A STUDY OF THE NATURAL RADIOELEMENTS WITH $Z < 80$.

BY

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PREFACE.

This thesis contains an account of research conducted at the University of Glasgow between October 1953 and July 1956.

The introduction consists of a review of the experimental clarification of β-decay theory and a survey of previous investigations on the radioactivity of certain naturally occurring elements with $Z < 80$.

In Chapter 1 the low background proportional and scintillation counter systems are described. This equipment and the associated techniques were developed by Messrs. D. Dixon and A. McNair, only minor modifications being introduced by Mr. D.E. Watt and the author. The improvement in sensitivity achieved by the use of an underground site is discussed.

Chapter 2 describes the measurement of the branching ratio of K^{40} by two different methods, the results of which are in excellent agreement, and coincidence studies which were undertaken to provide direct evidence in favour of the generally accepted decay scheme and to prove that the discrepancy between the geologically and physically determined values of the branching ratio was not due to a

defective decay scheme. The author assisted Mr. A. McNair in the prosecution of these experiments and was also responsible for the initial draft of the published paper describing them. The attempt to determine the branching ratio by observation of the Auger electrons following K-capture was carried out by the author assisted by Mr. D.E. Watt.

An exhaustive examination of the decay of La^{138} is reported in Chapter 3. Investigation of the γ-ray spectrum confirms the presence of only two of three previously reported γ-rays and coincidence studies conclusively establish an entirely new decay scheme. A previously unobserved 3rd forbidden β-ray spectrum is also detected. The experiments described in this chapter and in Chapters 4 and 5 were conducted jointly by the author and Mr. D.E. Watt but the author was responsible for the analysis and publication of the results.

Chapter 4 details attempts to detect radioactivity in V^{50} which fail to detect β-rays but result in the observation of a very weak gamma activity apparently consistent in energy with a transition, following electron capture in V^{50}, from the first excited state of Ti^{50} to the ground state.

There is no evidence of γ-rays or internal conversion electrons which could be ascribed to an electron capture branch of Lu^{176} (Chapter 5), but the half-life deduced from the γ-ray spectrum is in marked disagreement with the value proposed in a previous independent study of the β-rays.

A summary of the results of unsuccessful searches for activity in three naturally occurring isobaric pairs is given in Chapter 6. A complete description will be contained in Mr. Watt's thesis. In this case the author assisted Mr. Watt with the experimental work.

The problem of obtaining materials free from traces of the naturally radioactive members of the uranium and thorium series is discussed in Chapter 7. Possible methods of increasing the sensitivity of the apparatus sufficiently to render further investigations worthwhile are indicated, but the author considers that any results which might be obtained are unlikely to be of sufficient importance to justify the necessary trouble and expense.

The appendix contains a description of experiments on backscattering into 2π solid angle. The scintillation and proportional counter measurements were carried out by Mr. A. McNair and by the author respectively, the results being analysed jointly.

ACKNOWLEDGMENTS.

The author wishes to thank Professor P.I. Dee for his interest and encouragement throughout the course of this research. The investigations concerned with the measurement of the K^{40} branching ratio and with backscattering were carried out under the supervision of Dr. S.C. Curran. The author records his gratitude to Dr. G.M. Lewis for helpful discussions throughout the period in which the other investigations were undertaken. He is also indebted to Mr. J.T. Lloyd and his staff for technical assistance, particularly photographic; to Mr. R. Irvine and the workshops staff for the construction and modification of proportional counters, and to Mr. T. Pollok, Mr. J. Lindsay and the electronics group for much advice and servicing of equipment.

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CONTENTS.

A STUDY OF THE NATURAL RADIOELEMENTS WITH Z < 80.

INTRODUCTION.

1. THE EXPERIMENTAL CLARIFICATION OF β-DECAY THEORY.

It is not proposed to derive β-decay theory, but merely to present theoretical results and to demonstrate how the experimental evidence, consisting mainly of data on comparative half-lives (ft values) and spectrum shapes, indicates the possible combinations of interactions which can give the correct β-coupling. The material presented here has been drawn from Blatt and Weisskopf's "Theoretical Nuclear Physics" Chap. 13 and from review articles by Konopinski and Langer (1953), and by Rose, Konopinski, Wu and Kofoed-Hansen being Chaps. 9-12 inclusive of Seigbahn's "β and γ-ray spectroscopy" (1955).

As a basic principle one requires that the process of β-decay should obey the laws of conservation of energy, linear momentum, angular momentum, statistics and charge. Simple well-known arguments then lead to the postulate that one very light neutral particle of spin $\frac{1}{2}$, christened the neutrino by Pauli, is emitted in every β-decay process.

The theory actually assumes zero rest mass for the neutrino. The requirements of relativistic invariance lead to five possible types of interaction namely scalar (S), vector (V), tensor (T), axial vector (A) and pseudoscalar (P). The S and V types are called Fermi interactions since these were the types considered in Fermi's original formulation of the theory. The T and A types are Gamow - Teller (GT) interactions so named after the workers who first studied them. The complete β-interaction is a linear combination of these five types such that $H = \sum C_X H_X$ where X = S, V, T, A and P in turn, C_X is a coupling constant and H_X the interaction energy density for an interaction type X.

In the study of allowed β-decay only the non-relativistic limit of the nucleon wave functions is necessary, so that the treatment is relativistic with respect to the light particles only. These can be represented by a plane wave of the form $\exp(i \underset{\sim}{k}.\underset{\sim}{r})$, neglecting the Coulomb effect for electrons or positrons. This exponential can be expanded in powers of the exponent corresponding physically to expansion in waves of increasingly higher angular momentum. The major contribution arises from the first term in each expansion. Hence $\exp(i \underset{\sim}{k}.\underset{\sim}{r})$ can to a first approximation be replaced by unity, which means physically

that zero orbital angular momentum is carried off by the electron-neutrino pair. Obviously transitions with the largest decay constant will be those for which the matrix elements do not vanish when the wave functions are thus replaced by their long wavelength limit. These are referred to as the allowed transitions and are expected to show the shortest comparative half-lives. Inspection of the matrix elements leads to the selection rules for allowed transitions.

S, V $\quad \Delta J = 0$; no

T, A $\quad \Delta J = 0, \pm 1$, except $0 \rightarrow 0$; no

where 'no' indicates that the parity does not change. That the presence of the GT interaction is necessary is shown by the existence of $\Delta J = \pm 1$, no transitions e.g. $He^6 \rightarrow Li^6$, which are not allowed by Fermi selection rules. On the other hand the two β^+ decays $O^{14} \rightarrow N^{14*}$ and $C^{10} \rightarrow B^{10**}$ are known to undergo $0 \rightarrow 0$, no transitions which are strictly forbidden by GT selection rules. Hence both Fermi and GT interactions are necessary and β-decay therefore cannot be explained by one particular interaction alone, but only by a linear combination. The P interaction requires a parity change and hence its matrix elements cannot coexist

with those of the other interactions. Furthermore its selection rule permits only $\Delta J = 0$ transitions. The theoretical comparative half-lives are also much longer than the shortest which have been experimentally observed. Therefore the P interaction does not give allowed transitions.

The energy distribution of the emitted β particles is given in its most general form by

$$N(W)dW = \frac{g^2}{2\pi^3} F(\mp Z, W) pW(W_0 - W)^2 \left[(C_S^2 + C_V^2)|M_F|^2 + (C_T^2 + C_A^2)|M_{GT}|^2 \right] \left(1 \mp \frac{b}{W}\right) dW$$

where N(W) is the number of particles with total energy between W and W + dW;

g is the coupling constant indicating the strength of the β-decay interaction;

$F(\mp Z, W)$ is the Fermi function representing the effect of the Coulomb field;

p and W_0 are the momentum and maximum energy of the emitted β particle respectively;

C_X is the coupling constant for an interaction type X;

M_F and M_{GT} are the matrix elements for Fermi and GT interactions respectively;

b/w is the Fierz interference term.

It follows that, if b/w is zero, the energy distribution of the β-particles for allowed transitions is given by

$$\left(\frac{N(W)}{pWF}\right)^{1/2} = K(W_o - W)$$

where K is a constant independent of energy.
Hence a plot of $\left(N(W)/pWF\right)^{1/2}$ against the energy W, called a Kurie (or Fermi) plot should be linear, intersecting the energy axis at W_o the maximum energy of the β-particle. If the interference term exists, which is possible only if the β-interaction involves both the S and V types or both T and A, since the numerator of the constant 'b' contains only terms involving the products $C_S C_V$ and $C_T C_A$, then the Kurie plot should not be linear. Careful measurement of the spectrum shape for many allowed transitions confirms the linearity of the Kurie plot thus showing that the Fierz interference term b/w must be very small, if it exists, and hence that $C_S C_V \simeq 0$ and $C_T C_A \simeq 0$. Several evaluations of the best available experimental data indicate that $C_{A,T}^2 / C_{T,A}^2 \leq 4\%$ and $C_{V,S}^2 / C_{S,V}^2 \leq 4\%$. Pohm et al.(1956) examined the allowed P^{32} spectrum closely for Fierz deviation

and reported that $C_{A,T}^2 / C_{I,A}^2 \leq 0.02\%$. A theory containing such small components in a fundamental law of interaction is hardly acceptable, and the conclusion is that C_A or $C_T = 0$ and C_V or $C_S = 0$ i.e. that the fundamental law must contain either the S or V interaction, but not both, and either the T or A, but not both.

The decay constant for a β-transition is given by

$$\lambda = \int_1^{W_0} N(W)\,dW = \frac{g^2}{2\pi^3}\left[C_F^2 |M_F|^2 + C_{GT}^2 |M_{GT}|^2\right] f(Z, W_0)$$

where

$$C_F^2 = C_S^2 + C_V^2 \; ; \; C_{GT}^2 = C_T^2 + C_A^2$$

and $$f(Z, W_0) = \int_1^{W_0} F(Z, W)\, p W (W_0 - W)^2\, dW$$

Numerical values of $f(Z, W_0)$ have been extensively tabulated. The half-life $t = \log_e 2 / \lambda$ is not itself characteristic of a transition since t is strongly dependent on W_0. However, theoretically the matrix elements M_F and M_{GT} should be of the same order of magnitude for all allowed transitions. The characteristic quantity is therefore the comparative half-life ft.

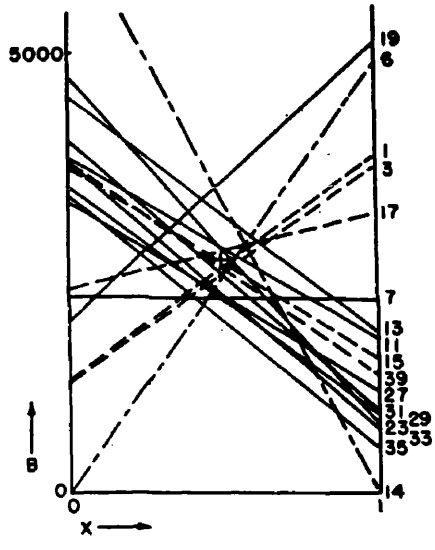

Fig.1. B-x lines for mirror transitions and for He^6 and O^{14}. Mass numbers are indicated.

----- closed shell ± 1 transitions

_____ other mirror transitions

_.-._. He^6 and O^{14} decay

$$\therefore ft = \frac{B}{(1-x)|M_F|^2 + x|M_{GT}|^2}$$

where $x = C_{GT}^2 = 1 - C_F^2$ and $B = 2\pi^3 \hbar g_v^2 / g^2 = 43/g^2$

If the matrix elements are known accurately then the experimentally measured ft value determines a straight line in a diagram of B against x. There will be a separate line for each allowed transition, and all these lines should intersect at a single point, giving the values of x, the relative strength of GT to Fermi interactions, and B (hence g). The matrix elements are most reliably evaluated for mirror transitions (transitions for which N - Z = $\pm$ 1 for both parent and daughter nucleus) especially for those with closed shells in both neutrons and protons $\pm$ one nucleon. The intersection points exhibit a spread (Fig.1), but cluster around $x \simeq 0.5$. Thus the Fermi and GT interactions are of approximately equal strength. Independent analyses by Kofoed-Hansen and Winther (1952 a,b; 1953), Trigg (1952) and Bouchez and Nataf (1952 a,b; 1953) all arrived at this conclusion.

One extremely important experiment remains to be mentioned. The β^--decay of He^6 must be a pure GT interaction since $\Delta J = -1$. Rustad and Ruby (1953) have examined the angular correlation between the β-ray and the recoil ion. In their instrument the angular correlation could be measured for different intervals of β-ray energy, or the energy distribution of the β-rays could be found for a certain angular interval. The experimental results were compared with the theoretical expectations for T and A interactions corrected for the finite resolution of the instrument, and were found to be strongly in favour of the tensor (T) interaction. This experiment provides the only evidence in favour of the T rather than A interaction.

To summarise, experimental evidence shows that both Fermi and GT interactions are required to account for allowed transitions, and that they are of approximately equal strength. Hence the complete β-interaction cannot be given by any single type, but must be a linear combination. The observed linearity of Kurie plots indicates that not both the S and V nor both the T and A interactions can be present. The He^6 angular correlation experiment shows that the GT interaction is given by the tensor form. Thus study of the allowed transitions alone reduces the fundamental law of

interaction to either an ST(P) or a VT(P) combination.

An important deduction can be made from the study of the spectrum shape of the allowed transition $H^3 \rightarrow He^3$. The precise shape in the vicinity of the end-point depends on the rest mass of the neutrino. Careful measurement of the H^3 β-spectrum shows that the mass must be < 1 kev. The theoretical assumption of zero rest mass is thus justified.

In the theory of forbidden β-decay the small effects neglected in allowed theory must be considered. Firstly, the interaction energy densities for the V, T and A forms must be modified to produce invariance to a Lorentz transformation i.e. the nucleons are now treated relativistically. Secondly, effects arising from the variation of the electron and neutrino waves across the nucleus must be considered. These were ignored when only the first term of the expansion of $\exp(i\, \underset{\sim}{k}.\underset{\sim}{r})$ was retained, but this term will now disappear since the allowed selection rules are violated.

Theory leads to the following general selection rules for an 'n'-th forbidden transition.

$$\Delta J = n, n + 1 \qquad \text{where } n \neq 1$$

The parity change, given by $(-1)^n$, alternates with successive

degrees of forbiddenness.

$$\Delta J = 0, \pm 1, \pm 2; \quad \text{yes} \qquad \text{where } n = 1$$

Pseudoscalar coupling gives only negligible contributions in all but once forbidden transitions. This peculiarity is due to the fact that its general 'n'-th degree matrix element vanishes unless $\Delta J = n-1$. It is then a negligible correction to the GT contribution which is, by comparison, only (n-2)-times forbidden if $n > 1$. The singularity of the $n = 1$ case is because no (n-2)-times forbidden transitions exist for $n = 1$ i.e. no transitions to overwhelm the $\Delta J = n - 1 = 0$ contribution. Hence one can hope to detect the presence of the P interaction only in once forbidden transitions with $\Delta J = 0$.

The transitions with $\Delta J = n + 1$ are called the unique forbidden transitions since they arise only from GT coupling, in fact from the T interaction alone since the A interaction has already been ruled out. Since only one nuclear matrix element is involved, the shape factor, which gives the deviation from allowed shape, can be calculated unambiguously. Many examples of unique first forbidden transitions ($\Delta J = 2$, yes) whose Kurie plots are linearised by the unique first forbidden shape factor have now been

observed. Only one case is known for each of the unique 2nd and 3rd forbidden transitions. These are Be^{10} ($\Delta J = 3$, no) and the natural source K^{40} ($\Delta J = 4$, yes). The mere existence of the unique transitions is the second important piece of evidence for GT coupling (the first being the allowed spectra with $\Delta J = \pm 1$). The excellent agreement between theory and experiment convincingly confirms that the present theoretical approach to β-decay is correct.

The first forbidden transitions are now studied to differentiate between the linear combinations ST and VT, and to investigate the contribution of P coupling. The selection rule is $\Delta J = 0, \pm 1$; yes. The first forbidden shape factors for each type of interaction contain two terms one of which is energy dependent, the other, the Coulomb term, not. The energy dependent term is usually swamped by the Coulomb term so that, if one considers only a single interaction at a time, the first forbidden transitions would be expected to yield an allowed spectrum shape and hence a linear Kurie plot. However since the evidence from the study of allowed transitions dictates the adoption of a linear combination of interactions, a Fierz-type interference term can exist, just as for allowed transitions. Experimental investigation has shown that, with the exception

of RaE the Kurie plots are linear. Now considering only the two remaining possible linear combinations, VT leads to an energy dependent shape factor, ST does not. Analysis of first forbidden Kurie plots for a deviation from linearity shows that $C_V^2/C_T^2 \lesssim 1\%$, whereas from the study of allowed spectra it is known that the Fermi and GT interactions are of approximately equal strength ($C_{S,V}^2 = C_T^2$). This is the only evidence that the Fermi interaction is scalar (S) rather than vector (V).

