ \frac{1}{10}(W_0 - W)^4 \left[\left(\frac{\alpha Z}{2\rho}\right) + \frac{1}{3}\frac{p^2}{W} \right] + \frac{5}{18}(W_0 - W)^2 \left[\left(\frac{\alpha Z}{2\rho}\right) + \frac{2}{5}\frac{p^2}{W} \right] + \frac{7}{90} p^4 \left[\left(\frac{\alpha Z}{2\rho}\right) + \frac{3}{7}\frac{p^2}{W} \right] \right\}$$

Where W_0 is the maximum energy in Mc^2 units, W is the [illegible] and the factor $\left(\frac{\alpha Z}{2\rho}\right) \approx (2Z)^{2/3}$ the calculation is being made, p is the momentum and the factor $\left(\frac{\alpha Z}{2\rho}\right) \approx (2Z)^{2/3}$, y is an adjustable factor (the ratio of the matrix elements $Q_3(\beta\alpha, r)$ and $Q_3(\beta\sigma \times r, r)$ in Greuling's notation) for which different values must be substituted until a straight line Fermi plot is obtained. Thus the correction factor C_{3T} is not unique and is very laborious to calculate; hence the calculation was not completed. It is probable, however, that the adjustment can be made. *

It will be seen that the value of E_0 obtained here is in agreement with that estimated by Klemperer on the basis of data obtained by Mühlhoff. The value obtained by Libby and Lee is much lower, viz. 132 KeV. This can be understood in terms of the peculiar shape of the spectrum and of the thick source Libby and Lee are believed to have used. If one considers fig. 51, it is easily seen that the addition of a large background spectrum

* See section 8.

to the Rb^{87} spectrum, with its long tail, could easily lead one to estimate the end-point to be ~130 KeV. The background spectrum of Libby and Lee was relatively much larger in relation to the Rb^{87} counting rate. Also the use of a thick source tends to reduce the number of high energy electrons and increase the number of low energy ones, thus further emphasising the peculiar spectrum shape. Also it should be noted that the values of E_0 for other activities obtained at this time by Libby and Lee using the screen - wall counter are also low by later and more precise work. In the following list of values of E_0 obtained by these workers, the modern value of E_0 follows in brackets:-

S^{35}, 107 KeV (169 KeV); Au^{198}, 770 KeV(970 KeV);

K^{40}, 725 KeV (1.4 MeV); Rb^{87}, 132 KeV(275 KeV).

It might be supposed that the long tail of the spectrum is due to the integrating properties of the proportional counter i.e. due to the addition of one or more coincident conversion electrons to the beta-particles. However work still to be discussed shows that there is no appreciable X-ray or gamma-ray intensity and also, since 2π geometry is employed, one would expect to observe strong conversion lines superposed on the spectrum if such a process were taking place.

Saha's value (144 KeV) was also obtained with a screen-wall counter of almost identical design to Libby's and he, too, used a very thick source (o.3 gm/cm^2). His value of E_0 for S^{35}, 103 KeV, is likewise low and is almost the same as Libby's. Hence the same criticism applies to his work. Although Ollano is quoted as giving a value of 130 KeV for E_0, he merely points out that the spectral limit which he obtained was not inconsistent with Libby's and that "the spectrum limit ..... does not fall precisely at 130 KeV, as is said by Libby and Lee, but a little above this value."

As a matter of fact the electron spectrum published by Ollano consists mainly of a series of peaks and the intensity actually approaches a maximum near 130 KeV, beyond which energy he did not make any observations. Further discussion of his results will follow in section 4.

The work of Haxel, Houtermans and Kemmerich seemed to indicate that two particles of energies ~10 KeV and ~135 KeV were emitted in coincidence per disintegration. They examined the radiations from the two sides of a thin layer of rubidium chloride evaporated on a 0.1 mgm/cm^2 silver coated Zapon foil which formed the dividing wall between two Geiger counters. Their suggestion that the 135 KeV electrons were photoelectrons and that the 10 KeV electrons formed the beta-spectrum was based on the following grounds: (a) 10 KeV suits the Sargent Diagram better, (b) Ollano's conversion lines and (c) it is difficult to imagine a conversion line of such low energy as 10 KeV in coincidence. Anticipating some later findings, the suggestion of the emission of two electrons per disintegration can be eliminated. It is clear that the unusually high intensity of the beta-spectrum at low energies explains to some extend their observations and it seems fairly certain that the coincidences were due to electrons making counts and then being reflected by the counter wall or gas into the other counter, thus giving two counts in coincidence. They used this experiment to determine the half life of Rb^{87} but in such a way that the question as to the decay scheme did not affect the result. They thus got a value of τ in good agreement with the geological method and with the value obtained in the present research. *

4. Gamma Radiation and Conversion.

As was mentioned earlier, the non-appearance of conversion lines superposed on the spectrum was in disagreement with Ollano's results, since

* See also section 8.

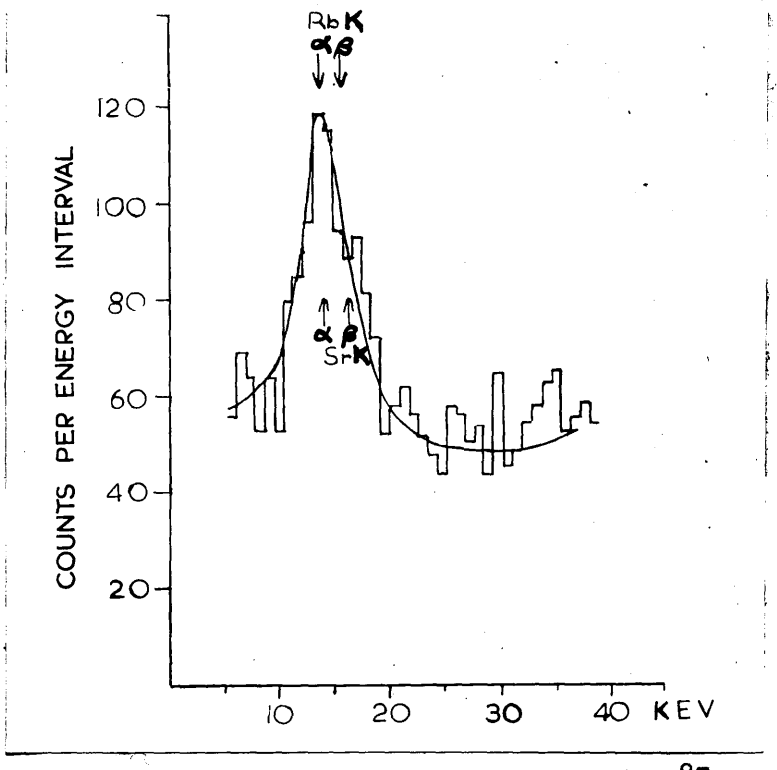


Fig. 54. K X-rays observed with Rb^{87}.