The shapes of the 2nd forbidden spectra so far investigated are definitely not allowed, but these transitions provide no clear-cut evidence because of the number of independent parameters contained in the shape factors of the various linear combinations.

Lastly the contribution of the pseudoscalar (P) interaction must be considered. Formerly the only direct evidence for its existence was the shape of the RaE spectrum, which could only be fitted by a TP combination for a $\Delta J = 0$ transition with its large energy independent terms cancelling out. However the spin of RaE has recently been measured to be unity (Smith 1955), so that the transition is definitely $\Delta J = 1$. The validity of the power series expansion of $\exp(i\, \underset{\sim}{k}.\underset{\sim}{r})$ depends on the fact that the electron de Broglie

wavelength, k, is large compared to the nuclear radius, r. When k and Z are large, this approximation is insufficient. It has been shown that if the electron radial wave functions are calculated accurately then the ST combination for a $\Delta J = 1$, yes transition fits the observed spectrum (Rose and Perry 1953; Rose et al. 1953). Thus there is now no direct evidence favouring the presence of P coupling. An apparent difference in the ft values of first forbidden transitions with $\Delta J = 0$ and $\Delta J = 1$ has been suggested to be partly due to a substantial contribution from the P interaction for $\Delta J = 0$, and thus to be indirect evidence of its existence (King and Peaslee 1954). However for the P contribution to be comparable with that from the other interactions, C_P/C_T must be large (~ 50), or the presence of pseudoscalar coupled forces in the nucleus must be assumed.

Now the shape of allowed spectra will be affected if a large amount of P interaction is included, the distortion being more pronounced for β-transitions of large maximum energy. The theoretical spectrum shape is independent of the amount of P coupling which might exist in nuclear forces so that an upper limit of C_P/C_T i.e. of the contribution of the P interaction to β-decay can be deduced by comparison of the theoretical and observed spectrum shapes.

Experimental data on He^6 and B^{12} is as yet not accurate enough to give more than a rough limit, but seems to suggest that a large amount of P interaction is not present. Thus at the moment the role of the P interaction is very uncertain.

In short, it can be seen that the experimental data has conclusively established the validity of the theoretical approach to β-decay, and has determined that the fundamental law is given by a linear combination consisting of equal amounts of the scalar and tensor interactions, with the pseudoscalar neither definitely included or excluded by the present experimental data.

2. β-DECAY ENERGETICS.

This subject has been reviewed at length by Coryell (1953). The energetics of the processes of β^- and β^+ emission and electron capture are given by the following relationships between the atomic masses of the parent and daughter nuclides where M(Z, A) denotes the atomic mass of the parent nuclide with atomic number Z and mass number A. The neutrino rest mass is taken to be zero.

$$E_{\beta^-} = \left[M(Z,A) - M(Z+1,A)\right]c^2$$

$$E_{\beta^+} = \left[M(Z,A) - M(Z-1,A) - 2m_0\right]c^2$$

$$E_{EC} = \left[M(Z,A) - M(Z-1,A)\right]c^2 \geq E_{K,L\ldots}$$

where E_K is the binding energy of the K electron.
If the available energy is such that K - capture is not energetically possible, then if the mass difference is greater than the binding energy of the L - electron, L - capture will occur.

From consideration of the energy relationships for β^--decay and electron capture, it follows that, when two adjacent isobars exist, one must decay to the other.

It can be deduced, using the semi-empirical mass formula, that no odd-odd nucleus should be stable against β^- decay. For the very light nuclei (H_1^2, Li_3^6, B_5^{10} and N_7^{14}) the condition Z = N overrides the instability caused by the existence of both an unpaired neutron and an unpaired proton. These nuclei are, in any case, too light for the semi-empirical

mass formula to be strictly valid. Similarly no odd-odd nucleus is expected to be stable against electron capture. In contrast, even-even nuclei are expected to be extremely stable.

Suess and Jensen (1951) first correlated the total decay energy, i.e. the energy associated with $\beta^{\pm}$-emission or electron capture together with the energy of any γ-rays in cascade with whichever of these processes occurs, with nuclear composition. Way and Wood (1954) plotted the total decay energy against the number of neutrons in the parent nucleus. A linear relationship, which is implicit in the semi-empirical mass formula, was shown to exist for the decay energies of disintegrating atoms of the same Z. A striking feature was the sudden decrease in neutron and proton binding energies just after the magic numbers given by the nuclear shell model i.e. just after completion of a shell. From a practical point of view, these graphs allow the energy available in an unknown decay to be crudely estimated.

3. THE NATURAL RADIOELEMENTS WITH $Z < 80$.

The significance of these energetics as regards the natural radioelements with $Z < 80$ will now be indicated, and

the importance of the study of these elements will be discussed.

Since it has just been shown that two adjacent isobars cannot both be stable, it follows that one member of each of the following naturally occurring isobaric pairs should be radioactive:- Rb^{87}_{37}, Sr^{87}_{38}; Cd^{113}_{48}, In^{113}_{49}; In^{115}_{49}, Sn^{115}_{50}; Sb^{123}_{51}, Te^{123}_{52}; and Re^{187}_{75}, Os^{187}_{76}. Only one isotope Rb^{87} has definitely been identified as radioactive and thoroughly investigated. Re^{187} was found to emit low energy β-rays by Naldrett and Libby (1948), Sugarman and Richter (1948) and Suttle and Libby (1954), the same apparatus being used in all three cases. The end-point was found by aluminium absorption methods but, although the intensity observed in each case was the same, the maximum energy quoted by Suttle and Libby (1954) was a factor of five smaller than that given in the two previous investigations. In our own experience, commercial aluminium is extremely impure compared with the purity required in low activity investigations. Experiments by Dixon and McNair (1954) failed to detect this reported activity. In^{115} has been reported to decay by β^--emission by Martell and Libby (1950) and Cohen (1951). The former attempted a measurement of

the maximum β-energy. In view of the large discrepancies between the maximum energies reported by Libby and co-workers for several activities and the now accepted values, their method of energy measurement is suspect. Cohen (1951) merely reported that some activity had been observed. The spectrum shape has never been investigated. The radioactive members of the pairs Cd^{113}, In^{113} and Sb^{123}, Te^{123} have not been identified. The systematics of Way and Wood (1954) indicate that an energy of 250 - 350 kev is available for β^--decay of Cd^{113} and the same amount for electron capture by Te^{123}. The energy available for all these transitions is low, as is to be expected since whether the odd nucleon is a proton or a neutron makes little difference to the binding energy of an odd-A nucleus.

Four naturally occurring isobaric triplets are well known viz. A^{40}_{18}, K^{40}_{19}, Ca^{40}_{20}; Ti^{50}_{22}, V^{50}_{23}, Cr^{50}_{24}; Ba^{138}_{56}, La^{138}_{57}, Ce^{138}_{58} and Yb^{176}_{70}, Lu^{176}_{71}, Hf^{176}_{72}. In every case the middle member is an odd-odd nucleus. Since these nuclei have been shown to be inherently unstable, and also since neighbouring isobars cannot both be stable, the middle member is expected to decay both by electron capture (or β^+-emission) and β^--emission. The radioactivity of K^{40} is well established

and has been the subject of many investigations, but there is considerable doubt as to the correct value of the branching ratio. The β^--decay of Lu^{176} has been conclusively demonstrated but there is no evidence for the existence of the electron capture branch. Gamma activity ascribed to the electron capture branch and β^--decay have been reported for La^{138} but the results are not conclusive and the decay scheme proposed has been contradicted by coincidence studies on the 33 min. β^--activity of Cs^{138}. No activity has been reported for V^{50}. Recently a new isotope of tantalum Ta^{180} was discovered by White et al.(1955). Its existence creates a fifth isobaric triplet Hf^{180}_{72}, Ta^{180}_{73}, W^{180}_{74}. The energy available for all the natural odd-odd decays is high, being >1 Mev in all the transitions so far established. This is to be expected because of the existence of an unpaired proton and an unpaired neutron so that the decay produces either a pair of neutrons or a pair of protons, in both cases the daughter nucleus being even-even and therefore of much greater stability.

It can thus be seen that relatively little is known about the natural radioelements with $Z < 80$. It only remains to answer the query "Are they worth further investigation?"

Obviously the mere detection of their radioactivity confirms the theoretical predictions, if any confirmation were needed. However since the half-lives of these natural radioelements are very long ($> 10^9$ yrs) even when there is considerable energy available, the transitions must involve a large spin change. The spin changes are 3, 4, 4, 3 and 2 respectively for the isobaric pairs in the order listed earlier. The spins of the odd-odd nuclei are 4 for K^{40}, 6 for V^{50}, 5 for La^{138} and 10 for Lu^{176}. Thus the prime reason originally was to provide experimental evidence on highly forbidden β-decay. For example, the K^{40} β^- spectrum has been shown by several investigators to have a forbidden shape such that the Kurie plot is linearised by the 3rd forbidden unique correction factor. This spectrum provides one of the arguments for the existence of GT coupling. Research done at Glasgow by Curran et al.(1952) using a large proportional counter and Lewis (1952) by scintillation methods using a RbI crystal first demonstrated the unusual shape of the Rb^{87} β^--spectrum. This is the only $\Delta I = 3$, yes transition known. Tomozawa et al.(1952) have shown that both the 3rd forbidden V and T correction factors linearise the Kurie plot. In view of the number of adjustable parameters, there should be no difficulty in fitting an ST combination to the

observed spectrum. In addition, at Glasgow, Dixon et al. (1954) investigated the β^--spectrum of Lu^{176} and were able to determine the maximum energy but not the shape, since the range of the higher energy β-rays in the counter at the pressure then attainable was such that many would either hit the wall or pass out of the counting volume into the "dead space" at the ends, and thus give pulses which were too small.

Furthermore, since odd-odd nuclei have a very high spin and the available decay energy is large, the daughter is often former in an excited state with the subsequent emission of γ-radiation. Thus the study of the natural radioelements also provides information about the excited states of nuclei. In particular, following the β-decay of Lu^{176}, Hf^{176} is formed in a highly excited state, and is de-excited by the emission of three γ-rays in cascade. The energies of these excited states are found to be in excellent agreement with those predicted by the Bohr-Mottelson theory (1953), and this result has been extensively used as support for the theory.

From a more practical and less academic viewpoint, measurement of the amount of A^{40} or Sr^{87} present in certain minerals due to the decay of K^{40} or Rb^{87} enables the age of

the mineral to be determined. These methods are of great importance to geologists, and it is possible that more of the isobaric pairs or triplets listed might be useful in this respect.

The above evidence clearly shows that much important information has been derived from the study of the natural radioelements with $Z < 80$, and, in the opinion of the author, more than justifies further investigation.

CHAPTER ONE

THE APPARATUS FOR THE DETECTION OF WEAK ACTIVITIES.

The proportional counter is well adapted to such investigations as the decay of the naturally occurring radioelements. Sources of large area can be mounted internally, an important consideration where very feeble activities are concerned. This technique was first applied to the Geiger counter by Libby (1934). However the proportional counter has a great advantage over the Geiger counter in that it is both detector and spectrometer. Because of this latter property the effective background may be considerably less than the total background. For example, homogeneous radiation would be concentrated in the energy spectrum relative to the background which would be spread out in a smoothe continuous manner. The same argument applies, but less effectively so, to sources of low energy β-rays since only the background in a limited energy range need be considered. The energy range over which the counter can be used as a β-ray spectrometer is limited by the fact that the β-rays must be totally absorbed within the sensitive volume. In the present studies, when the

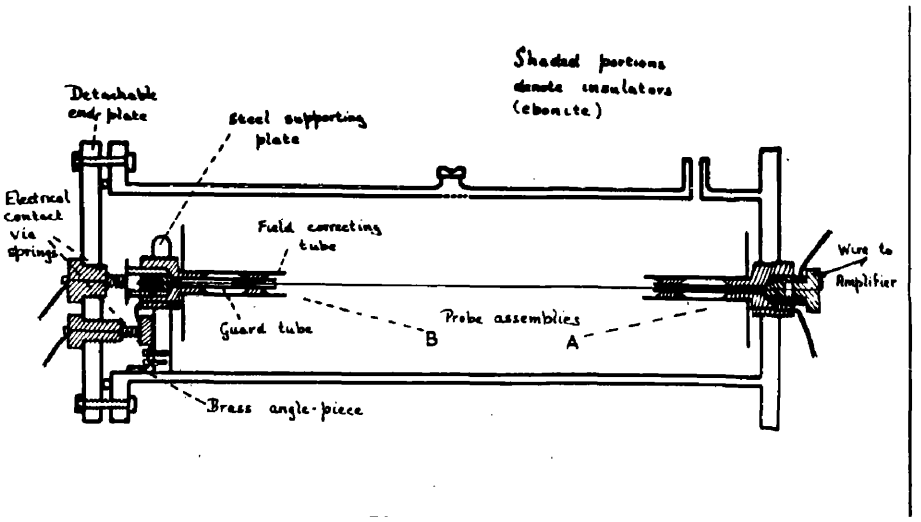

Fig.2. Diagram of the proportional counter.

counter was used at a pressure of 5 atm. (argon-methane mixture), β-spectra of maximum energies less than 350 kev could be obtained undistorted. The end-points of spectra of energies up to 700 kev could be determined, but these spectra would be distorted due to β-rays either penetrating into the insensitive volume at each end of the counter or hitting the cylindrical wall. However, from the point of view of the present studies, this limitation was of no importance.

The counter used by Dixon and McNair has been modified considerably. It was desirable that the linings on which the sources were mounted could be inserted and removed without breaking the central wire. To achieve this, Dixon and McNair brought the field and guard-tube assembly connections out through the side of the counter about 10cm from one end. Now the connections are made via the end-plate (see fig.2), a system first utilised by McNair et al. (1955, 1956) in another counter used for experiments in connection with the measurement of the K^{40} branching ratio. This assembly, apart from being much more robust, was also at the end of the counter so that the counting length was altered from 24.8 to 34cm. The maximum source area was thus increased from 1100cm^2 to 1450cm^2, an increase of

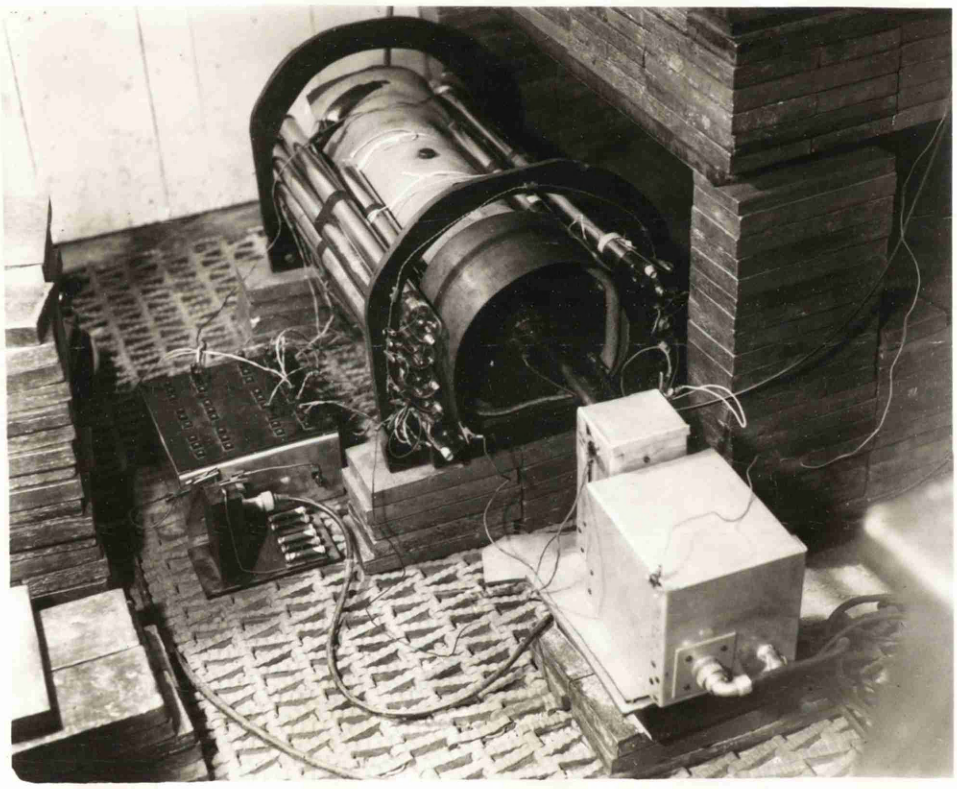

Fig.3. View of the proportional counter in the mine showing Geiger rings partly assembled.

roughly 30%. Also the ebonite insulators were fitted with an O-ring seal inside the counter instead of being waxed in, and were made to screw from the inside of the end-plate outwards rather than from the outside inwards, so that high pressure sealed the plug tighter instead of tending to force it out and so cause leaks rather more frequently than desirable.

Obviously successful detection of low activities required, as well as maximum source area, that the background be reduced to a minimum, since its statistical nature would tend to obscure the presence of an additional very weak activity. The arrangement used, essentially the same as that of Dixon and McNair, consisted of the proportional counter surrounded by two rings of Geiger counters operated in anticoincidence with it, and enclosed in a shield of 4in lead lined with 1in iron (Fig.3). The amplified pulses from the proportional counter were applied to the X-plates of a cathode ray tube. The output pulses from the Geiger counters, all connected in parallel, were amplified, shaped and fed to the Y-plates. When coincident events occurred in the two systems, the Geiger pulse deflected the spot vertically on the screen to a position behind an opaque covering which hid the coincident proportional counter pulse

from the camera. When an event occurred only in the proportional counter, the spot was deflected across the screen a distance proportional to the size of the applied pulse and recorded by the camera. Instead of turning the camera by hand, the experiments were made much less tedious by driving it with a geared-down electric motor. Pulse height analysis was subsequently carried out visually in a microfilm reader. During the experiments of Dixon and McNair the equipment was sited in an alcove surrounded by 10 ft. of concrete in the beam room of the 300 Mev synchrotron about 10 ft. below ground level. However when this machine came into operation the site was hardly suitable for low activity work. Unfortunately on moving to a room at ground level the background was found to have risen considerably. In an attempt to improve even on the beam room background the complete equipment including the $3\frac{1}{2}$ ton lead shield was shifted to a specially constructed laboratory about 90 ft. underground in a disused coal-mine beneath this University. Measurements with an unshielded small Geiger counter indicated that the particle component of the cosmic ray flux was reduced by a factor of three compared with ground level. With the proportional counter under the shielding the background (120 cpm) was improved by the same factor

relative to that in the beam room. However when the anticoincidence Geiger counters were switched on the background was reduced only to 28 cpm, practically the same as that in the beam room. Using a scintillation spectrometer, considerable γ-activity was detected and was traced to radioactive materials, mainly K^{40}, in the surrounding rock and brickwork. Therefore the very disappointing reduction was due to the fact that the proportional counter was now detecting γ-rays which could not be completely cut out by the available shielding. There is no doubt that the background would be considerably improved if the thickness of the lead shield was doubled as will be seen from the following paragraph. Thus the sensitivity of the apparatus is roughly equivalent to that achieved by Dixon and McNair (1954), Mulholland and Kohman (1952a), Kulp and Tryon (1952) and by the modified screen-wall Geiger counter first described by Libby (1934).

For the detection of weak γ-activities a scintillation spectrometer consisting of a 2 in. high x $1\frac{3}{4}$ in. diam. NaI crystal mounted on a Dumont 6292 photomultiplier was used. The pulses were amplified, fed through a cathode follower into a cable 400 ft. long, and analysed at the surface by means of a 100-channel Hutchinson-Scarrott kicksorter.

The available lead was rearranged to give the spectrometer an 8 in. lead shield on all sides. The background above 70 kev was 80 cpm compared with 250 cpm at ground level. This result indicates that the 8 in. shield was sufficient to cut out the γ-ray contamination from the surroundings. This spectrometer was considerably more sensitive than any reported before the present investigations were undertaken. Most of the background is believed to be due firstly to impurities in the aluminium can and secondly to the presence of potassium in the photomultiplier and in the NaI crystal. It has been reported (Miller et al. 1956) that mounting a NaI crystal size 4 x 1½ in. in a copper or steel can reduced the shielded background in the range 0.1-2.5 Mev from 360 cpm to 150 cpm. This spectrometer is of comparable sensitivity to that already described which, however, could now be further improved by recanning the NaI crystal.

Disadvantages of working underground, apart from the dimensions of the room (length 10½ ft., breadth 6½ ft. and height 5ft.) soon became apparent. To prevent dampness the room had to be heated continuously. In the event of an electronic breakdown test equipment had to be transported a distance of over 200 yd. A more troublesome feature was that the large and heavy proportional counter

had to be filled at the surface and then carried about 150 yd. to the underground site. This was necessary because, when the counter was filled in the mine, it was found that it was being contaminated with a fine dust which was feebly radioactive producing an α-rate of 6 cpm immediately after filling and 3 cpm after the dust had time to settle on the walls, thus giving 2π geometry. Hence the counter was never opened underground and care was taken to keep it and the interior of the lead castle scrupulously clean. The α-activity in the counter was also checked regularly. However the site did ensure comparative freedom from electrical or mechanical disturbances.

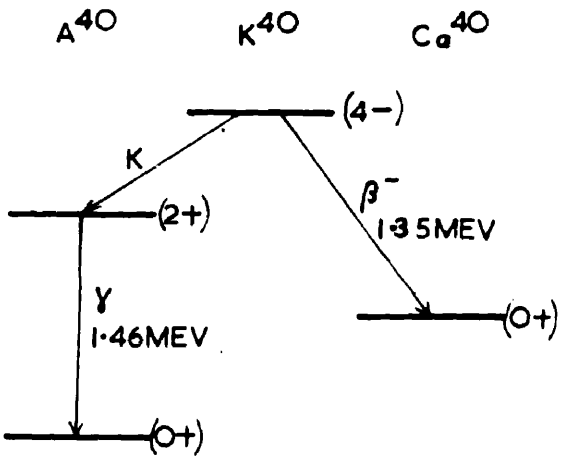

Fig.4. Decay scheme of K^{40}.

CHAPTER TWO

THE BRANCHING RATIO OF K^{40}.

1. INTRODUCTION.

Considerable interest has been shown in the decay of K^{40}, mainly because of its importance in geological dating. The earlier investigations are of historical interest only, being severely hampered by lack of suitable experimental techniques. Within the last ten years, the study of potassium has resulted in an accurate knowledge of the γ-ray energy, the maximum energy of the β-ray and the shape of the β-spectrum. Evidence, which will be discussed later, has been accumulated supporting the decay scheme (Fig.4) first proposed in a slightly modified form by Suess (1948). However a wide range of values has been reported for the branching ratio, defined here as the ratio of the gamma or electron capture transition rate to the beta transition rate. This ratio may be determined by two methods 1) direct counting methods and 2) by measurement of the amount of argon or calcium or both in a potassium-bearing mineral of known geological age. The results obtained by the first

method vary between 0.05 and 0.127. Examination of the experimental techniques used shows that many investigations must be of very limited accuracy. Furthermore most of the results are not independent since only the γ or β-activity was measured and the results of the others relied on to derive the ratio. Only the investigations by Sawyer and Wiedenbeck (1949, 1950) and Houtermans et al. (1950) seem at all reliable. These indicate a branching ratio ~ 0.12, the generally accepted value. However the most recent results obtained by the geological method tend to a value of 0.09 (Shillibeer and Russell 1954, Wasserburg and Hayden 1954) although a value of 0.12 was reported by Inghram et al. (1950). The geological method will be discussed later in more detail in the light of our own results and those of other investigations which have become available since our work was completed. A discrepancy obviously exists between the geological and physical measurements of the branching ratio, and in view of the importance of this ratio in the age determination of potassium-bearing minerals, an accurate measurement by direct counting methods was felt necessary.

2. MEASUREMENT OF THE BRANCHING RATIO.

Two methods were employed to measure the branching ratio, in both of which it was assumed that the decay scheme of Fig.4 was correct and that the ratio of the absolute rate of emission of γ-rays to that of β-rays yielded the branching ratio.

FIRST METHOD.

A. EXPERIMENTAL DETAILS.

In this method difficult and uncertain calculation of the γ-detector efficiency was avoided, to a first order, by comparing the known γ/β ratio of Co^{60} with that of K^{40}. Co^{60} emits a β-ray of maximum energy 300 kev followed by two γ-rays in cascade, the energies of which (1.17 and 1.33 Mev) are fairly close to the K^{40} γ-ray energy (1.46 Mev). Had the γ-ray energies, the maximum β-ray energies and the β-spectrum shapes been identical, it is easy to show that the K^{40} branching ratio, R say, would have been given by

$$R = 2 \frac{\gamma_K}{\gamma_{Co}} \frac{\beta_{Co}}{\beta_K}$$

where γ_K and β_K are the observed gamma and beta counting rates of a potassium source and γ_{Co} and β_{Co} the corresponding rates for a Co^{60} source, determined under the same conditions. As the γ and β-spectra were not identical, various corrections, detailed later, had to be applied.

The γ-detector consisted of a NaI crytal, 2 in. long x $\frac{3}{4}$ in. diam., which was surrounded by sufficient absorber to absorb all β-rays, mounted on an EMI Type 6262 photomultiplier. The crystal was surrounded by a cylindrical vessel of capacity 186 cc. which was filled to a fixed mark with the solutions to be measured. Hence the geometrical conditions were identical for all sources. To ensure maximum operational stability of the apparatus, arrangements were made to empty, rinse and fill the vessel without in any way altering the position of the crystal on the face of the photomultiplier or exposing the tube to light. Between experiments the vessel was thoroughly rinsed with acid, distilled water and acetone in that order. Air was then blown through to remove moisture completely and thus ensure that the correct volume of source was used each time. Background measurements were made between each experiment to check the efficiency of the rinsing. No difficulty was encountered in removing every trace even of Co^{60} solution

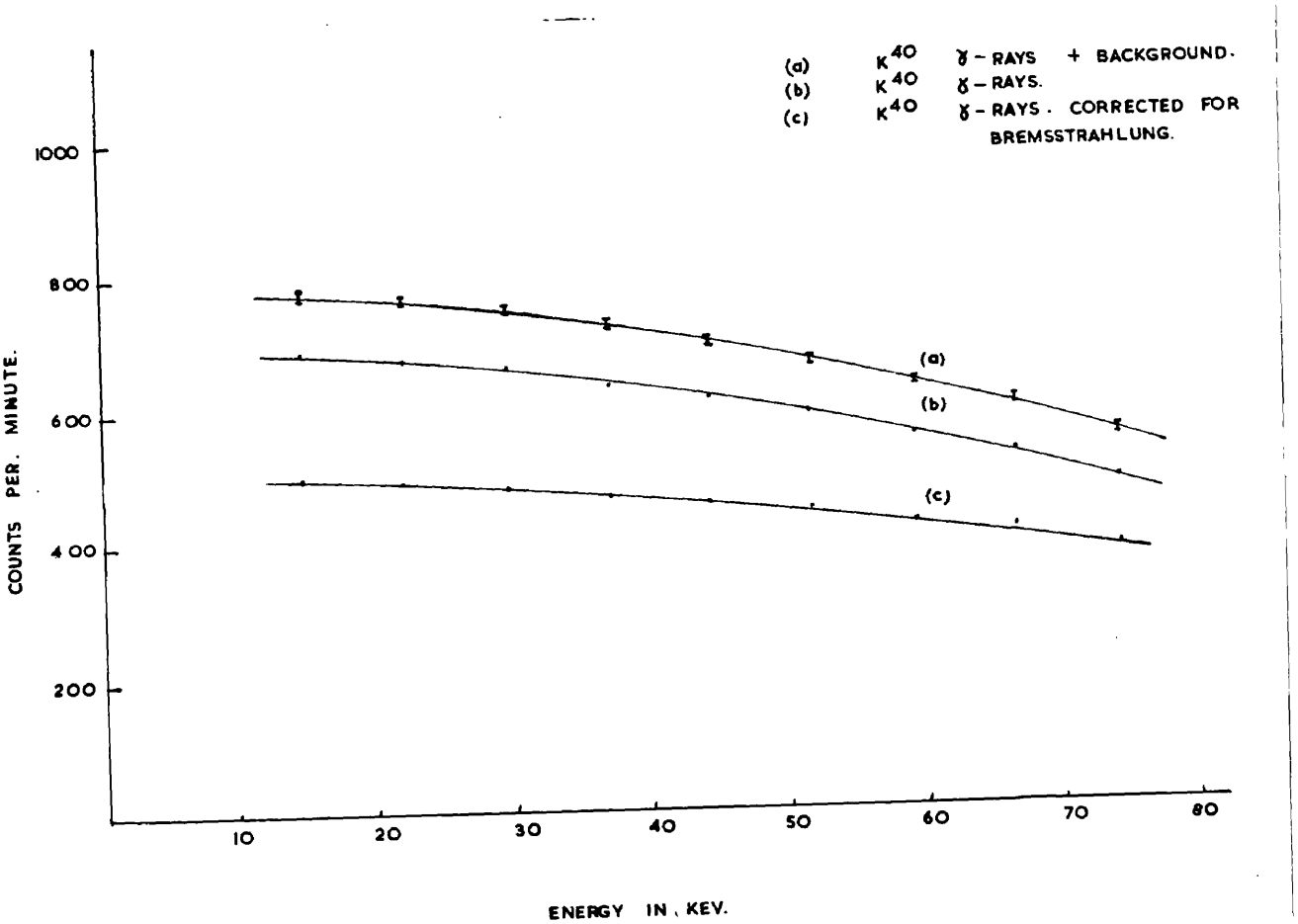

Fig.5: Discriminator bias curves for K^{40} γ-rays.

from the vessel. The solutions used were water containing a known aliquot of a standard $Co^{60}Cl_2$ solution and an aqueous solution of potassium hydroxide (493 gm K/litre) giving 700 cpm above the background of 87 cpm, which was attained by surrounding the apparatus with 4 in. lead. It was found that the background with water, carbon tetrachloride, or aqueous solutions of calcium chloride or sodium chloride in the vessel was slightly higher than when it was empty, the actual rate being independent, within experimental error, of which liquid was employed. Hence the background rate was always checked with distilled water in the vessel. The output pulses from the photomultiplier after amplification were passed through a discriminator and recorded by a scaler. Integral bias curves in the range 10-100 kev were obtained for the test solutions, calibration being effected by means of the 46.7 kev γ-ray of RaD. The results for potassium before and after subtraction of the background are shown in Figs. 5(a) and 5(b).

The β-detector was a cylindrical end-corrected proportional counter filled to a pressure of 1 atm. with the usual argon-methane mixture. The methods of reducing the background described earlier were used, except that these experiments were carried out at ground level. Accurately

known volumes of the two solutions were spread over an area of 700 cm^2 on thick stainless steel sheets. This large source area, combined with the low background (45 cpm) ensured reasonable counting rates from thin potassium sources (1 mg/cm^2). Integral bias curves were taken from 0.2 to 10 kev. The anti-coincidence ring of Geiger counters was not used during the measurement of the Co^{60} β-emission rate as a reduction in counting rate of 4% was found due to real coincidences between β-rays detected by the proportional counter and γ-rays detected by the Geiger ring.

B. CORRECTIONS.

(i) Allowance must be made for the absorption and scattering of K^{40} γ-rays by the dense and relatively high Z KOH solution. This correction was determined by adding a known small volume of $Co^{60}Cl_2$ solution to the KOH solution and comparing the increase in counting rate with that due to the same quantity of $Co^{60}Cl_2$ solution diluted with distilled water to fill the vessel. It was found that the effect of the KOH solution was to increase the counting rate slightly showing that absorption of the γ-rays was more than compensated for, in the geometry used, by scattering into the crystal. To allow for this effect,

a correction of about 6% had to be added to the Co^{60} γ-ray counting rate.

(ii) The effect on the K^{40} γ-ray counting rate of bremsstrahlung, excited by the K^{40} β-rays in their passage through the dense KOH solution, must be considered. No such correction to the Co^{60} γ-rate was necessary since (a) the intensity of the K^{40} β-rays relative to its γ-rays is a factor $\sim$16 up on the relative intensity of the β and γ-rays from Co^{60} (b) the energy of the Co^{60} β-transition is considerably less that that of the K^{40} (c) the Co^{60} β-rays pass through a solution which consists almost entirely of water whereas the K^{40} β-rays traverse a solution containing a high concentration of relatively heavy atoms.

The correction was obtained experimentally by observing the increase in the γ-rate when a small volume of a P^{32} solution, a pure β-emitter whose spectrum covers nearly the same energy range as that of K^{40}, was added to the KOH solution, the P^{32} β-activity being determined by means of the proportional counter. Fig. 5(c) shows the K^{40} γ-ray bias curve after application of this correction.