his spectrum seemed to consist mainly of conversion lines. Hence a sensitive search for the X-rays which would result from such conversion was undertaken. The thick source (1.5 mgm/cm^2) of RbCl was covered with sufficient aluminium to stop all beta-rays (58 mgm/cm^2). This absorber reduces the intensity of the K X-rays of strontium to about 50%. The spectrum of such X-rays was observed at two different values of gain and one of the resulting histograms is shown in fig. 54, in which a peak of low intensity sppears. The average efficiency of the counter, at two atmospheres pressure of argon, for such rays was assessed at ~84%. By estimating the area under the peak, the total number of X-rays emitted in unit time was determined from the histogram and comparison with the number of beta-rays emitted in unit time showed that the X-rays were down in intensity relative to the beta-rays by a factor of more that 500. The extremely low intensity of X-radiation made accurate atomic number assignment of the X-rays difficult but from fig. 54 it appears that allocation to rubidium rather than strontium is preferable. This intensity of X-rays is not inconsistent with the view that the X-rays are generated by bombardment of the rubidium by the beta-radiation. In any case it seems certain that internal conversion in the K-shell of Sr^{87} does not occur in more than 0.2% of the disintegrations. This result completely contradicts the conclusions of Ollano and leads to the suggestion that the electron groups observed by him are spurious and are the result merely of statistical fluctuations.

However it was still possible, though only remotely so, that the gamma-radiation, if any, might convert in the L-shell only. Hence a separate search for L X-ray quanta was carried out. Since the L X-rays of strontium (~1.8KeV) would be almost completely absorbed by an aluminium

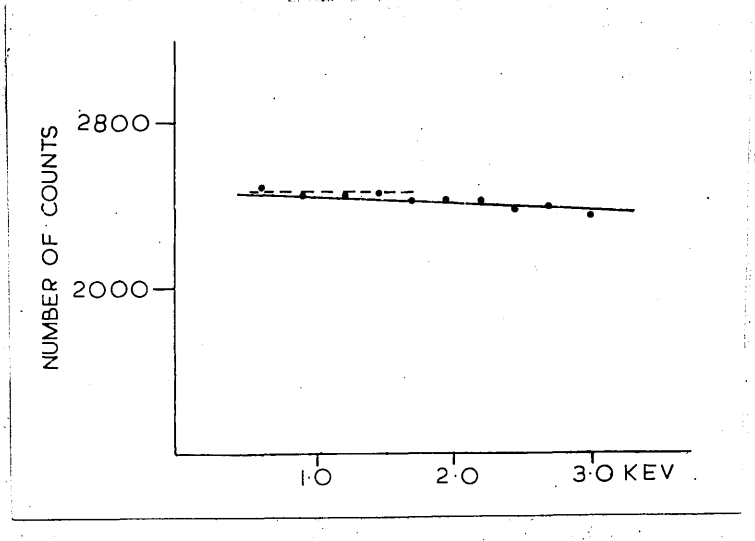


Fig. 55. Search for L X-rays. Counting rate as discriminator bias is varied over a range including the L-radiation.

beta absorbing screen, another method had to be devised, as follows. The counter with the thicker rubidium source, but with no absorber, was connected through a discriminator unit to a scaler. Using as calibration the X-rays emitted in the decay of Ge^{71}, the discriminator bias setting corresponding to an energy of 1.8 KeV was determined. Now if L X-rays are emitted in some disintegrations, in 50% of such cases the X-ray will not be detected in coincidence with a beta-particle (due to the use of 2π geometry). Hence if the bias setting is varied over a wide range below and above the setting corresponding to 1.8 KeV, a sharp rise in counting rate should accur at that setting. Fig. 55 shows that no appreciable discontinuity can be found. Even if the slow rise on the curve is distorted to create a discontinuity as shown by the dashed line in the figure an upper limit of 3% is found for the number of disintegrations accompanied by L X-rays. It is very much more probable, however, that the upper limit is not greater than for the K-radiation, ie. 0.2%. Thus there is no real evidence of conversion of gamma-radiation in either K- or L-shells in more than 0.2% of the disintegrations.

A sensitive search for gamma-radiation itself was carried out by Mr. Bannerman of the Natural Philosophy Department. He used a sodium iodide (thallium activated) scintillation crystal surrounded by ~ 5.6 gm. of rubidium chloride. The crystal was shielded from the beta-rays with aluminium. There appeared to be a very small additional counting rate with the source in position, but there was no indication whatever of homogeneity of the pulses produced, which were almost certainly due to bremsstrahlung excited by the passage of beta-rays through the aluminium. Whatever the origin of the pulses, it can be said with certainty that gamma-radiations in the range 20-100 KeV cannot be released in more than one per 5000

disintegrations and in more than one per 1000 disintegrations in the range 100-300 KeV.

Thus as a result of the search for K X-rays, L X-rays and gamma-quanta and because of the lack of conversion peaks in the beta-ray spectrum, it appears certain that the probability of the beta-decay leaving the nucleus Sr^{87} in an excited state is negligibly small. The decay process consists of a beta-transition between the ground states of Rb^{87} and Sr^{87}.

5. The Period τ.

The most recent accurate value of τ obtained directly from the radioactivity is that due to Haxel, Houtermans and Kemmerich. It has already been pointed out that the work was based on wrong assumptions regarding the mode of decay of Rb^{87}, though, as they themselves pointed out, their method of deducing the number of disintegrations gives a value which is independent of their assumptions regarding the decay scheme. In fact it is equivalent to correcting for the reflection of the beta-particles. It was thought of importance to make a separate, very careful determination of τ to compare with theirs, and with the geological value.

The thin source used formthe examination of the low energy part of the spectrum (0.128 mgm/cm^2) was also used for the determination of τ. The rubidium chloride employed was spectroscopically pure and was shown to contain only 0.018% of moisture, i.e. a negligible amount. 0.10075 gm. of the chloride was weighed out and deposited very carefully and evenly on an aluminium backing as described in section 2. This source gave rise to an additional 1477 counts per minute with a statistical accuracy of better than 1%. Allowing for the solid angle of 2π, chemical composition and isotopic abundance, this gives directly a value for τ of 5.95×10^{10} years.

This value must, however, be corrected for (a) reflection of particles

in the source and support and (b) the self-absorption of the source. In the estimation of (a), data on reflection due to both Yaffe (1950) and Burtt (1950,1949) was used. Their results are in reasonably good agreement but both plot the reflection coefficient as a function of the maximum energy E_o. However in view of the remarkable shape of the Rb^{87} spectrum it appears more desirable to estimate the reflection in terms of the average energy, $\bar{E}$, of the beta-rays. Thus, although the value of E_o for Rb^{87} (275 KeV) is considerably in excess of those of S^{35} (169 KeV) and C^{14} (158KeV), the average energy of Rb^{87} is only 44 KeV, less than the values of 54 KeV and 52 KeV for S^{35} and C^{14} respectively. As result of much consideration a value of 7.5% was finally adopted for the reflection coefficient. In the estimation of (b) it was assumed that the source was equivalent in thickness to electrons of energy 5 KeV. Then the number of 4 KeV, 3 KeV, 2 KeV and 1 KeV electrons absorbed by the source was calculated and thus the fraction of all electrons of energies less than 5 KeV, failing to get out of the source, was found. The ratio of the number of these electrons to the total number emitted was calculated by extrapolating the spectrum of fig.51 as a straight line to zero energy. The value deduced was 4.5%. Thus the total contribution to the counting rate due to these causes is (7.5 - 4.5)%, giving a value of τ of 5.95 x 1.03 or 6.15 x 10^{10} years. The chief uncertainties in the value arise from the estimation of the errors and, allowing for these and for the statistical error, an error of ~4% was believed to be possible. Thus with these rather conservative limits the final value of τ becomes

$$\tau = 6.15 \pm 0.3 \times 10^{10} \text{ years.}$$

6. Conclusions and Discussion.

The experiment showed that the beta-decay of Rb^{87} is simple, involving a transition from the ground state of Rb^{87} to the ground state of Sr^{87}.

The maximum energy, E_o, is found to be 275 KeV. The shape of the spectrum is qualitatively in agreement with that predicted for a spin change of three with a change of parity. So far as future work is concerned it does not seem that any outstanding experimental work requires still to be done (unless for confirmation) and further interest in the decay must lie principally with the theorists. However, since NaI and KI crystals have been used successfully as scintillators, the writer feels it would be of interest to re-investigate the beta-spectrum and possible very weak electromagnetic radiations by the use of a RbI crystal.

There are, however, many other interesting problems in the field of natural radioactivity and for this purpose a new type of counter has been designed by the writer specially for this purpose. It is shown in fig.56. It is so arranged that one of the end plates can be removed to insert a source without removing the wire as is always the case in the ordinary design of counter. It is often necessary in studies of this kind to open the counter frequently e.g. to compare the counting rate with and without the source present, to investigate the beta activity of a source and then the gamma activity with the beta-particles absorbed out, to compare different sources, etc. The counter is end-corrected and possesses two 0.002" aluminium windows, one at the centre of the counting volume, the other at one end of the counting volume. The counting volume is 10" long and 5.5" in diameter.

A very interesting fact with regard to the natural radioactivities has been pointed out by Dr. S.C. Curran. In table 2 are listed the calculated log ft values for the natural radioactivities.

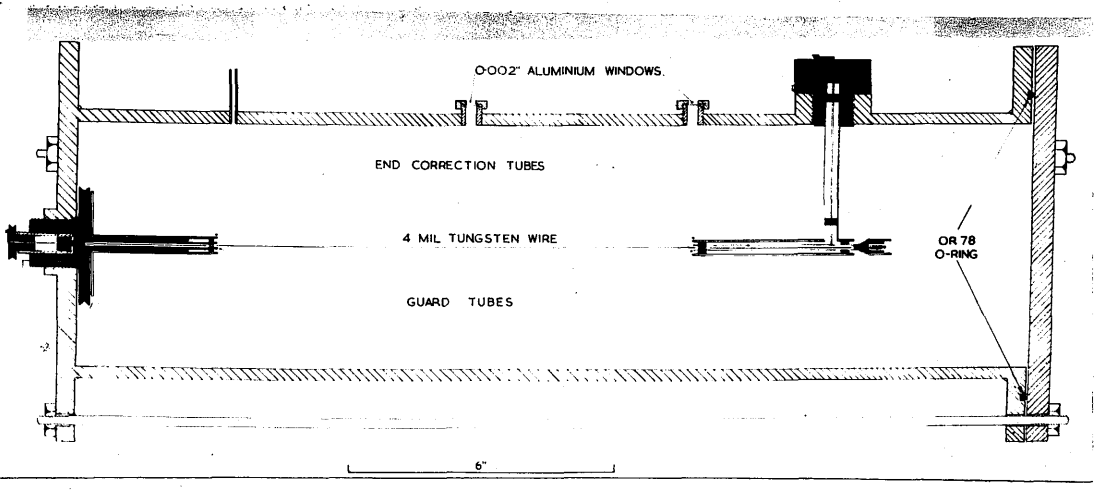


Fig. 56. Special counter built for investigation of natural radioactivities.

Table 2.

Source	E_o (MeV)	10^{10} yr	log ft
Rb^{87}	0.275	6.15	17.6
K^{40}	1.36	0.11	18.05
Lu^{176}	0.215 or 0.4	2.4	18.02 or 18.91
Re^{187}	0.043	4.0	17.73
Nd^{150}	0.011	5	13.73

Using the new value of E_o, 275 KeV, the value of log ft for Rb^{87} rises from 16.5 to 17.6. A remarkable grouping about log ft = 18 is immediately apparent. A considerable gap exists between this value and that of the next well - established value of 13.65 for the artificially radioactive isotope Be^{10} though a fairly uniform distribution exists below this value. The value of log ft for Nd^{150}, calculated for E_o = 11 KeV and $\tau = 5 \times 10^{10}$ years appears as an obvious exception. However preliminary work carried out in Glasgow and in America has shown that these values, which are due to Libby (1934), are almost certainly wrong. This work will be briefly described in the following section. One may predict, on the basis of this fact, that the half life and/or the value of E_o should be greater. If Libby's value of the half life were used, an upper energy limit of 250 KeV would be predicted.

A brief account of this work was published by Curran, Dixon and Wilson (1951) and a fuller account by the same authors appears in the Philosophical Magazine (1952).

7. Preliminary Work on Nd^{150}.

Libby (1934) using the screen-wall counter obtained a half life of

1.5×10^{12} years for the activity and an upper energy limit for the beta-spectrum of 11 KeV. Kohman (1948) corrected Libby's value of the half life for isotopic abundance on the assumption that Nd^{150} was the active isotope and this gave a half life of $\sim 8 \times 10^{10}$ years.

On the basis of these figures it was calculated that 1 gm. of neodymium should give a total counting rate of 5920 counts per minute, an easily detectable activity. Of course, only half this rate is expected if the source is mounted on the wall of the counter. The source was prepared by disolving 0.413 gm. of spectroscopically pure neodymium in hydrochloric acid, the solution evaporated to dryness, the residue dissolved in a 5% solution of insulin in water and the solution brushed evenly over a hot aluminium liner, as in the case of rubidium. The liner was inserted in a large proportional counter which was filled to a pressure of two atmospheres of argon plus methane to a partial pressure of 10 cm. of mercury. From 0.413 gm. one would expect a counting rate of 1225 counts per minute above background. Actually the increase in counting rate was certainly not more than 50 counts per minute above background and the spectrum obtained by pulse analysis did not differ to a significant extent from the background spectrum.