(iii) A correction for the slight difference in detection efficiency for Co^{60} and K^{40} γ-rays was determined by postulating an average path length in the crystal.

Then it can be shown that the ratio of the true Co^{60} and K^{40} γ-rates, I_{Co} and I_K respectively, is given by

$$\frac{I_{Co}}{I_K} = 1.015 \frac{\mu_K}{\mu_{Co}} \frac{\gamma_{Co}}{\gamma_K}$$

where μ_{Co} and μ_K are the mass absorption coefficients of NaI for the Co^{60} and K^{40} γ-rays and γ_{Co} and γ_K are the observed counting rates.

(iv) A small correction was calculated to allow for coincident detection in the crystal of the two γ-rays which are emitted in cascade by the Co^{60} source.

(v) If the amount of backscattering of the K^{40} and Co^{60} β-rays from the wall of the proportional counter back into the sensitive volume differs, then the observed counting rates would not be in the same ratio as the true rates of β-emission. Unfortunately the literature on the subject of the variation of backscattering factor with energy was rather contradictory. As a result, a series of experiments on backscattering was carried out and is described in the Appendix. Within the experimental error, the backscattering factor for steel showed no energy dependence, and hence no correction to the observed β-rates was necessary.

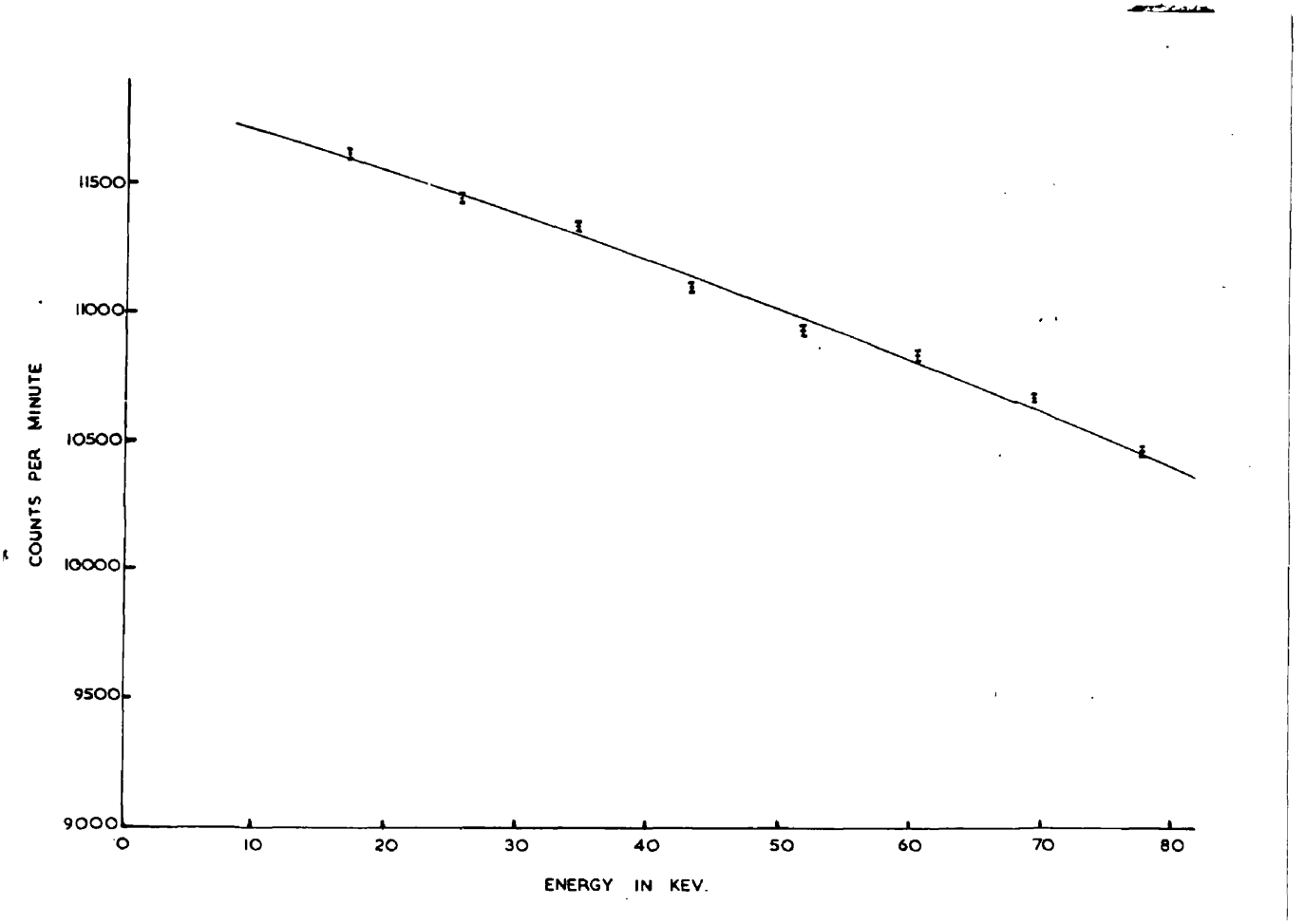

Fig.6. Discriminator bias curve for Co^{60} γ-rays.

C. RESULT.

The bias curves for the K^{40} and Co^{60} γ-rates after the necessary corrections have been applied are shown in Fig. 5(c) and Fig. 6 respectively. The counting rates γ_K and γ_{Ca} to be substituted in the formula were obtained by extrapolating these curves to zero energy. The value of the branching ratio obtained was 0.124 ± 0.002 i.e. 11% of the transitions take place by electron capture to A^{40} and 89% by β^--decay to Ca^{40}.

SECOND METHOD.

A. EXPERIMENTAL DETAILS.

This method eliminated many of the correction factors involved in the first. It made use of the fact that in the decay of Na^{24} a γ-ray occurs of energy 1.38 Mev, close to the energy of the K^{40} γ-ray, while the β-ray energies are almost identical being 1.39 and 1.32 Mev for Na^{24} and K^{40} respectively. In principle the areas under the total absorption peaks of these γ-rays were compared, the sources being monitored by their β-emission. Na^{24} also emits a 2.76 Mev γ-ray which precluded its use in the first method.

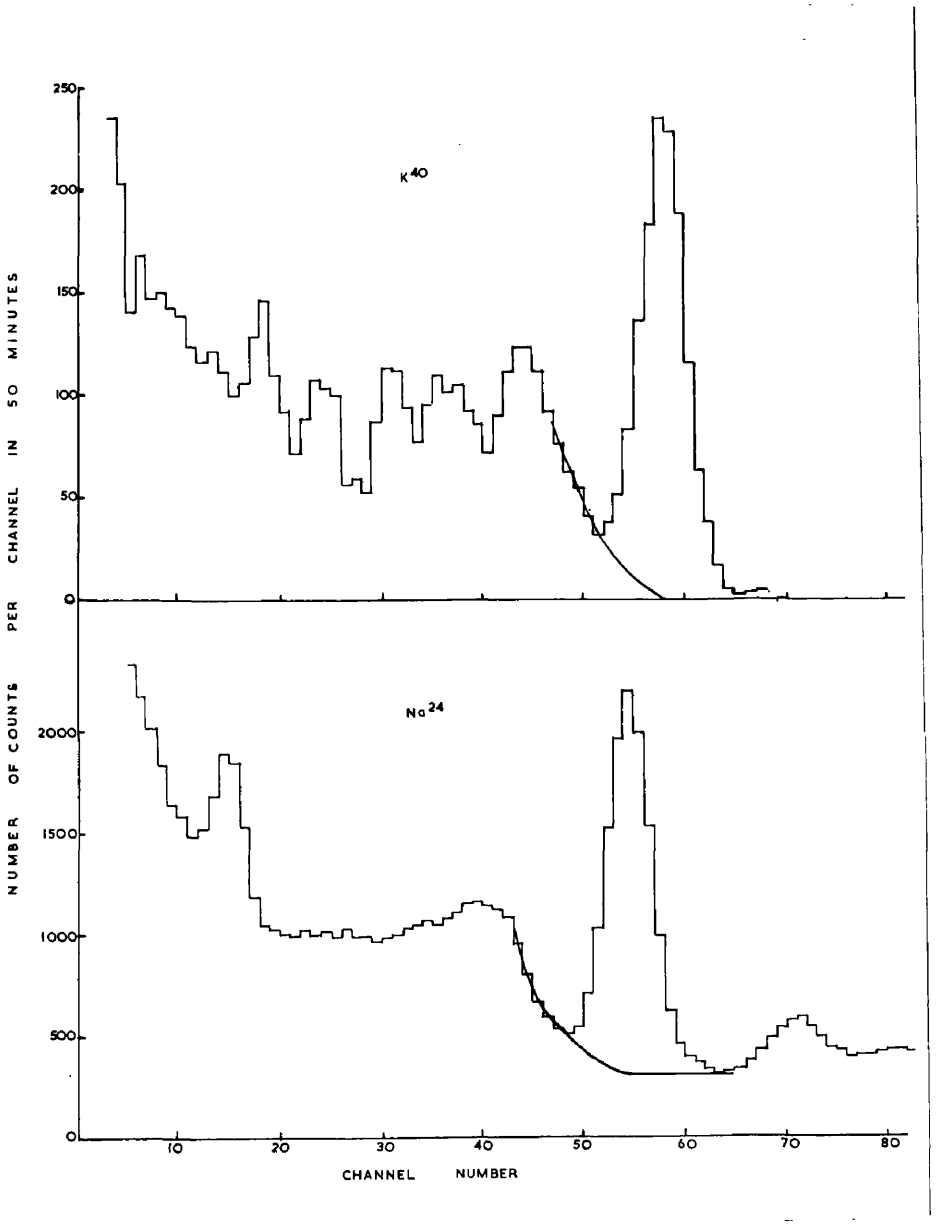

Fig.7. γ-ray spectra of K^{40} and Na^{24}.

The branching ratio is now given by the expression

$$R = \frac{\gamma_K}{\beta_K} \frac{\beta_{Na}}{\gamma_{Na}}$$

where γ_K, β_K and γ_{Na}, β_{Na} refer to the γ and β-emission rates of potassium and Na^{24} respectively.

A cylindrical glass vessel 1 in. high x $1\frac{1}{2}$ in. diam., filled with concentrated KOH solution, was placed on top of a 2 in. long x $1\frac{3}{4}$ in. diam. NaI crystal mounted on a Dumont 6292 photomultiplier, the whole being shielded by lead. The γ-ray spectrum was analysed by means of a 100 channel Hutchinson-Scarrott kicksorter. A minute quantity of $Na^{24}Cl$ solution was added to the KOH solution and a second spectrum taken. By subtraction, and knowing the shape of the background spectrum, the spectra corrected for background were obtained, (Fig.7). The areas under the total absorption peaks, in the case of Na^{24} allowance being easily made for the contribution from the Compton electrons due to the 2.76 Mev γ-ray (see Fig.7), gave a relative measurement of the γ-ray intensities. The relative β-emission rates were obtained by examination in the proportional counter of a thin source made from the mixed solutions. Counting rates were taken over a period

of several days until the Na^{24} had completely decayed. Knowing the background of the counter, the intensity of the K^{40} β-emission was simply obtained, and the relative β-emission rates of the two sources was calculated, the Na^{24} β-rate being corrected to the time when the γ-spectra were obtained. The half-life of Na^{24} was assumed to be 15.05 hr., this being the mean value of the determinations listed by Hollander et al (1953).

B. CORRECTIONS.

Consideration of the corrections which have to be applied illustrates the superiority of the present method. Since the $Na^{24}Cl$ was added to the KOH solution, no correction for absorption and scattering of the K^{40} γ-rays was required. Bremsstrahlung could not affect the magnitude of the γ-ray peaks since for K^{40} the maximum energy of the β-ray was less than the γ-ray energy and for Na^{24} the two energies were so close that any contribution to the γ-ray peak would be completely negligible. As the β-ray energies were practically the same, no correction for backscattering was necessary. (This experiment was devised before completion of the experiments showing no dependence of the backscattering factor on β-ray energy).

Furthermore the use of a mixed source in the proportional counter, together with the fact that the β-ray energies are similar, ensured that no correction would be required for source thickness, since it was effectively the same for both the K^{40} and Na^{24} measurements. Hence the only corrections to be made were:-

(i) for coincident detection of the Na^{24} 1.38 and 2.76 Mev γ-rays. Knowing the absolute disintegration rate from the β-ray experiments in the proportional counter and using our experimentally determined backscattering coefficient of 0.45 for steel, the efficiency of the crystal for 1.38 and 2.76 Mev γ-rays was calculated. The error due to coincident detection was found to be $\sim 7\%$.

(ii) for the slightly different efficiency of the crystal for 1.38 and 1.46 Mev γ-rays. This was calculated as described earlier for the first method.

(iii) for the dead time of the kicksorter, which was 0.75 milliseconds. This correction was very small, since the counting rates were low.

Thus it can be seen that, unlike the first method, the necessary corrections are easily calculable. Indeed the determination by the second method took approximately

one week, compared to six months by the first.

C. RESULT.

The value of the branching ratio was 0.121 ± 0.004, the error being mainly associated with estimating the areas under the γ-ray peaks.

3. THE DECAY CONSTANTS OF K^{40}.

The weighted mean of these two results gives a branching ratio of 0.123 ± 0.003.

From the first method, the specific β-activity was calculated by extrapolating to zero energy the bias curves obtained in the proportional counter experiment, and using a backscattering factor for steel of 0.45. The extrapolation presented no difficulty since the bias curves were very flat at low energy. The result was 27.6 ± 0.3 betas/sec/gm K. The specific γ-activity is therefore 3.41 ± 0.06 gammas/sec/gm K.

These values lead to a total half-life of $(1.28 \pm 0.02) \times 10^9$ yr assuming the abundance of K^{40} to be 0.0119 ± 0.001% (Nier 1950).

The results are in excellent agreement with the

values of 0.123, 27.6 and 3.4 selected by Endt and Kluyver (1954) as the weighted averages of all previous measurements of the branching ratio and specific β and γ-activities respectively.

4. THE EVIDENCE FOR THE MODE OF DECAY OF K^{40}.

The bulk of the evidence favouring the decay scheme of Fig. 4 is of an indirect nature since the low specific activities make coincidence experiments difficult to carry out. Perhaps the best proof is that of Johnson (1952a) who showed by measurement of the masses of A^{40}, K^{40} and Ca^{40} that the energy available for decay to A^{40} and Ca^{40} was 1.49 ± 0.07 and 1.30 ± 0.07 Mev respectively. The latter value is in good agreement with the maximum β-energy of 1.32 Mev, and is also less than the energy of the γ-ray which must therefore be associated with the decay to A^{40}. Examination of the shape of the β-spectrum failed to reveal the presence of a β-ray of lower energy, and Endt and Kluyver (1954) reported no energy level of Ca^{40} lower than 3.35 Mev. Hence it seems certain that the β-decay proceeds to the Ca^{40} ground state. Unsuccessful searches for β-γ coincidences have been carried out by Meyer et al.(1947)

who, however, give no experimental data whatsoever, and by Houtermans et al.(1950) who showed that $< 0.6\%$ of the β-rays could be in coincidence with a γ-ray. The observation by Sawyer and Wiedenbeck (1950) that the electron capture disintegration rate was approximately equal to the γ-emission rate showed that the probability of electron capture to the ground state of A^{40} was small, a result supported theoretically by Morrison (1951). Ceccarelli et al.(1950) claimed that coincidences between the X-rays, and Auger electrons, and the γ-rays had been detected. The absence of annihilation quanta (Bell and Cassidy 1950 a,b; Good 1951a) and of 180° coincidences (Colgate 1951) showed that the probability of decay by positron emission was very small, $< 0.1\%$ according to Good and $< 0.06\%$ according to Colgate. The theoretical investigation by Morrison confirmed these results. The first and second excited states of A^{40} were reported to be 1.46 and 2.4 Mev above ground (Endt and Kluyver 1954), thus indicating the existence of only one γ-ray in the electron capture branch since the available energy is only 1.49 ± 0.07 Mev. Numerous investigations of the γ-ray spectrum have failed to reveal any other transition of appreciable intensity.