It appears, therefore, that the activity, if it exists, is of very much longer half life than that stated by Libby. Also there is no evidence, within the limitations of the apparatus, of a spectrum ending at 11 KeV. However this experiment was operated without the anticoincidence counters with the result that the background rate was rather high, ~ 400 counts per minute. It is hoped to make another attempt in the near future with larger quantities of source and lower backgrounds, since, at the present time, nothing definite appears to be known regarding this decay.

In a very recent article of Mulholland and Kohman (1952) and in a private communication of Kohman's to the author, an investigation of the activity of Nd^{150} was described. A 15 gm. layer of highly purified neodymium oxide in a proportional tube gave a counting rate in excess of background ($\sim$60 counts per minute) of 4 counts per minute. If this were due to Nd^{150}, it would correspond to a half life of 2×10^{15} years. However it is believed to be due to the increase in the gamma-ray detection sensitivity of the counter, caused by the source layer. This increase in sensitivity was calculated to be $\sim$11%. All that can be said, therefore, is that the half life of the activity is greater than 2×10^{15} years.

8. Recent Work on Rb^{87}.

Two very recent papers on this activity should be mentioned. Firstly, MacGregor and Wiedenbeck (1952) made another determination of the half life obtaining a value of $6.23 \pm 0.3 \times 10^{10}$ years which is in good agreement with our own value. They also repeated Haxel, Houtermans and Kemmerich's experiment but showed that the decay is simple, in agreement with our own work and in contradistinction to the results of Haxel et al. Secondly, Tomozawa, Umezawa and Nakamura (1952) applied the C_{3T} correction factor to the Fermi plot obtained in this work and showed that it gives a straight line from 100 KeV to the end-point at 275 KeV. If the effect of screening is allowed for, theplot is linear down to 50 KeV. They suggest that the lack of linearity below this could be due to "internal bremsstrahlung" electrons or to experimental difficulties. They also showed that the C_{3V} factor, as well as the C_{3T}, fits the spectrum, but that no other factor does. It will be remembered that vector (V), scalar (S) and pseudoscaler (P) interactions all yield Fermi selection rules, while tensor (T) and axial vector (A) interactions yield Gamow-Teller selection rules. Thus, in view

of the weight of evidence supporting Gamow-Teller rules, this experiment suggests strongly that the "tensor" interaction is the correct choice, though it may finally prove impossible to explain all spectra by the use of any one interaction alone.

Mr. G.M. Lewis has recently examined the beta-spectrum of Rb^{87} by the technique suggested in section 6, viz. using a rubidium iodide crystal in a scintillation spectrometer arrangement. He obtained a spectrum whose shape is in good agreement with that obtained in this work.

APPENDIX.

EXPERIMENTS ON THE FLUORESCENCE OF ANTHRACENE.

The experiments to be described were undertaken at the suggestion of Dr. J.B. Birks in order to test a prediction which he had made.

The absorption spectrum of anthracene in the crystalline form has been investigated several times, for example, by Kortüm and Finkh (1942) and it may be represented, in a somewhat simplified form, as in fig. 57. The fluorescence spectrum is a mirror image of the first absorption band. This phenomenon is discussed by Pringsheim (1949, p.302) and is to be expected on theoretical grounds.

Since another absorption band has been shown to exist about 2500 Å (fig.57), one might expect to find a fluorescence band displaced somewhat to longer wavelengths and, probably, forming a mirror image of the second absorption band. The predicted fluorescence band is shown dotted in fig. 57.

Before describing the experiments designed to test the prediction, it will be useful to consider the "potential curve" or "Jablonski" method of indicating excited and ground states which is more commonly used in the present type of study than the ordinary energy level diagrams and is more useful since it gives some indication of the intensities of the various transitions. Fig.58 shows such a diagram for the case of a diatomic molecule, which is the simplest one to consider. U is the potential energy of the molecule at different distances r of one atom from the other. One atom may be considered to be at the origin. D corresponds to the dissociation energy of the molecule. The horizontal lines represent the vibrational energy levels and the length of each line corresponds to the amplitude of that vibration. Now, since (a) the atoms spend most of their

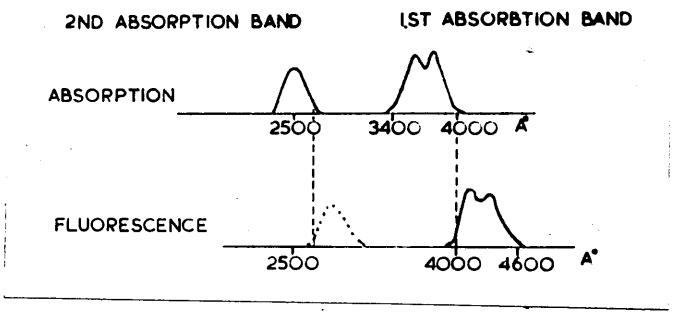


Fig. 57. Absorption and fluorescence spectra of crystalline anthracene.

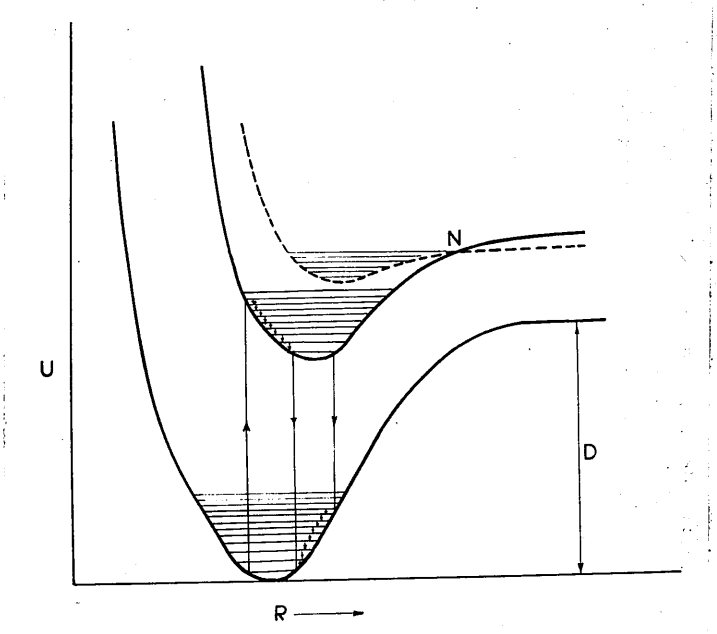


Fig. 58. Jablonski diagram for diatomic molecule.

time at the ends of the vibrational motions, the most probable transitions take place from the ends of the vibrations and since (b) the electronic motion is very much more rapid than the nuclear motion, the transitions are represented by vertical lines - since r does not change appreciably in the time for an electronic transition to take place (Frank - Condon principle). Such transitions are shown in fig. 58. The molecule in the diagram is excited from the lowest vibrational level of the ground state to the eighth level of the first excited state and then energy is lost by transitions between vibrational levels. Eventually a transition is made from some level (in the diagram, the lowest level) of the first excited state to the ground state (fluorescence radiation). Two possible transitions are shown. One transition goes directly to thelowest vibrational level in the ground state while the other goes to the eighth vibrational level, the remaining energy being reduced by further transitions between vibrational levels. An important point must be noted here. From what has been said one might expect all the transitions to take place in one molecule alone. This is not the case. Transitions between vibrational levels are normally associated with a collision between the excited molecule and another, which carries off the energy, less the small amount corresponding to the energy difference between the vibrational levels. In this way fig. 58 might correspond to transitions taking place in seven or fifteen different molecules.

The existence of the vibrational levels gives the reason, of course, for the broadness of the absorption and fluorescence bands. In the case of a more complex molecule like anthracene, the picture becomes much more complicated. Fig.58 may be considered to represent the case of anthracene, though not quantitatively, if r is interpreted as the distance between two

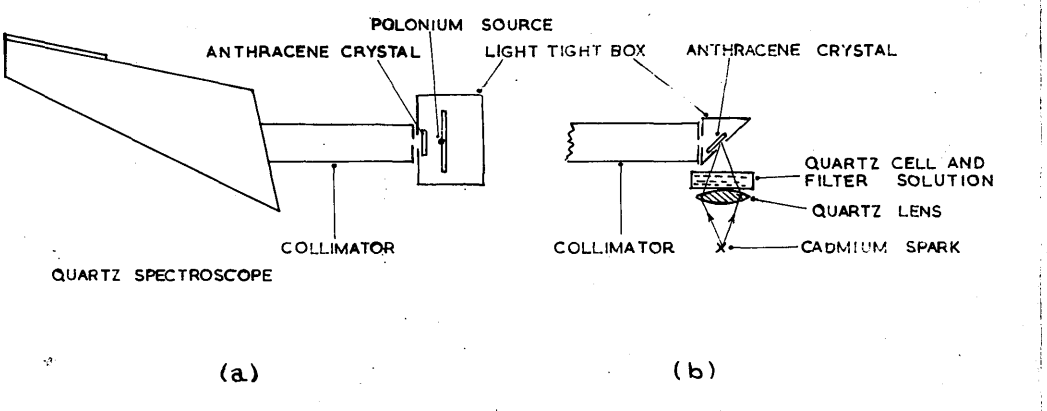


Fig. 59. Experimantal arrangement (a) using alpha-source, (b) using ultra-violet illumination.

atoms, or two parts, of the molecule.

Experimental Procedure.

In the first instance it was decided to escite the molecules of anthracene by exposing an authracene crystal to alpha-particle bombardment. A strong polonium source ($\sim$ 6mC) was placed close to a crystal and the emitted spectrum examined using a Hilger Quartz Spectroscope, which could be used to measure wavelengths between 1800 Å and 7000 Å. The experimental arrangement is shown in fig. 59(a). An exposure of several hours duration was made using the fastest plate obtainable, i.e. Ilford HP3. This exposure gave only a very faint image at the first fluorescent band, but nothing at the supposed position of the second. An exposure of $\sim$ 48 hours was then made. It was noticed at the end of this period that the crystal fluoresced scarcely at all, although the initial fluorescence had been quite apparent. The crystal itself was discovered to have a very distinct brownish discolouration. This was believed to be due to decomposition of the anthracene molecules. It was known that many fluorescent materials were somewhat easily altered (generally for the worse from the fluorescent standpoint) by strong overexposure to ultra-violet light, but it is believed that it had not been observed before with particle bombardment, at any rate for organic phosphors. The result is not surprising but it is nevertheless important in view of the considerable and extending use of scintillation counting techniques. The fact was reported by Birks in a letter to the Physical Society (Birks, 1950) and the phenomenon was further investigated by Birks and Black (1951), who found that the number of scintillations remained constant but that the pulse size was reduced considerably (to $\sim$3% of the original) after bombardment by $\sim 3 \times 10^{12}$ alpha-particles per square centimetre. The law governing the reduction

of pulse size was approximately

$$\frac{I}{I_o} = \frac{1}{1 + AN}$$

where I/I_o is the ratio of the pulse amplitudes after and before bombardment by N alpha-particles per cm^2. A is a constant having the value $\sim 10^{-11}$.

Returning to the search for the proposed second fluorescence band, it was decided to use instead of the polonium source, the cadmium 2469.8 Å line whose wavelength coincides approximately with the centre of the second absorption band. The apparatus used is shown in fig. 59(b). The light from the cadmium spark was passed through a quartz condensing lens of short focal length, through a filter which absorbed light above 3500 Å and focussed on the anthracene crystal. To cut down the stray light reaching the plate the crystal was arranged at such an angle that the light from the cadmium spark was reflected away from the slit. This, however, did not prevent the fluorescence radiations entering the collimator. The filter solution was prepared using a formula due to Bowen, i.e. 49 gm. of hydrated nickel sulphate and 13 gm. of hydrated cobalt sulphate dissolved in 100 c.c. of water. The sulphates must be pure and, especially, iron-free. The solution thickness was 1.5 cm. Several exposures were made, on HP.3. plates, with different exposures and different distances of the cadmium spark. In none of these plates was there any indication whatsoever of a second fluorescence band. Indeed, if one does exist, the intensity relative to the fluorescence band is less than one part in two hundred.

The result can be explained by saying that the transition from the second excited state to the first is radiationless and is accomplished entirely by a series of transitions from one vibrational level to another.

In this case the potential curve of the second excited state must intersect the first excited state as in the tentative curve drawn (dashed) in fig. 58. (Pringsheim, 1949, p.246). This means that the energy could be transferred from a low vibrational energy level in the second excited state of one molecule to a higher vibrational energy level of the first excited state of a second molecule and no fluorescent radiation being emitted.

This experiment, therefore, showed no evidence of a fluorescence band corresponding to the second absorption band. However it would be interesting to find out whether excitation to higher energy levels (probably by electron or X-ray excitation) would give rise to other fluorescence bands.

REFERENCES

Agnew, H.M., 1950, Phys. Rev. 77, 655.

Agnew, H.M. and Anderson, H.L., 1949, Rev. Sci. Inst.20, 869.

Albert, R.D. and Wu, C.S., 1948, Phys. Rev. 74, 847.

Alburger, D.E., 1950, Phys. Rev. 79, 236.

Allen, J.S., 1942, Phys. Rev. 61, 692.