It can thus be seen that there is a considerable

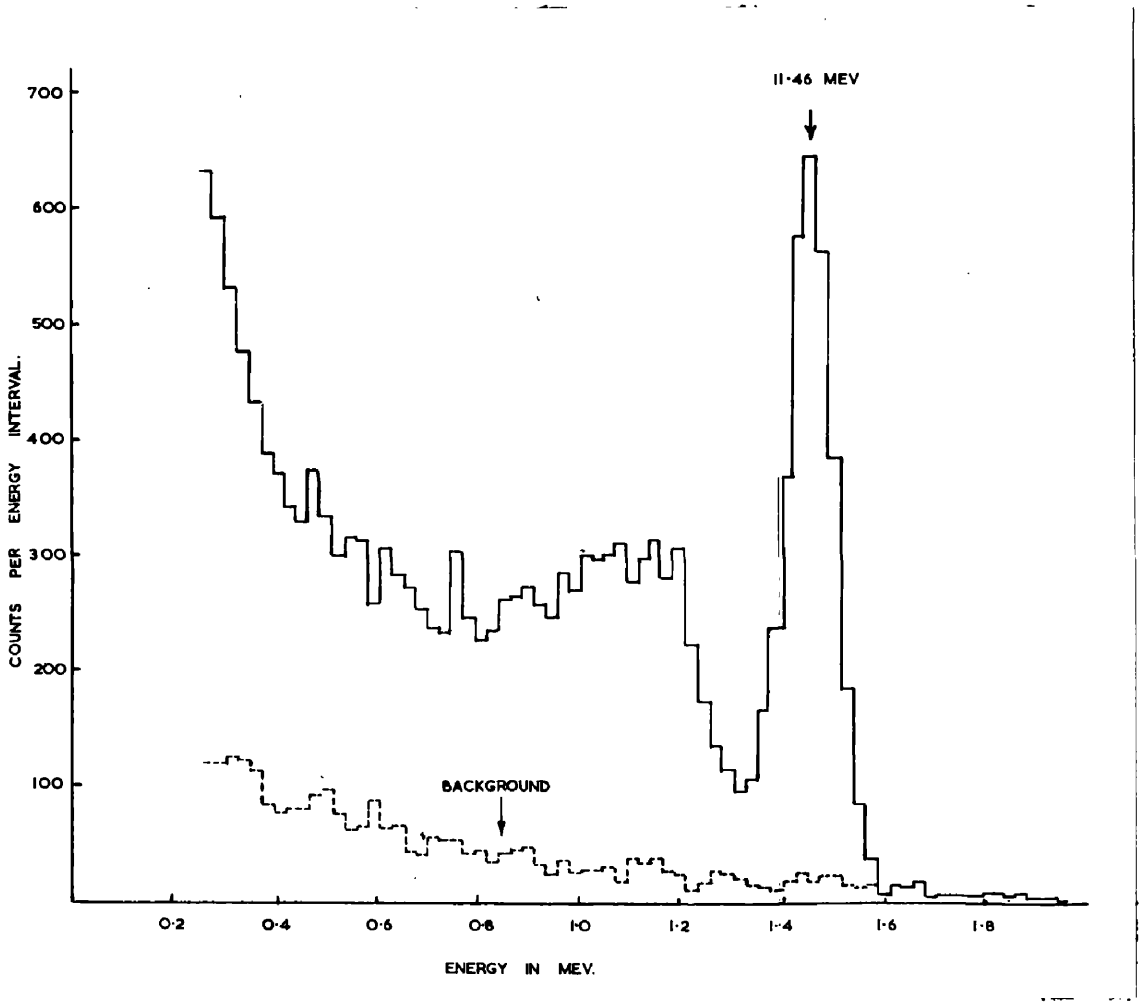

Fig.8. K^{40}. γ-ray spectrum.

body of indirect evidence in favour of the present decay scheme. However, in view of the importance of the branching ratio, it was thought worthwhile to undertake β-γ and γ-γ coincidence measurements using a scintillation counter as the γ-detector, instead of, as in the experiments of previous investigators, a Geiger counter which, of-course, has a very low efficiency for high energy γ-rays, and thus definitely to exclude any possibility that the apparent disagreement with the branching ratio determined geologically was due to a defective decay scheme.

5. EXPERIMENTAL CONFIRMATION OF THE DECAY SCHEME.

A. Gamma-ray experiments.

The γ-ray spectrum (Fig.8) emitted by 330 gm. of pure potassium chloride was examined using the crystal and kicksorter which were employed in the measurement of the branching ratio by the second method. Comparison with the γ-rays of Na^{22} (1.277 Mev) and Na^{24} (1.38 Mev) gave an energy for the K^{40} γ-ray of 1.46 ± 0.01 Mev in good agreement with the results of previous investigations (Bell and Cassidy 1950b; Good 1951b). Examination of the spectrum revealed no evidence in favour of the existence of

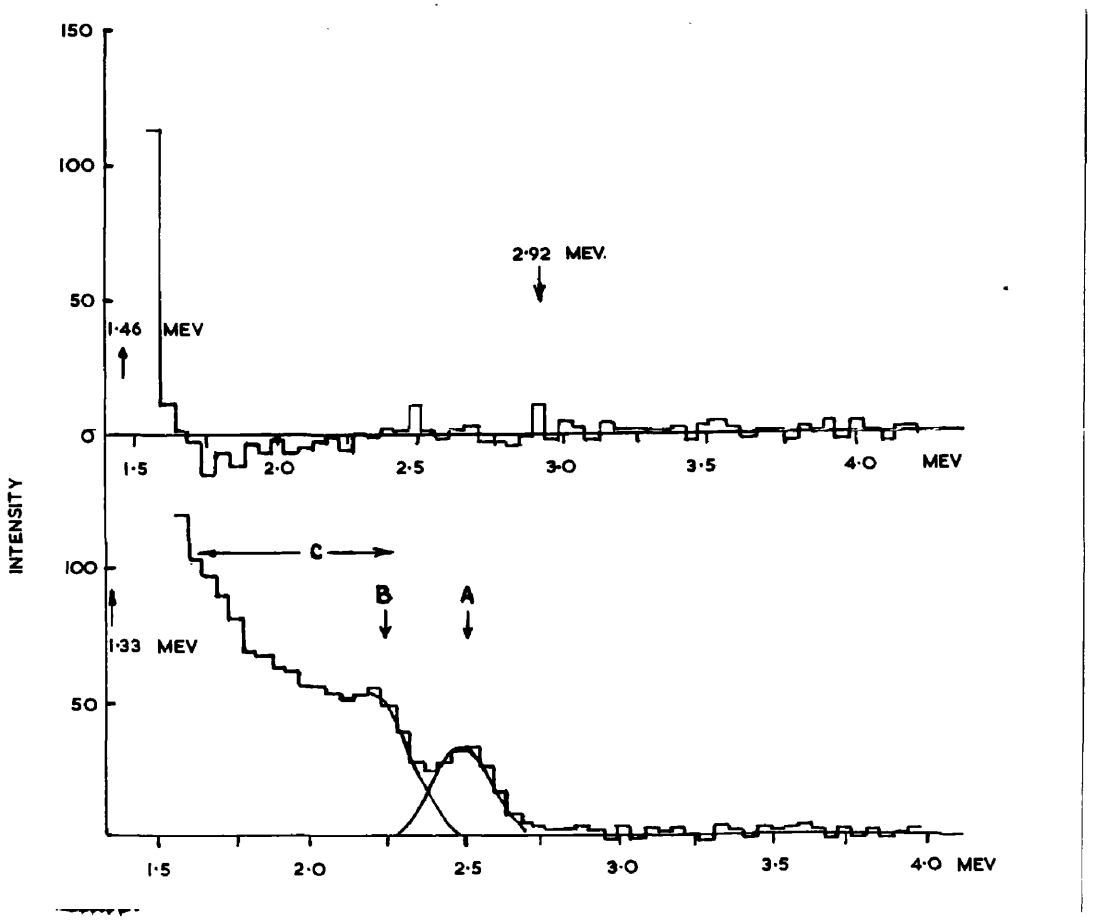

Fig.9. K^{40} and Co^{60} γ-ray spectra above 1.46 and 1.33 Mev. respectively.

another γ-ray. Experiments with the proportional counter showed no γ-ray of energy < 100 kev.

Although the mass measurements of Johnson (1952a) fairly conclusively ruled out the possibility of two high energy γ-rays, experimental confirmation was sought. Fig.9(a) shows the K^{40} γ-spectrum above 1.46 Mev while Fig.9(b) shows the corresponding region for Co^{60}, the spectra being normalised so that the γ-ray peaks were of equal intensity. In the Co^{60} spectrum there is a fairly well defined peak A at 2.5 Mev due to coincident detection of the 1.17 Mev and 1.33 Mev γ-rays. B represents the Compton edge of one γ-ray carried forward by coincident total absorption of the other. The region C is due to the integration of the two Compton distributions. No such effect is observed in the K^{40} spectrum, and hence it is deduced that the emission of two high energy γ-rays could occur in fewer than 10% of the transitions.

A γ-γ coincidence experiment was also carried out in which a 1 in. cube NaI crystal was placed against the side of the vessel used in the determination of the branching ratio by the first method, and the coincidence rate observed when the vessel was filled in turn with KOH solution and water. No γ-γ coincidences were observed,

but the overall efficiency of the system, determined by measuring the coincidence rate when the vessel was filled with the standard $Co^{60}Cl_2$ solution, was rather low.

Thus, although none of these experiments is capable of great sensitivity, it can certainly be concluded that no other γ-ray exists with an intensity approaching that required to explain the discrepancy (~30%) between the geological and physical measurements of the branching ratio.

B. SEARCH FOR β-γ COINCIDENCES.

60 mgm. of KCl enriched to 0.57% K^{40} were placed on aluminium foil of thickness 0.001 in., which acted as the reflector for the β-detector, a $2\frac{1}{2}$ in. diam. plastic phosphor, and covered with a thin plastic sheet for protection and a steel sheet 0.05 in. thick to prevent β-rays being observed by the γ-detector, a 1 in. cube NaI crystal. The whole system was surrounded by 4 in. of lead. The outputs from the two photomultipliers were amplified, passed through discriminators and then into a coincidence circuit of resolving time 0.62 microsecond. The β, γ, and β-γ coincidence rates were recorded as a function of the beta discriminator voltage.

The presence of the enriched source was found to

increase the counting rate by $(1.43 \pm 0.03) \times 10^{-3}$ coincidences per detected β-ray. Since part, at least, of this positive effect could be due to coincidences between β-rays and bremsstrahlung, a control experiment was performed using P^{32}, a pure β-emitter, and an increase of $(1.04 \pm 0.02) \times 10^{-3}$ coincidences per detected β-ray was observed. Calculations based on the efficiency of the apparatus showed that the remainder of the positive effect could reasonably be accounted for by scattering of the K^{40} γ-rays in the system. Hence there is no possibility of β-γ coincidences.

C. CONCLUSION.

The results of these experiments contribute to the mass of evidence in favour of the simple decay scheme. It is therefore concluded that the geologically determined branching ratio must be in error.

6. DETECTION OF THE AUGER ELECTRONS.

An unsuccessful attempt was made to measure the branching ratio directly in a small proportional counter (internal diam. 2 in.) by observation of the K X-rays and

Auger electrons from a thin solid source of enriched potassium. A preliminary experiment was carried out by McNair (1956). A more detailed investigation was carried out by the author assisted by Mr. D.E. Watt.

1 mg. of the enriched KCl was dissolved in alcohol and spread over an area of 150 cm^2 of the counter lining, giving an average source thickness of 7 microgm/cm^2. The anticoincidence Geiger ring was used to reduce the background to a minimum. The pulses were photographically recorded and subsequently analysed. A further experiment was undertaken using an extremely thin weak source of P^{32} and the results fitted to the K^{40} β-spectrum obtained in the first experiment. Because of the smallness of the counter and the high energy of the β-rays, the shapes of the K^{40} and P^{32} spectra in the limited energy range considered (1 - 5 kev) would be completely distorted so that fitting of the spectra is quite legitimate. The excess counts appeared not as a peak at $\sim$2.8 kev but distributed between 2.0 and 3.0 kev, the intensity being very much less than that expected. Because the source was in the form of KCl, argon K X-rays were not observed, since they are strongly absorbed by chlorine. Using a gaseous source Drever and Moljk (1955) observed a peak due to Auger

electrons following K-capture in Cl^{36}. However, with a solid source of average thickness 0.03 microgm/cm^2, only a broad distribution, not a peak, of intensity < 10% of the peak previously observed was found. It is therefore obvious that in the very low energy region reliable results cannot be obtained even with extremely thin solid sources.

The only really satisfactory method of measuring the branching ratio directly would be the use of a gaseous source, but no simple potassium compounds are gaseous around room temperature. However the success of Moljk et al. (1955) in operating proportional counters at temperatures up to 900°C suggests a possible solution of this difficulty.

7. DISCUSSION.

The agreement between these two determinations of the branching ratio is very gratifying. Since experimental evidence denies the existence in the decay scheme of any other transition of sufficient intensity to explain the discrepancy between the physical and geological methods, some discussion of the latter seems necessary.

In general, the gases, including argon, present in the mineral, usually a feldspar, were extracted by fusing

in a high vacuum system. After purification the volume of argon might be measured by means of a McLeod gauge. Mass spectrometer analysis was necessary in order to correct for, if necessary, the presence of non-radiogenic argon. The total potassium content was determined by chemical analysis. Hence, knowing the decay constants of K^{40}, the age of the mineral could be calculated, or, assuming the specific β-activity of K^{40} and the age of the mineral, usually from lead-ratio age measurements on other minerals from the same district, the branching ratio could be deduced. Two possible sources of error exist-

a) incomplete extraction of argon from the mineral or failure to remove all the extracted argon from the vacuum system, and

b) loss of argon from the mineral by diffusion over the geological period since its formation.

Until recently these possibilities had never been investigated. Carr and Kulp (1955), using A^{37} as a tracer, have shown that minute quantities of argon can be circulated round a vacuum system and then completely extracted. Furthermore, it has been demonstrated in a very ingenious manner that complete extraction of argon from a mineral is possible (Kulp 1955). A quartz capillary tube containing a minute

known quantity of A^{37} was heated with a feldspar specimen, the latter having the lower melting point. The activity of the argon extracted from the molten material was shown to be the same as the original activity, demonstrating complete recovery of the A^{37}. Thus the experimental technique seems to be free of systematic error. However Wetherill et al. (1955) found that micas consistently gave higher branching ratios than feldspars obtained from the same region. This observation has been confirmed recently by Wasserburg and Hayden (1956) and Reynolds (1956). Wetherill et al. (1956) have shown that micas give potassium-argon ages close to the uranium-lead ages, the conclusion being that there is serious leakage of argon from feldspars, but not from micas, over the geological period since their formation. The results obtained from the mica samples were analysed and the decay constants re-evaluated, a branching ratio of 0.117 ± 0.015 being found consistent with the data. The cause of the discrepancy between the geological and physical measurements has thus been revealed.

It is very satisfying that the results of several investigations published recently are in excellent agreement with the values obtained in the present experiments. A careful determination of the specific γ-activity by

Backenstoss and Goebel (1955) yielded a value of 3.50 ± 0.14. The specific β-activity is quoted by Suttle and Libby (1955) to be 29.6 ± 0.7, but, in the present author's opinion, their paper contains an arithmetical error, the corrected value being 26.9 ± 0.7. Using a KI crystal, Soji Kono (1955) obtained a specific β-activity of 29.0 ± 1.2. The low specific γ-activity reported by Suttle and Libby is not considered significant, the measurements having been made by means of a Geiger counter.

In conclusion, the physically determined value of the branching ratio is now acceptable to geologists. The old geological value, which was based on the examination of feldspars, has been shown to be too low due to argon leakage. Data recently obtained from mica samples leads to a branching ratio in agreement with the physically determined value.

CHAPTER THREE

THE NATURAL RADIOACTIVITY OF LANTHANUM.

1. INTRODUCTION.

La^{138} is an odd-odd nucleus, and the middle member of the isobaric triplet (Ba^{138}_{56}, La^{138}_{57}, Ce^{138}_{58}). Since, theoretically, no odd-odd nucleus above N^{14} should be stable, La^{138} may be expected to decay both by electron capture (and perhaps β^+-emission) and by β^--emission, the long half-life being due to a large spin change involved in its decay.