Anderson, E.C., Arnold, J.R. and Libby, W.F., 1951, Rev.Sci.Inst.22,225.

Angus, J., Cockroft, A.L. and Curran, S.C., 1949, Phil. Mag.40, 522.

Backus, J., 1945, Phys. Rev. 68, 59.

von Baeyer, O. and Hahn, O., 1910, Phys. Zeit. 11, 488.

von Baeyer, O., Hahn, O. and Meitner, L., 1911, Phys. Zeit. 12, 273, 378;
1912, Phys. Zeit. 13, 264.

Bannerman, R.C. and Curran, S.C., 1952, Phys. Rev. 85, 134.

Bannerman, R.C., Lewis, G.M. and Curran, S.C., 1951, Phil. Mag.42, 1097.

Bethe, H.A., 1947, "Elementary Nuclear Theory" (Wiley).

Bethe, H.A. and Bacher, R.F., 1936, Rev. Mod. Phys. 8, 194.

Binder, D., 1950, Phys. Rev. 78, 325.

Birks, J.B., 1950, Proc. Phys. Soc. A 63, 1294.

Birks, J.B. and Black, F.A., 1951, Proc. Phys. Soc. A 64, 511.

Bleuler, E. and Zünti, W., 1946, Helv. Phys. Acta. 19, 375.

Bonetti, A., 1950, Bull. du Centre de Phys. Nuc. de Bruxelles, May, No.19.

Braden, C.H., Owen, G.E., Townsend, J., Cook, C.S. and Shull, F.B., 1948,
Phys. Rev. 74, 1539.

Brosi, A.R., Borkowski, C.J., Conn, E.E. and Griess, J.C., 1951,
Phys. Rev. 81, 391.

Brosi, A.R. and Griess, J.C., 1949, ORNL - 499 (unpublished)

Brown, H. and Perez - Mendez, V., 1949, Phys. Rev. 75, 1286.

Burtt, B.P., 1950, "Absolute Beta Counting", Nuclear Science Series, Prelim. Report No. 8.

Burtt, B.P., 1949, Nucleonics 5, No. 2, 28.

Butement, F.D.S., 1951, Nature 167, 400.

Campbell, N.R. and Wood, A., 1906, Proc. Camb. Phil. Soc. 14, 15.

Chadwick, J., 1914, Verh.d.D. Phys. Ges. 16, 383.

Cockroft, A.L. and Curran, S.C., 1951, Rev. Sci. Inst. 22, 37.

Cockroft, A.L. and Valentine, J.M., 1950, Jour. Sci. Inst. 27, 262.

Conn, E.E., Brosi, A.R., Swartout, J.A., Cameron, A.E., Carter, R.L., and Hill, D.G., 1946, Phys. Rev. 70, 768.

Cook, C.S., Langer, L.M. and Price, H.C., 1948, Phys. Rev. 73, 1395.

Cork, J.M., Shreffler, R.G. and Fowler, C.M., 1948, Phys. Rev. 74, 240.

Corson, D.R. and Wilson, R.R., 1948, Rev. Sci. Inst. 19, 207.

Crane, H.R., 1948, Rev. Mod. Phys. 20, 278.

Crane, H.R. and Halpern, J., 1938, Phys. Rev. 53, 789.

Crane, H.R. and Halpern, J., 1939, Phys. Rev. 56, 232.

Curran, S.C., 1950, Atomics 1, 221.

Curran, S.C., 1952, Atomics 3, 5.

Curran, S.C., Angus, J. and Cockroft, A.L., 1948, Nature 162, 302.

Curran, S.C., Angus, J. and Cockroft, A.L., 1949a, Phil. Mag. 40, 36.

Curran, S.C., Angus, J. and Cockroft, A.L., 1949b, Phil. Mag. 40, 53.

Curran, S.C., Angus, J. and Cockroft, A.L., 1949c, Phys. Rev. 76, 853.

Curran, S.C. and Baker, W., 1944, Rad. Lab. Rep. 7.6.16,Nov.17.

Curran, S.C. and Baker, W., 1947, Rev. Sci. Inst. 19, 116.

Curran, S.C., Cockroft, A.L. and Angus, J., 1949, Phil. Mag. 40, 929.

Curran, S.C., Cockroft, A.L. and Insch, G.M., 1950, Proc. Phys.Soc. A 63, 845.

Curran, S.C. and Craggs, J.D., 1949, "Counting Tubes" (Butterworths).

Curran, S.C., Dee, P.I. and Petrizilka, V., 1938, Proc. Roy.Soc. A 169, 269.

Curran, S.C., Dixon, D. and Wilson, H.W., 1951, Phys. Rev. 84, 151.

Curran, S.C., Dixon, D. and Wilson, H.W., 1952, Phil. Mag. 43, 82.

Danysz, J., 1913, Le Radium, 10, 4.

Deutsch, M., Elliott, L.G. and Evans, R.D., 1944, Rev. Sci.Inst.15, 178.

Deutsch, M. and Wright, W.E., 1950, Phys. Rev. 77, 139.

Dunworth, J.W., 1940, Rev. Sci. Inst. 11, 167.

Dzelepow, B., Kopjova, M. and Vorobjov, E., 1946, Phys. Rev. 69, 538.

Ellis, C.D. and Wooster, W.A., 1927, Proc. Roy. Soc. A 117, 109.

Ellis, C.D. and Skinner, H.W.B., 1924, Proc. Roy. Soc.A 105, 60.

Eklund, S., 1946, Arkiv. Mat. Astron. Fysik 33A, 60.

Fazzini, T. and Franchetti, S., 1948, Nuovo Cimento, 5, 311.

Feather, N., 1930, Phys. Rev. 35, 1559.

Feather, N., 1938, Proc. Camb. Phil. Soc. 34, 599.

Feather, N. and Richardson, H.O.W., 1948, Proc. Phys. Soc. 61, 452.

Feenberg, E. and Trigg, G., 1950, Rev. Mod. Phys. 22, 399.

Feingold, A.M., 1951, Rev. Mod. Phys. 23, 10.

Feister, I., 1950, Phys. Rev. 78, 375.

Feldman, L. and Wu, C.S., 1949, Phys. Rev. 76, 697.

Fermi, E., 1934, Zeits. für Phys. 88, 161.

Fermi, E., 1950, "Nuclear Physics" (University of Chicago Press)

Fernelius, W., 1946, "Inorganic Syntheses", Vol.2 (McGraw-Hill), p.234.

Flammersfeld, A., 1946, Naturwiss. 33, 280.

Friedlander, G.F., 1949, BNL - AS - 2 (unpublished).

Friedlander, G.F. and Wu, C.S., 1943, Phys. Rev. 63, 227.

Gamow, G. and Teller, E., 1936, Phys. Rev. 49, 895.