Gamma-activity was first observed by Pringle et al. (1950) who reported a γ-ray of energy 1.05 Mev. Subsequently Bell and Cassidy (1950c) detected γ-rays of energies 0.545 and 1.06 Mev and a reinvestigation by Pringle et al. (1951) showed three γ-rays of energies 0.535, 0.807 and 1.39 Mev, their explanation of the previously reported 1.05 Mev γ-ray being that what had appeared to be the γ-ray spectrum end-point in the earlier low resolution experiment was in fact the Compton edge. The 1.39 Mev γ-ray was considered to be a crossover transition. In

the absence of β-rays, the γ-radiation was assigned to the electron capture branch, and measurement of the relative intensities led to the proposal of electron capture to both the first and second excited states of Ba^{138}, which were suggested to be at 0.807 and 1.39 Mev respectively. However investigations of the γ-rays following the β^--decay of Cs^{138} to Ba^{138} (Langer et al. 1953; Thulin 1955) revealed no 0.81 Mev γ-ray. Furthermore the systematics of Scharff-Goldhaber (1953) predict an energy ~1.5 Mev for the first excited state of Ba^{138}. Pringle et al.(1951) did not consider the possibility of β^+-emission to either of the excited states or to the ground state to be excluded. Neither γ-γ nor K-γ coincidence experiments were attempted. The half-life was calculated to be 2×10^{11} yr.

In a rather insensitive experiment using a thin window Geiger counter Pringle et al. (1951) assigned an upper limit of 12 counts/min./gm. La_2O_3 to the number of electrons or positrons emitted with energy > 100 kev. With the aid of a large proportional counter Mulholland and Kohman (1952b) reported β^--emission of maximum energy 1.0 ± 0.2 Mev measured by aluminium absorption methods, and partial half-life 1.2×10^{12} yr. These authors mentioned the difficulties caused by differing radioactive

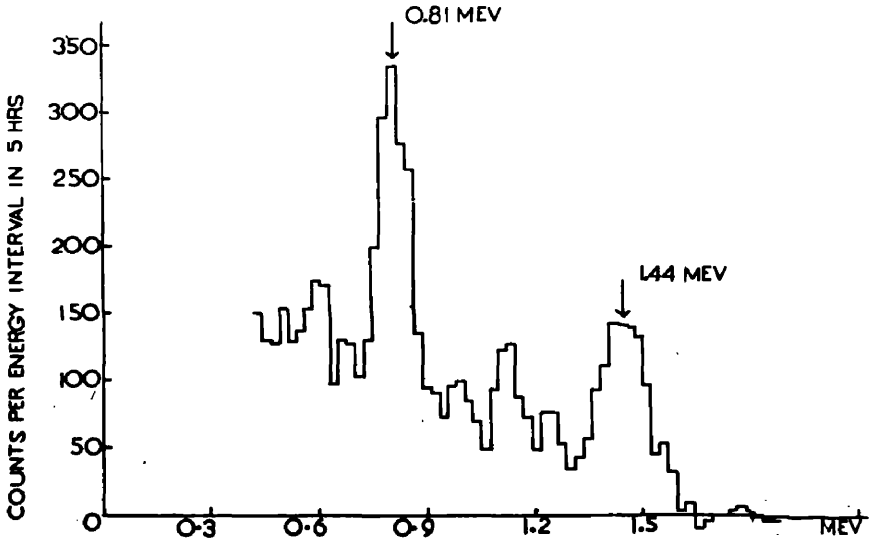

Fig.10. La^{138} γ-ray spectrum.

contamination of their absorbers, and, in view of the fact that Dixon and McNair (1954) discovered that a β^--spectrum which had been attributed to rhenium (Curran 1952) was in reality due to differing contamination of their aluminium counter linings, further investigation seemed essential especially since a search for β-rays of energy >200 kev carried out by Selig (1954) yielded negative results.

It can thus be seen that the validity of the decay scheme is doubtful and the existence of the β-ray questionable. Hence the present complete investigation of La^{138} was undertaken.

2. GAMMA RAY SPECTRUM.

The crystal was surrounded by 7 gms of lanthanum oxide La_2O_3, distributed uniformly to give an average source thickness of approximately 70 mg/cm^2. The activity due to the source was 19 cpm compared with a background of 49 cpm above $\sim$200 kev. The spectrum with background subtracted (Fig.10) shows the presence of two γ-rays of energies 0.81 ± 0.01 Mev and 1.44 ± 0.02 Mev. Although the measurements were extended to 2.5 Mev, no other γ-ray was observed, nor was there any indication of pulses due to

simultaneous detection of the two γ-rays as might be expected if they were in coincidence. Furthermore, closer investigation failed to reveal either a γ-ray of energy 0.535 Mev or annihilation radiation, which would be expected if β^+-emission occurred. It seems probable that statistical fluctuations caused Bell and Cassidy (1950c) and Pringle et al.(1951) to interpret the Compton edge of the 0.81 Mev γ-ray as evidence of the existence of a 0.54 Mev γ-ray.

The need for sources of very high chemical purity must be emphasised. Spectrographically pure La_2O_3 was found to give a spectrum showing several γ-rays at energies <0.4 Mev, their total activity being considerably greater than that of the high energy γ-rays. Another sample of different origin showed low energy γ-rays of almost the same intensity, hence suggesting that this γ-radiation was characteristic of La^{138}. Experiments showed that the low energy γ-rays were in coincidence with one another and with the 0.81 Mev γ-ray. It was extremely difficult to understand how so many previously unknown excited states presumably of Ce^{138} could be accommodated in the decay scheme. Furthermore in view of the observed relative intensities of the γ-rays, a complex β-spectrum was

indicated whereas, to anticipate our results, the β-spectrum appeared to be simple. However an extremely pure sample of lanthanum oxide, generously supplied, originally in connection with the β-ray experiments, by Dr. W. Turchinetz of the University of Manitoba, Canada, was found to show practically no low energy γ-rays, and was used to obtain the spectrum shown in Fig.10. Hence these low energy γ-rays must be due to contamination. In this sample, although the magnitude of the 1.44 Mev peak was unchanged, the ratio of counts in the 0.81 and 1.44 Mev peaks was approximately 1:1, compared with 2:1 in the previous sources. Thus almost half of the 0.81 Mev γ-rays in the earlier sources must have been contamination due to a γ-ray of energy very close to 0.81 Mev, and in coincidence with the low energy γ-rays. Estimation of the purity of the Canadian source relative to the earlier sources as indicated by the amount of low energy contamination showed that a maximum of 3% of the 0.81 Mev γ-rays still present could be due to impurity. Hence the 0.81 Mev γ-ray must be characteristic of the decay of La^{138}. It will be appreciated that considerable time and effort were required to reach these conclusions. This problem of source purity will be seen to occur frequently in low activity work in general,

and will be discussed in Chapter 7.

To summarise, only two γ-rays of energies 0.81 ± 0.01 and 1.44 ± 0.02 Mev were found to be associated with the decay of La^{138}.

3. BETA-RAY SPECTRUM.

0.927 gm of the extremely pure finely divided La_2O_3 powder was spread over 850 cm^2 of the removable copper lining of the proportional counter, the source hence being kept well within the effective counting volume. The average source thickness was 1.1 mg/cm^2. For the background run, a similar source of tin dioxide was used, since it was found that the background with an inert source in the counter was always slightly lower than with only a bare copper lining. This effect was most noticeable at energies <50 kev. Hence it is presumed that the inert source absorbs low energy electrons either produced by conversion of external γ-rays in the copper wall or due to a slight contamination of the copper itself. The counter was filled to a pressure of 4 atm. and calibrated by means of the 46.7 kev γ-rays of RaD. A counting rate of 4.3 cpm was observed above the background of 27.5 cpm. The spectrum

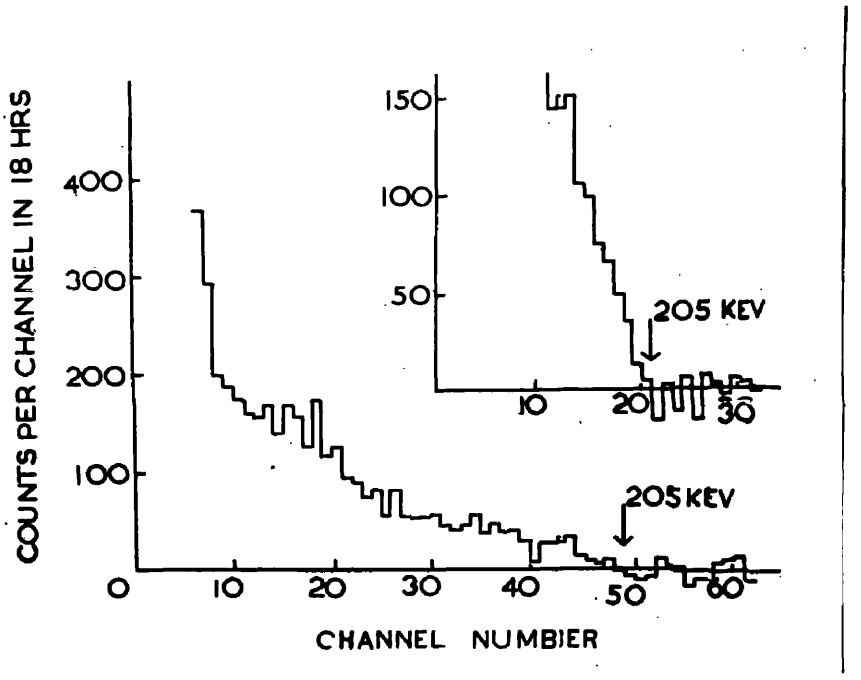

Fig.11. La^{138} β-ray spectrum. The inset spectrum was taken with a thicker source and lower amplifier gain to demonstrate the end-point more clearly.

obtained (Fig.11) indicated a maximum energy of 205 ± 10 kev for the β-ray. The end-point was observed more clearly (Fig.11, inset) by using a thicker source and lower amplifier gain. There was no indication of a β-ray of maximum energy ~1.0 Mev and half-life 1.2 x 10^{12} yr. as reported by Mulholland and Kohman (1952b) although such an activity would have been well within the limits of detection of the system used.

In earlier experiments using the contaminated sources a β-ray of the same maximum energy and intensity was detected, but these sources gave an extremely high α-rate (~750 cpm). Therefore a sample of very pure La_2O_3 was requested from Dr. Turchinetz in order to confirm this β-activity. The agreement between the intensity of the β-rays from the pure and contaminated sources shows that the β-activity cannot be associated with the α-contamination but must be due to the decay of La^{138}. As a result of this experience, the α-activity of every source required for the investigations described in this thesis was measured before use. If this exceeded a value of ~2 cpm, which appeared to be the minimum level of impurity attainable, the source was immediately rejected. This method is considerably more sensitive than any chemical or spectrographic test of

purity.

It was observed (Fig.11) that the spectrum appeared to exhibit a sharp rise at low energies. A thinner source of mass 0.316 gm. and average thickness 0.37 mg/cm^2 was used to examine the energy range 5 - 50 kev and gave a total counting rate of 2.6 cpm, considerably higher than that expected from comparison with the observed activity of the 1.1 mg/cm^2 source, hence suggesting the presence of low energy β-rays which were being seriously absorbed in the thicker source. Indeed at least 75% of the observed pulses were of energy < 50 kev. Therefore although the spectrum must still be distorted due to the necessary source thickness, the results suggest that the rise at low energy is real and that the shape is similar to that of Rb^{87} (Curran et al. 1952; Lewis 1952). Further investigation of this point using a sufficiently enriched source, as yet nowhere available, is obviously required.

4. COINCIDENCE EXPERIMENTS.

The coincidence experiments were carried out under a 6" lead shield, each detector being a 2 in. high x 1¾ in. diam. NaI crystal mounted on a Dumont 6292 photomultiplier.

To obtain the best possible geometry, the counters were separated only by the space required to insert the La_2O_3 source. The output pulses from one photomultiplier, after amplification, were passed through pulse lengthener and brightener circuits and viewed on a cathode ray tube as a spectrum of bright spots in the X-direction. The pulses from the other multiplier were amplified and lengthened, and then applied in the Y-direction, so that a coincidence appeared as a bright spot whose displacement from the X and Y axes was proportional to the energy spent in each crystal. The resulting pattern was filmed as a series of ten minute exposures and then analysed.

For the investigation of possible coincidences between the 0.81 and 1.44 Mev γ-rays only coincidences in which an energy >0.65 Mev was spent in both crystals were considered. The total coincidence rates for source + background and background respectively were 106 ± 10 and 96 ± 10 coincidences per 6 hrs. From a knowledge of the geometry and the γ-ray counting rates in each crystal a coincidence rate greater than twice the background rate would have been expected. Hence these two γ-rays cannot be in coincidence. Examination of the energy range 0.35 - 0.65 Mev showed no evidence of coincidences between

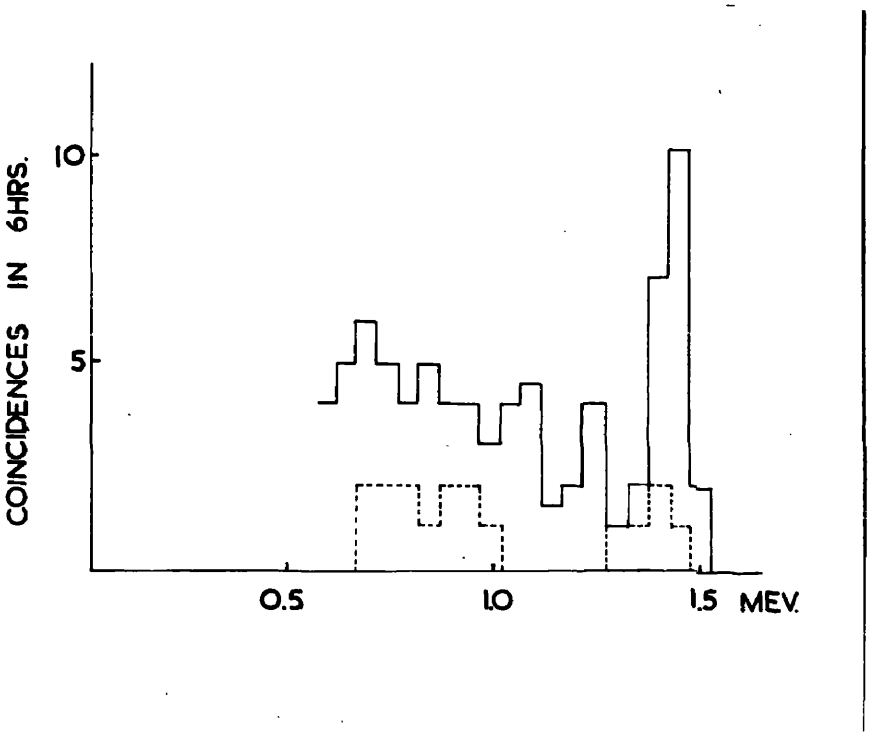

Fig.12. Spectrum of pulses in coincidence with Ba K X-rays following electron capture.

annihilation quanta, indicating that β^+-emission, if it occurs, must be of very low intensity, certainly $< 1\%$, compared with electron capture.

In the K-γ coincidence experiment a lead absorber 3 mm thick was placed between the γ-ray counter and the source to prevent any iodine escape X-rays, whose energy is close to that of the barium K X-rays following K-capture, being observed in the crystal used to detect X-rays. The spectrum of pulses in coincidence with the barium K X-rays (Fig.12) clearly shows that only the 1.44 Mev γ-ray follows electron capture. The number of coincidences under the 1.44 Mev peak alone is 21 in 6 hr. compared with a background of 4.

The decay scheme proposed on the basis of these experiments is shown in Fig.13. Observation of β-γ coincidences would provide final confirmation. However for such an experiment a thin source would be required to avoid appreciable absorption of the β-rays, and so, because of the very restricted source area available in an experiment of this nature, an attempt using unenriched material was not considered worthwhile.

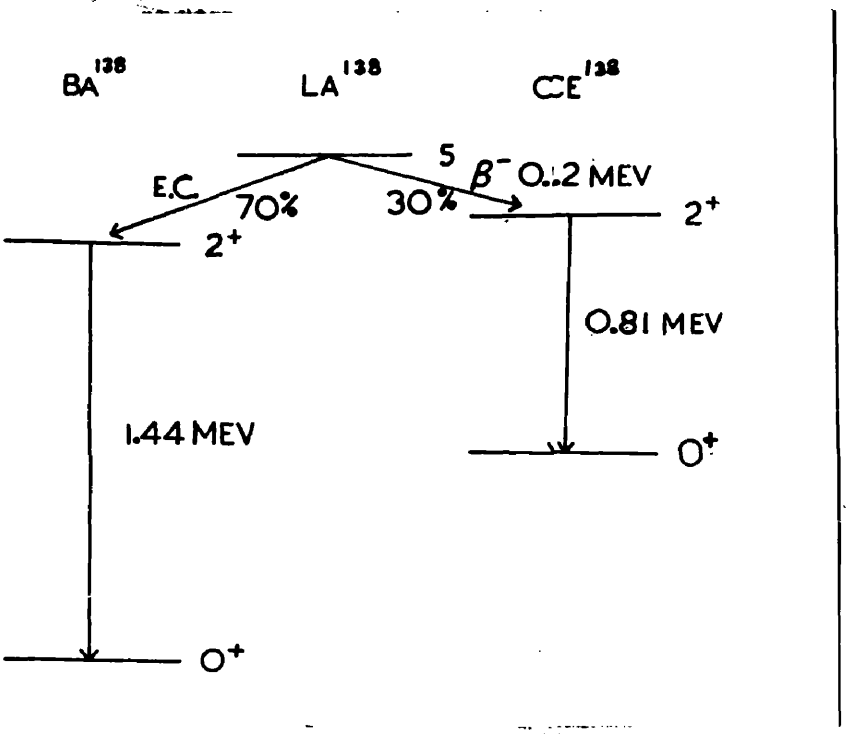

Fig.13. Proposed decay scheme of La^{138}.

5. DECAY CONSTANTS.

Since the 1.44 Mev γ-ray follows electron capture, the specific activity of this branch can be simply determined by comparison with the K^{40} 1.46 Mev γ-ray. Spectra were taken with sources of La_2O_3 and KCl under identical geometrical conditions. The detection efficiency of the crystal was taken to be the same for both γ-rays. By determining the total counts under the La^{138} 1.44 Mev and K^{40} peaks, knowing the mass of each source, the specific activity of the electron capture branch was found to be 30.9 ± 1.2 disintegrations/min/gm La. The specific activity of potassium was taken to be 3.47 gammas/sec/gm K, the mean of the results of Backenstoss and Goebel (1955) and McNair et al.(1956). Since the abundance of La^{138} is 0.089% (Inghram et al.1947) the electron capture partial half-life is $(1.64 \pm 0.06) \times 10^{11}$yr. This result is in agreement with that of 2×10^{11}yr obtained using a similar method by Pringle et al.(1951), but not with a value of 4×10^{13}yr estimated by Selig (1954) from observation of Ba K X-rays using a proportional counter.

By comparison with the K^{40} γ-ray spectrum extrapolated to zero energy the counts in the Compton

distribution of the La^{138} 1.44 Mev γ-ray were estimated. Hence, knowing the total counts N_1, N_2 due to the 1.44 and 0.81 Mev γ-rays respectively, the partial half-life of the β-branch can be deduced from the formula

$$t_\beta = \frac{N_1}{N_2} \cdot \frac{1 - e^{-\mu_2 m}}{1 - e^{-\mu_1 m}} t_K$$

where t_β and t_K are the partial half-lives of the β and K capture branches and μ_1, μ_2 the total mass absorption coefficients for the 1.44 and 0.81 Mev γ-rays respectively. 'm' is the average thickness of the NaI crystal in gm/cm^2. The partial half-life thus deduced was $(3.5 \pm 0.3) \times 10^{11}$ yr., the corresponding specific activity being 14.6 ± 1.2 disintegrations/min/gm La.

Using the thinner source in the proportional counter, the β-rays of energy >4 kev were counted. By subtracting the Ba K X-rays detected following electron capture, extrapolating the β-rate to zero energy, and correcting for self-absorption and backscattering (45%) (McNair et al. 1956), a specific activity of 12.4 ± 2.1 betas/min/gm La was obtained. The large probable error is mainly associated with poor statistics since the source counting

rate was only ~8% of the background. The corresponding partial half-life is $(4.1 \pm 0.7) \times 10^{11}$ yr. The specific activity is perhaps slightly lower than that found by the previous method. The suggested spectrum shape could provide a simple explanation, since even the thinner source would absorb an appreciable fraction of those β-rays with very low energy. However the agreement between the values found by these two different methods is sufficiently good to provide further indirect evidence that the β-ray and 0.81 Mev γ-ray are in coincidence.

From these results, using the mean of the two values for the activity of the β-branch, it is concluded that approximately 70% of the disintegrations proceed by electron capture to Ba^{138} and 30% by β-decay to Ce^{138}.

6. DISCUSSION.

The spin of La^{138} has recently been measured to be 5 (Sogo and Jeffries 1955), in agreement with the shell model which predicts a $(d_{3/2}, g_{7/2})$ configuration. On this basis the ground state of La^{138} has even parity. The survey of Scharff-Goldhaber (1953a) indicates that the first excited state of even-even nuclei is almost invariably

2+. Hence the electron capture and β^--transition might both be expected to be $\Delta I = 3$, no i.e. unique 2nd forbidden with log ft. ~ 13. However the experimental log ft. value for the β-decay is 19.0, corresponding to a 3rd forbidden transition, not 2nd forbidden. Goeppert-Mayer (1955) has pointed out that the odd parity of the ground states of ${}_{51}Sb_{71}^{122}$ and ${}_{53}I_{71}^{124}$ indicated that the $h_{11/2}$ levels could be occupied by an odd number of neutrons in the ground states of odd-odd nuclei. Hence in ${}_{57}La_{81}^{138}$ it is possible that the last odd neutron is in an $h_{11/2}$ level instead of $d_{3/2}$. The parity would then be odd and a 3rd forbidden $\Delta I = 3$, yes transition would result. However the spin of 5 would then not be so easily explicable by the shell model, although the validity of its application to odd-odd nuclei is questionable. In support of the assignment of odd parity to the La^{138} ground state it should be noticed that the shape of the β-spectrum seems to resemble that of Rb^{87} the only known $\Delta I = 3$, yes transition. Further investigation is required to establish the shape conclusively.

Since there are several excited states of Ba^{138} <2.5 Mev above ground (Thulin 1955), the presence of only the 1.44 Mev γ-ray following electron capture indicates

that β^+-emission to the first excited state of Ba^{138} is energetically impossible. Decay to the ground state is obviously highly forbidden because of the large spin change ($\Delta I = 5$) involved. The inability to detect annihilation quanta experimentally is thus easily understood. Similarly decay to the ground state of Ce^{138} ($\Delta I = 5$) with emission of a 1.0 Mev β-ray is highly forbidden relative even to the excited state. The absence of a 1.0 Mev β-ray (its intensity cannot be $>5\%$ of the 0.2 Mev β-ray) is thus not unexpected.

The present investigation confirms that the 1.44 Mev γ-ray, which also follows the β^--decay of Cs^{138}, is a transition from the first excited state of Ba^{138} to the ground state, in agreement with the prediction of the systematics of Scharff-Goldhaber (1953). A 0.80 Mev γ-ray was observed by Handley and Olson (1954) following β^+-decay of Pr^{138} to Ce^{138}, hence providing further evidence in favour of the proposed decay scheme.

After the completion of this investigation, Turchinetz and Pringle (1956) reported that the γ-ray spectrum had been re-examined, only two γ-rays of energies 1.43 ± 0.01 and 0.81 ± 0.01 Mev being found, and that the 1.43 Mev γ-ray had been shown to be in coincidence with the

Ba K X-ray following electron capture but not with the 0.81 Mev γ-ray. There is excellent agreement between this and our own investigation. These investigators estimated a log ft. of 16.9 for the electron capture decay which suggests a 3rd forbidden transition, and therefore tends to support our assignment of odd parity to the La^{138} ground state.

From the Ba K X-rays observed in our proportional counter the half-life of the electron capture branch was estimated, knowing the detection efficiency of the counter for 32 kev X-rays and using the L/K ratio quoted by Turchinetz & Pringle (1956), who measured the K X-rays with a scintillation counter. The result was in agreement with the value estimated from the activity of the 1.44 Mev γ-ray. It seems certain that the half-life of 4×10^{13} yr. calculated by Selig (1954), also from the K X-ray activity observed in a proportional counter, is erroneous.

The possibility of using La^{138} for geological dating exists. Since Ba^{138} is of 71.66% abundance whereas that of Ce^{138} is only 0.25%, only the β-ray branch would be useful since a very small change in the relative abundance of Ba^{138} due to the La activity could not be accurately measured. However a more accurate determination of both

the electron capture and β-decay half-lives would be required since the values found by Turchinetz and Pringle (1956) and in the present investigation are not in sufficiently good agreement, particularly for the β-decay branch.

CHAPTER FOUR

A SEARCH FOR NATURAL RADIOACTIVITY IN VANADIUM.

1. INTRODUCTION.

Mass spectrometer measurements by Johnson (1952b) have indicated that the odd-odd nucleus V_{23}^{50} has an energy of 2.39 ± 0.12 Mev available for decay to Ti^{50} and 1.19 ± 0.12 Mev for decay to Cr^{50}. The spin of V^{50} was measured to be 6 by Kikuchi et al. (1952, 1953) and Baker and Bleaney (1952). β^--decay should occur to the ground state of Cr^{50} with spin change $\Delta I = 6$, since its first excited state is at 9.5 Mev (Endt and Kluyver 1954). Only one excited state of Ti^{50} exists below 2.5 Mev, namely a 2+ level at 1.58 Mev (Pieper 1952). Hence an electron capture transition with $\Delta I = 4$ accompanied by a γ-ray of energy 1.58 Mev might be expected.

Due to the low abundance of the isotope, 0.25% (White et al. 1956), and the degree of forbiddenness of the transition, detection of activity has proved to be extremely difficult. Unsuccessful attempts have been made by Sheline, Strome (1954) and Selig (1954). The minimum

TABLE 1.

	t_γ	t_κ	t_{β^+}	t_{β^-}
Sheline	10^{14}			
Strome		10^{14}		
Selig		2×10^{14}		3×10^{14}
Heintze	3×10^{15}	3×10^{14}	10^{15}	3×10^{14}

half-lives quoted by these authors are given in Table 1. However it is impossible to assess the worth of these results since details of none of these experiments, particularly the energy down to which the measurements were extended, have been published. The longest minimum half-lives have been quoted by Heintze (1955). However the ferrovanadium source used was stated to be contaminated, the minimum half-lives being arrived at by estimating the contribution to the observed activity of various possible contaminants. Furthermore in deducing the minimum half-life for K-capture no allowance appears to have been made for the facts that firstly, only a small fraction of the 4.5 kev X-rays could escape from a thick source (27% from a 10 mg/cm^2 source) and secondly, in only 25% of the K-capture transitions are X-rays emitted, Auger electrons which would be absorbed in the source being emitted in the remaining 75%. Hence the minimum K-capture half-life which can be deduced from Heintze's experiment is considerably shorter than the quoted value of 3×10^{14} yr.

Because of the sensitivity of the present apparatus, especially the scintillation spectrometer, an investigation of V^{50} was undertaken.

2. SEARCH FOR β^--DECAY AND ELECTRON CAPTURE.

This investigation was carried out using the proportional counter and associated apparatus described earlier. The source consisted of vanadium pentoxide spread over an area of 850 cm^2, the average thickness being 10 mg/cm^2. For the background a similar source of chromium oxide was used. Obviously if the β-rays were of maximum energy 1.2 Mev, then an undistorted spectrum could not be obtained using the proportional counter. Therefore this experiment was undertaken only to determine if β-emission could be detected. The difference between the source + background and background counting rates over a period of 8 hr. was $+113 \pm 137$. It was therefore concluded that β-emission must be beyond the limit of detection of the apparatus. Assuming a detectable limit of thrice the statistics and an isotopic abundance of 0.25% (White et al. 1956), then, taking into account β-ray absorption in the thick source, the half-life for emission of β-rays of energy >50 kev must be $> 2.4 \times 10^{14}$ yr. In case the β-rays were emitted predominantly with very low energy as in the Rb^{87} spectrum, the measurements were extended so that pulses of energy > 1.5 kev could be detected. No activity

was observed. For a 1 mg/cm^2 source, again allowing for absorption, the corresponding half-life for β-emission must be $> 3.5 \times 10^{13}$yr.

Using the 10 mg/cm^2 source, the K X-ray region was closely examined, the source + background and background counting rates per 8 hr. being 1155 ± 34 and 1110 ± 33 i.e. a difference of $+45 \pm 47$. Assuming the same detectable limit as above, then, allowing for absorption in the source of the X-rays which would be produced in only 25% of the transitions, the K-capture half-life must be $> 4 \times 10^{13}$yr.

3. SEARCH FOR γ-EMISSION.

Using the scintillation spectrometer shielded by 4 in. lead lined with 4 in. steel, a search for γ-rays was undertaken. The large NaI crystal was surrounded by 110 gms of vanadium pentoxide, and by the same mass of chromium oxide for the background run, both experiments thus being done under identical conditions. The source + background and background counting rates above an energy of 200 kev were consistent over several periods adding up to 10 hr., the totals being 28503 ± 169 and 27644 ± 166 respectively, an excess of 859 ± 237. Since this excess

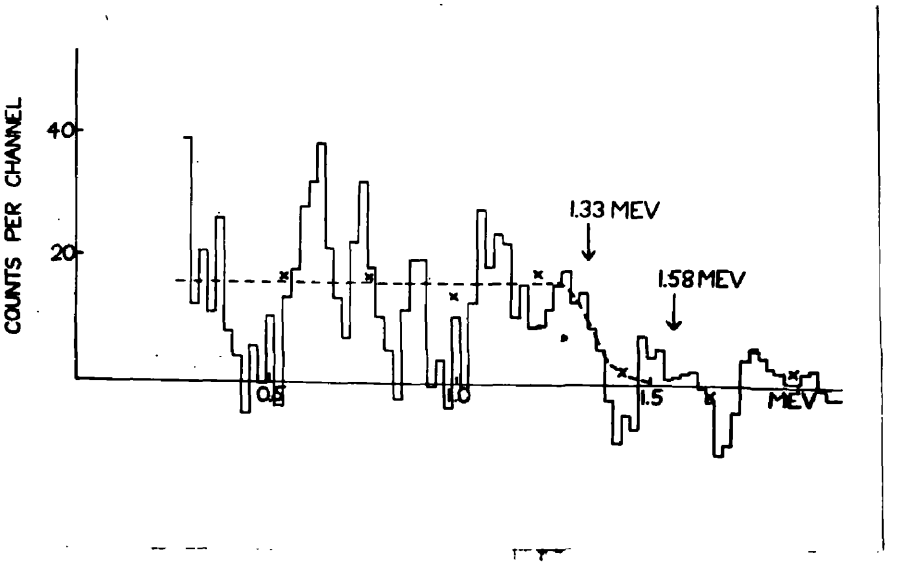

Fig.14. V^{50} γ-ray spectrum. The average excess counting rate in each group of ten channels is indicated by a cross.

counting rate is only 3% of the total background, the statistics are very poor, as can be seen from Fig.14. There is no appreciable peak at 1.58 Mev, but it seems significant that the spectrum drops sharply at 1.33 Mev, the energy at which the Compton edge would be expected. Also the result obtained from averaging the excess counting rate in each group of ten channels (see Fig.14) suggests that the counts below 1.33 Mev are spread evenly over the spectrum, as would be expected for a Compton distribution. It is unlikely that the activity detected is due to impurity. Failure to detect β-activity eliminates the possibility of β-contamination. The α-activity of both the vanadium pentoxide and chromium oxide was found to be only ~2 cpm, the minimum amount of impurity apparently attainable. Also although tests have been carried out on the oxides of six different elements which gave this α-rate of 2 cpm, no γ-activity has been found to be associated with the α-activity. These results therefore indicate that the γ-activity observed follows the decay of V^{50} by electron capture to the first excited state of Ti^{50}. By surrounding the crystal with a similar mass of potassium bicarbonate in the same geometry, the electron capture half-life of V^{50} was estimated to be $(4.0 \pm 1.1) \times 10^{14}$ yr.

4. DISCUSSION.

Failure to detect β-activity was not unexpected since this transition ($\Delta I = 6$) would be highly forbidden, even relative to the electron capture transition ($\Delta I = 4$). The sensitivity of the present apparatus is greater than that of previous investigators, but informative comparison with the lower limits of the beta half-life given in Table 1 is impossible since no other investigator has indicated the energy down to which his measurements extend, on which energy the lower limit depends since the background decreases, and therefore the lower limit which can be placed on the half-life increases , as this energy increases.

Comparison of the half-life deduced from the observed γ-activity with the lower limit attainable in the proportional counter search for K X-rays shows the impossibility of detecting these X-rays using a thick source of unenriched material. It is estimated that if the search for X-rays was carried out using a minimum of 250 mg of vanadium pentoxide, 50 times enriched in V^{50}, then a significant counting rate should be obtained.