Geiger, H. and Klemperer, G., 1928, Zeits. für Phys. 49, 753.

Geiger, H. and Rutherford, E., 1912, Phil. Mag. 24, 618.

Geoffrion, C., 1949, Rev. Sci. Inst. 20, 638.

Gibson, W.M., Green, L.L. and Livesley, D.L., 1947, Nature 160, 534.

Glendenin, L.E., 1948, Nucleonics 2, No.1, 12.

Good, M.L., 1951, Phys. Rev. 83, 1054.

Greuling, E., 1942, Phys. Rev. 61, 568.

Gross, L. and Hamilton, D.R., 1950, Phys. Rev. 78, 318.

das Gupta, N.N. and Ghosh, S.K., 1946, Rev. Mod. Phys. 18, 225.

Hahn, O., Strassman, F. and Walling, E., 1937, Naturwiss. 25, 189.

Hamilton, D.R. and Gvoss, L., 1950, Rev. Sci. Inst. 21, 912.

"Handbook of Chemistry and Physics", 1949, 31st. ed.(Chemical Rubber Publishing Co.), p. 2031.

Hanna, G.C. and Pontecorvo, B., 1949, Phys. Rev. 75, 983.

Hayden, R.J., Reynolds, J.H. and Inghram, M.G., 1949, Phys. Rev.75, 1500.

Haxby, R.O., Shoupp, W.E., Stephens, W.E. and Wells, W.H., 1940, Phys. Rev. 58, 1035.

Haxel, O. and Houtermans, F.G., 1948, Zeits. für Phys. 124, 705.

Haxel, O., Houtermans, F.G. and Kemmerich, M., 1948, Phys.Rev. 74,1886.

Haxel, O., Jensen, J.H.D. and Suess, H.E., 1950, Zeits. für Phys.128, 295.

Hemmendinger, A.H. and Smythe, W.R., 1937, Phys. Rev. 51, 1052.

Heyden, M. and Kopfermann, H., 1938, Zeits. für Phys. 108, 232.

Hofstadter, R. and McInture, J.A., 1950a, Nucleonics 7, No.3, 32.

Hofstadter, R. and McIntyre, J.A., 1950b, Phys. Rev. 78, 619.

Hopkins, J.I., 1951, Phys. Rev. 77, 406.

Hughes, A.L. and Rojansky, V., 1929, Phys. Rev. 34, 284.

Inghram, M.G., Hayden, R.J. and Hess, D.C., 1947, Phys. Rev. 71, 643.

Inghram, M.G., Hess, D.C., Hayden, R.J. and Parker, G.W., 1947, Phys. Rev. 71, 743.

Insch, G.M., 1950, Phil. Mag. 41, 857.

Isotope Committee, 1946, Science 103, 697.

Ivanenko, D. and Lebedev, V., 1950, Jour. Exp. Theor. Phys. U.S.S.R. 20, 91 (Jan.). (English translation in Guide Russ. Sci. Period. Lit., Brookhaven, 1950, 3, June, 162 - 163).

Jackson, D.A., 1933, Proc. Roy. Soc. A 139, 673.

Jacobsen, J.C. and Kofoed - Hansen, O., 1948, Phys. Rev. 73, 675.

Kapitza, P.L., 1924, see p. 454 of Tricker (1924) reference.

Kemmerich, M., 1949, Zeits. für Phys. 126, 399.

Ketelle, B.H. and Parker, K.W., 1949, Phys. Rev. 76, 1416.

Klemperer, O., 1935, Phil. Mag. 20, 545.

Klemperer, O., 1935, Proc. Roy. Soc. A 148, 638.

Kohman, T.P., 1948, Phys. Rev. 73, 16.

Konopinski, E.J., 1943, Rev. Mod. Phys. 15, 209.

Konopinski, E.J. and Uhlenbeck, G.E., 1935, Phys. Rev. 48, 7.

Kopfermann, H., 1933, Zeits. für Phys. 83, 417.

Korff, S.A., 1942, Rev. Mod. Phys. 14, 1.

Korff, S.A., 1946, "Electron and Nuclear Counters" (Van Nostrand).

Korff, S.A., 1950a, Nucleonics 6, No.6, 5; 1950b, Nucleonics 7, No.5, 46; 1951, Nucleonics, 8, No.1, 38.

Kortüm, G. and Finkh, B., 1942, Z. Physikal. Chem. B 52, 263.

Krisberg, N.L., Pool, M.L. and Hibdon, C.T., 1948, Phys. Rev. 74, 44.

Kurie, F.N.D., Richardson, J.R. and Paxton, H.C., 1936, Phys. Rev. 49, 368.

Langer, L.M., 1949, Rev. Sci. Inst. 20, 216.

Langer, L.M. and Cook, C.S., 1948, Rev. Sci.19, 257.

Langer, L.M., Motz, J.W. and Price, H.C., 1950, Phys. Rev.77, 798.

Leipunski, A.I., 1936, Proc. Camb. Phil. Soc. 32, 301.

Lewis, W.B., 1942, "Electrical Counting" (Cambridge)

Li, K.T., 1937, Proc. Camb. Phil. Soc. 33, 164.

Libby, W.F. 1934a, Phys. Rev. 45, 845; 1934b, Phys.Rev.46,196.

Libby, W.F., 1939, Phys. Rev. 56, 21.

Libby, W.F. and Lee, D.D., 1939, Phys. Rev. 55, 245.

Longmire, C. and Brown, H., 1949a, Phys. Rev. 75, 264, 1102.

Longmire, C. and Brown, H., 1949b, Phys. Rev.75, 328.

MacGregor, M.H. and Wiedenbeck, M.L., 1952, Phys. Rev.86, 420.

Marinsky, J.A., 1949, PhD. Thesis, Massachussetts Institute of Technology, and, 1949, Prog.Rep.Lab. Nuc. Sci. Eng.No.13,July.

Marshak, R.E., 1949, Phys. Rev. 75, 513.

Mattauch, J., 1937a, Naturwiss. 25, 189; 1937b, Phys.Z.S. 38, 951.

Maze, R., 1946, J. Phys. Radium 7, 164.

Mellor, J.W., 1932, "Comprehensive Treatise of Inorganic and Theoretical Chemistry", Vol.V (Longmans), p.953.

Mellor, J.W, and Thomson, H.V., 1938, "Treatise on Quantitative Inorganic Analysis" (Griffins), p.420.

Millar, L.C. and Curtiss, L.F., 1945, Pu. Proj. Rep. CP-3102 (August).

Millman, S. and Fox, M., 1936, Phys. Rev. 50, 220.

Millman, S. and Kopfermann, H., 1938, Zeits. für Phys. 108,232.

Millman, S. and Kusch, P., 1940, Phys. Rev. 58, 438.

Mitchell, A.C.G., 1948, Rev. Mod. Phys. 20, 296.

Miyamoto, G., 1942, Proc. Phys. Math. Soc. Japan, 24,676; 1943,

Proc. Phys. Math. Soc. Japan 17, 587. (In Japanese, translations available from author).

Montgomery, C.G., and Montgomery, D.D., 1940, Phys. Rev. 57, 1030.

Moon, P.B., 1949, "Artificial Radioactivity" (Cambridge).

Mühlhoff, W., 1930, Ann. Physik. 7, 205.

Mulholland, G.I. and Kohman, T.P., 1952, Phys. Rev. 85, 144.

Nordheim, N.W. and Yost, F.L., 1937, Phys. Rev. 51, 942.

Ollano, Z., 1941, Nuovo Cimento 18, 11.

Parker, G.W., and Lantz, P.M., 1948, AECD-2160.

Parker, G.W., Lantz, P.M., Ruch, J.W. and Hebert, G.M., 1948, Pu. Proj. Rep. ORNL-65, 105.

Persico, E., 1949, Rev. Sci. Inst. 20, 191.

Persico, E. and Geoffrion, C., 1950, Rev. Sci.Inst. 21, 945.

Pomerance, H., 1949, Atomic Energy Commission Declassified Document No.2502 (unpublished).

Pontecorvo B., 1946-1947, Reports on Progress in Physics 11, 32.

Pringsheim, P., 1949, "Fluorescence and Phosphorescence" (Interscience Inc.).

Reitz, J.R., 1950, Phys. Rev. 77, 10.

Rogers, M.M., McReynolds, A.W. and Rogers, F.T., 1940, Phys. Rev.57,379.

Rose, M.E. and Korff, S.A., 1941, Phys. Rev. 59, 850.

Rosenfeld, L., 1948, "Nuclear Forces" (North-Holland Publishing Co., Amsterdam), Vol. 1, p.506.

Ross, M. and Story, J.S., 1948 - 1949, Reports on Progress in Physics 12, 291.

Rossi, B.B. and Staub, H.H., 1949, "Ionization Chambers and Counters" (McGraw-Hill).

Rothwell, P. and West, D., 1950, Proc. Phys. Soc. A 63, 541.

Rutherford, E, 1919, Phil. Mag. 37, 581.

Rutherford, E., Chadwick, J. and Ellis, C.D., 1930, "Radiations from Radioactive Substances" (Cambridge).

Rutherford, E. and Geiger, H., 1908, Proc. Roy. Soc. A 81, 141.

Rutherford, E. and Robinson, H., 1913, Phil. Mag. 26, 717.

Sagane, R., Gardner, W.E. and Hubbard, H.W., 1951, Phys. Rev. 82, 577.

Sagane, R. and Giles, P.C., 1951, Phys. Rev. 81, 153.

Saha, A.K., 1946, Proc. Nat. Inst. Sci. Ind. 12, 159.

Sakai, M., 1950, J.Phys. Soc. Japan 5, 178.

Sargent, B.W., 1933, Proc. Roy. Soc. A 139, 659.

Saxon, D., 1948, Phys. Rev. 74, 849.

Scharff-Goldhaber, G., der Mateosian, E., McKeown, M. and Sunyar, A.W., 1950, Phys. Rev. 78, 325.

Schwartz, H.M., 1952, Phys. Rev. 86, 195.

Seaborg, G.T. and Perlman, I., 1948, Rev. Mod.Phys. 20, 585.

Segre, 1948, Isotope Chart AECD 2404

Senftle, F.E. and Leavitt, W.S., 1950, Nucleonics 6, No.5, 54.

Shull, F.B., 1948, Phys. Rev. 74, 917.

Shull, F.B. and Dennison, D.D., 1947a, Phys. Rev.71,681; 1947b, Phys. Rev. 72, 256.

Shull, F.B. and Feenberg, E.F., 1949, Phys. Rev. 75, 1768.

Siegbahn, K., 1946, Phys. Rev. 70, 127.

Siegbahn, K., 1943, Arkiv. Mat. Astron. Fys. Bd. 30A, No. 1.

Skyrme, T.H.R., 1950, "Progress in Nuclear Physics", Vol.1. (Butterworths), p.115.

Slätis, H. and Siegbahn, K., 1949, Phys. Rev. 75, 1955.

Slätis, H. and Siegbahn, K., 1949, Phys. Rev. 75, 318.

Smith, P.B. and Allen, J.S., 1951, Phys. Rev. 81, 381.

Strassman, F. and Walling, E., 1938, Ber.d.d. Chem. Ges. 71. 1.

Sullivan, W.H., 1949, "Trilinear Chart" (John Wylie and Sons, Inc.)

Svartholm, N. and Siegbahn, K., 1946, Arkiv. Mat. Astron. Fys. Bd.33A, No.4, p.22.

Swartout, J.A., Boyd, G.E., Cameron, A.E., Keim, C.P. and Larson, C.E., 1946, Phys. Rev. 70, 232.

Taylor, H.M. and Mott, N.F., 1932, Proc. Roy. Soc. A 138, 665.

Thomas, D.G. and Kurbatov, J.D., 1950, Phys. Rev. 77, 151.

Thomson, J.J., 1905, Phil. Mag. 10, 584.

Tomozawa, V., Umezawa, M. and Nakamura, S., 1952, Phys. Rev. 86,791.

Tricker, R.A.R., 1924, Proc. Camb. Phil.Soc. 22, 454.

Tyler, A.W., 1939, Phys. Rev. 56, 125.

Walker, R.L. and McDaniell, B.D., 1948, Phys. Rev.74, 315.

Wang, K.C., 1942, Phys. Rev. 61, 97.

Watt, R.J. and Williams, D., 1946, Phys. Rev. 70, 640.

West, H.W., Meyerhof, W.E. and Hofstadter, R., 1951, Phys.81, 141.

Wigner, E.P., 1939, Phys. Rev. 56, 519.

Wilson, H.W.,1950, Phys.Rev. 79, 1032.

Wilson, H.W., 1951, Phys. Rev. 82, 548.

Wilson, H.W. and Curran, S.C., 1949, Phil.Mag.40, 631.

Wilson, H.W. and Curran, S.C., 1951, Phil. Mag. 42, 762.

Wilson, H.W. and Lewis, G.M., 1952, Proc. Phys. Soc.A65, 656.

Winsberg, L., 1950, NNES 9,paper 199.

Witcher, C.M., 1941, Phys. Rev. 60, 32.

Wu, C.S., 1950, Rev. Mod. Phys. 22, 386.

Wu, C.S. and Feldman, L., 1949, Phys. Rev. 76, 693.

Yaffe, L., 1950, "Absolute Beta Counting," Nuclear Science Series, Prelim. Report No. 8.

Zajac, B. and Ross, M.A.S, 1949, Nature, 164, 311.