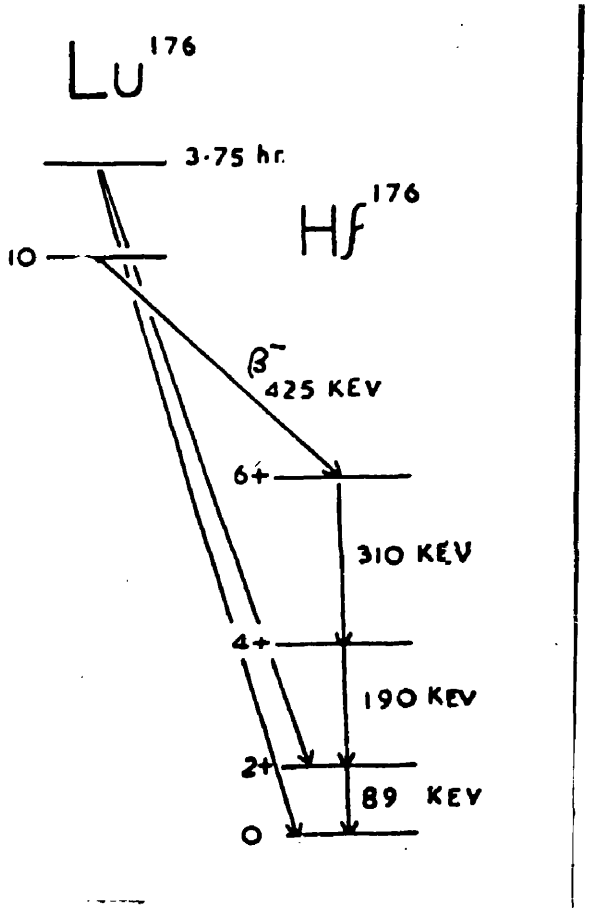

Fig.15. Decay scheme of Lu^{176}.

CHAPTER FIVE

A SEARCH FOR ELECTRON CAPTURE IN Lu^{176}.

1. INTRODUCTION.

Recent investigations by Arnold and Sugihara (1953), Arnold (1954) and Dixon et al. (1954) have conclusively confirmed the β^--decay scheme of Lu^{176} (Fig.15) first suggested by Goldhaber and Hill (1952) on the basis of unpublished work by Scharff-Goldhaber (1952). Lu^{176} being an odd-odd nucleus and the middle member of the isobaric triplet (Yb^{176}_{70}, Lu^{176}_{71}, Hf^{176}_{72}), decay by electron capture might also be expected. However such a mode of decay has never been established. Scharff-Goldhaber (1952) could not detect electron capture, and the results of Arnold (1954) indicated that the intensity of electron capture was certainly $<10\%$ of that of the β-rays. Dixon et al. (1954) examined the L X-ray peak. It consisted mainly of those Hf L X-rays due to K and L internal conversion of the 89 kev γ-rays which, because of the counter geometry, were detected unaccompanied by conversion electrons or β-rays. However they considered that the observed width of this

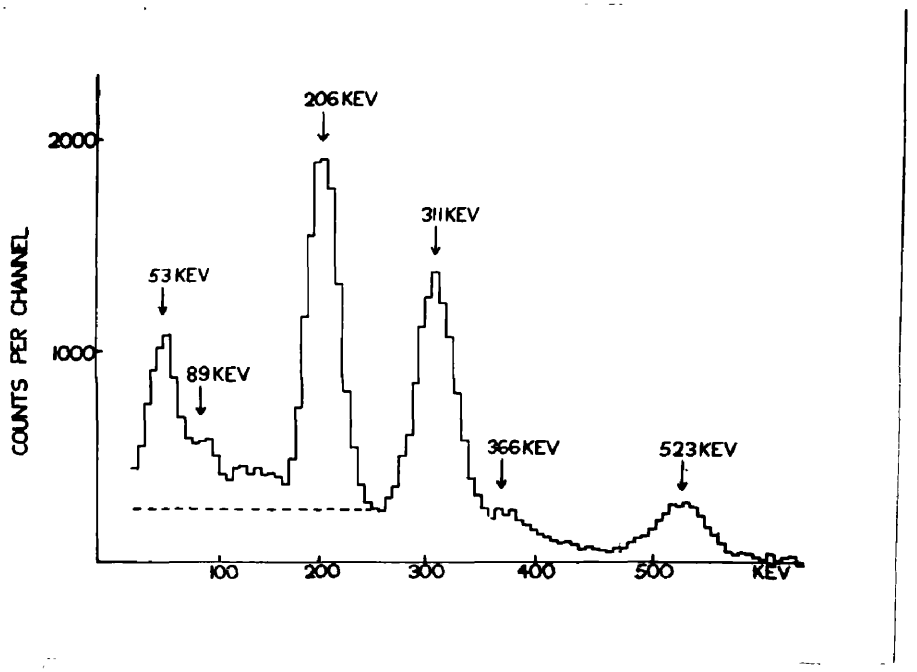

Fig.16. Lu^{176} γ-ray spectrum.

peak, compared with that of the Ga K X-ray calibration, was compatible with an admixture of ~15% of Yb L X-rays, and hence tentatively proposed that 3 ± 1% of the disintegrations took place by electron capture. In a private communication (1956) Dixon suggests that this might rather be regarded as an upper limit. Since the ground state spin of Lu^{176} is 10, it is unlikely that electron capture occurs to the ground state of Yb^{176}, since its spin is zero. After a re-examination of the experimental data contained in the paper of Dixon et al. (1954), McNair (1956) suggested that (8 ± 1)% of the transitions take place by electron capture, and that there was some evidence for the occurrence of a γ-ray of energy 115 ± 10 kev and possibly another of energy ~250 kev. To test these suggestions a search was carried out for γ-rays or conversion electrons which could be attributed to the electron capture branch.

2. THE γ-RAY SPECTRUM.

The γ-ray spectrum from 500 mgm. of lutetium oxide Lu_2O_3 placed on top of the 2 in. crystal is shown in Fig.16. The source counting rate was 700 cpm compared with the background of 80 cpm. The energies of the γ-rays were

accurately determined to be 206 ± 5 and 311 ± 7 kev in good agreement with the values of 203 and 306 kev found by Arnold (1954). The peak at 523 ± 10 kev was due to simultaneous detection of these two γ-rays in the crystal. Similarly that at ~365 kev corresponded to addition of the 311 kev γ-ray and a 55 kev K X-ray. The addition peak for the 206 kev γ-ray and an X-ray would occur in the valley between the two γ-ray peaks and was therefore obscured in the present spectrum. The incompletely resolved peak close to the K X-ray peak was due to 89 kev γ-rays which were not internally converted. There was no indication of any peak not attributable to the known γ-radiation.

Since the spectrum obtained by Dixon et al. (1954) also showed an indication of a peak at ~350 kev, the alternative explanation, not ruled out by McNair (1956), that the suggested peak at 250 kev was due to addition of the 206 kev γ-ray and an X-ray seems much more reasonable. There is no indication whatsoever of a 115 kev γ-ray despite the fact that the present scintillation spectrometer is considerably more sensitive than that used by Dixon et al. (1954). In their experiment the pulses were recorded on film, and then were visually analysed in amplitude using a microfilm reader, a very tedious process. Hence in the

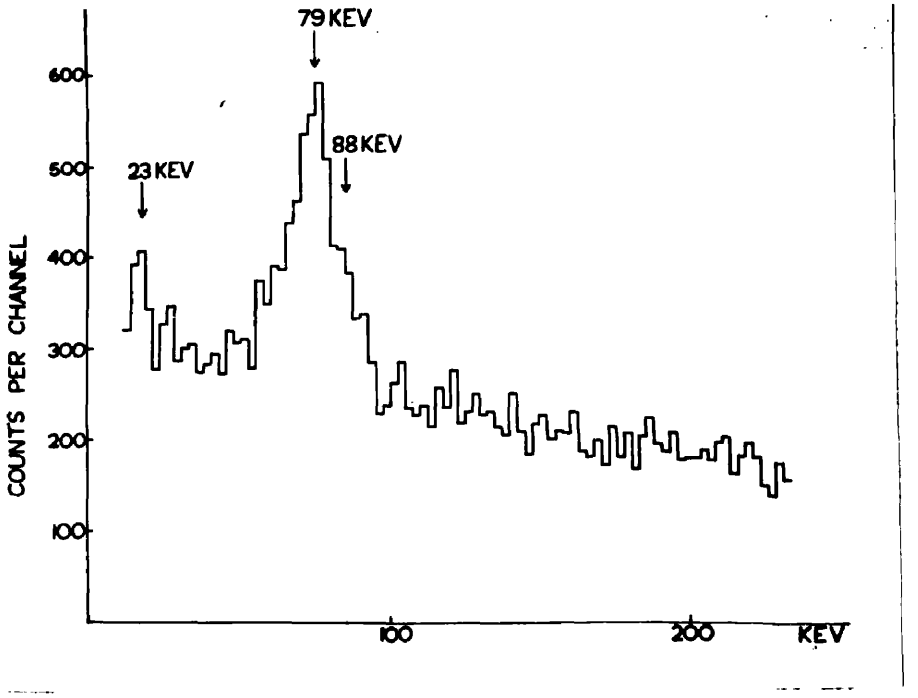

Fig.17. β-ray spectrum and electron lines of Lu^{176}.

region of 115 kev in their spectrum there were only about 30 counts per channel compared with 450 per channel in Fig.16. It is therefore the author's opinion that this suggested peak was due to statistical fluctuation. It is concluded that the γ-ray spectrum shows no evidence in favour of an electron capture branch.

3. SEARCH FOR INTERNAL CONVERSION ELECTRONS.

120 mgm of Lu_2O_3 was spread over an area of 800 cm^2 of the proportional counter lining giving a very uniform source of average thickness 0.15 mg/cm^2. The anticoincidence Geiger ring was not used since the background above 20 kev under the lead shielding was only 90 cpm compared to 250 cpm from the source. Hence the experiment was simplified since the pulses were now fed along the cable to the Hutchinson-Scarrott kicksorter, thus eliminating the tedious film analysis. Fig. 17 clearly shows the electron lines due to internal conversion of the 89 kev γ-ray in the K shell (23 kev) and L, M and N shells. There is no indication of a line at ~105 kev as suggested by McNair (1956) although a peak of intensity 5% of that at ~79 kev would have been clearly seen.

4. THE HALF-LIFE OF Lu^{176}.

The half-life was estimated from the observed γ-ray spectrum. Allowance was made for the Compton distribution due to the 206 kev γ-ray (see Fig. 16). Then the remainder of the counts in the spectrum represent the total number of 311 kev γ-rays detected by the crystal either alone or in coincidence with a 206 kev γ-ray or an X-ray. A source of potassium chloride was placed in the same geometry, and its spectrum taken. Knowing the specific γ-activity of potassium, and calculating the average thickness of the crystal, the geometry was deduced using a value for the mass absorption coefficient in NaI for 1.46 Mev γ-rays interpolated from the tables given in the appendix to Siegbahn (1955). Hence, using the mass absorption coefficient for 311 kev γ-rays obtained from the same tables, the half-life was calculated to be $(2.1 \pm 0.2) \times 10^{10}$yr. This is in excellent agreement with the value of $(2.15 \pm 0.10) \times 10^{10}$yr found by Arnold (1954), also from examination of the γ-ray spectrum, but not with that of $(4.13 \pm 0.20) \times 10^{10}$yr recalculated by McNair (1956) to replace the original value of $(4.6 \pm 0.3) \times 10^{10}$yr obtained by Dixon et al. (1954) from an examination of the

β-spectrum. The half-life was also calculated from our observed β-spectrum, but a routine check for α-activity revealed about 20 times the usual amount, so that the shorter half-life obtained (2.8×10^{10} yr) cannot be considered significant since the presence of α-contamination meant that β-contamination, which would explain the apparent reduction in half-life, could not be ruled out.

5. CONCLUSIONS.

Neither the scintillation nor the proportional counter experiment provides evidence supporting the electron capture branch suggested by McNair (1956). It is concluded, in the absence of γ-rays or conversion electrons, which would certainly be associated with a detectable electron capture branch in view of the spin change of 10 between the ground states, that a maximum of 3% of the disintegrations take place by electron capture as suggested by Dixon (1956).

There appears to be a marked disagreement between the half-lives deduced from γ and β-ray measurements. Unfortunately, for the reason given above, it was not possible in the present investigation to determine the β-ray half-life. Obviously further study of the β-rays using extremely pure lutetium is required.

CHAPTER SIX

AN INVESTIGATION OF THE ISOBARIC PAIRS (Cd^{113}, In^{113}), (In^{115}, Sn^{115}) and (Sb^{123}, Te^{123})

A detailed description of these experiments will be given in the thesis of Mr. D.E. Watt. Here only an outline is presented particularly to illustrate further the problem of radioactive contamination, the implications of which will be fully discussed in the next chapter. No evidence was obtained of instability in any of these three isobaric pairs.

1. (Cd^{113}, In^{113}).

According to the systematics of Way and Wood (1954) an energy of 250 - 350 kev should be available for β^--decay of Cd^{113} to In^{113}. The data on which this figure was based was considered by them to be reliable enough to eliminate the possibility of In^{113} being unstable with respect to electron capture. An unsuccessful search for electron capture was carried out by Heintze (1955), who claimed minimum half-lives of 10^{16}yr and 10^{14}yr for K and L-capture

respectively. No β-rays of energy >200 kev were observed by Kalkstein (1952) or Selig (1954).

In the present experiments sources of spectrographically pure cadmium oxide were inserted in the proportional counter to investigate the possibility of β-decay. No activity was observed and in consequence it was deduced that the half-life for the emission of β-rays of energy >50 kev must be greater than 1.7×10^{15} yr and of energy >10 kev must be greater than 7.6×10^{13} yr.

2. (In^{115}, Sn^{115}).

Martell and Libby (1950) reported the emission of β-rays from In^{115} of maximum energy 630 kev and half-life 6×10^{14} yr. Cohen (1951) also observed activity of unspecified energy and half-life.

Confirmation of this activity was first considered necessary and, with this intention, a source of spectrographically pure indium oxide of average thickness 2 mg/cm^2 was introduced into the proportional counter filled to a pressure of 5 atm. A β-spectrum of maximum energy and half-life similar to that reported by Martell and Libby (1950) was observed. However the routine check revealed

an α-activity of 11 cpm, roughly five times the normal value. A second spectrographically pure sample was obtained and found to give the normal α-rate. No β-activity was observed from it, although on the basis of the results of Martell and Libby a counting rate of approximately 15 cpm was to be expected. The half-life for β-rays of energy >200 kev therefore appears to be greater than 1.6×10^{16} yr. In view of the result obtained with the first sample it would seem extremely probable that the activity previously reported was due to an insufficiently pure source. At that time the extreme purity necessary was not fully appreciated.

3. (Sb^{123}, Te^{123}).

The systematics of Way and Wood (1954) indicate that an energy of 250 - 350 kev may be available for the decay of Te^{123} by electron capture to Sb^{123}. Unfortunately the experimental data on which this estimate was based was not sufficiently accurate to allow the possibility of β-decay of Sb^{123} to be eliminated. Kalkstein (1952) and Selig (1954) found no evidence of K or L-capture in Te^{123}, and an apparently more sensitive experiment by Heintze (1955) quoted the minimum half-lives as 10^{15} yr and 10^{13} yr

respectively. No β-rays of energy >50 kev were observed by Selig (1954) and hence the minimum half-life for β-emission from Sb^{123} was indicated to be 10^{13}yr.

Two methods were used for the detection of K X-rays. In the first spectrographically pure tellurium dioxide was packed round the 2 in. high x $1\frac{3}{4}$ in. diam. NaI crystal. No counting rate in excess of the background, taken with antimony trioxide round the crystal, was observed. Due to the small area available (approximately 100 cm^2) and because of the absorption of X-rays in the source and in the reflector surrounding the crystal, the half-life can only be claimed to be greater than 2.6×10^{14}yr. The experiment was repeated in the proportional counter using a source of thickness 25 mg/cm^2. Because of the much larger area available and the absence of absorber between source and detector, this method proved more sensitive in spite of the relatively low efficiency of the proportional counter even at 5 atms. for 26.2 kev X-rays. By this method a lower limit for K-capture of 2.3×10^{15}yr was deduced.

Evidence of a β-spectrum was obtained using spectrographically pure samples both of tellurium dioxide and tellurium powder. It is impossible for tellurium to decay by β-emission. It is extremely improbable that

there would be sufficient energy for β^+-emission. Indeed this possibility can be ruled out since a search for γ-activity using the scintillation spectrometer failed to reveal annihilation radiation, which, if the counting rate observed in the proportional counter was due to positrons, should have been easily detectable. The activity therefore seemed to be due to contamination. It is noteworthy that in this case the α-activity of neither sample was abnormally high.

A search for β^--activity in Sb^{123} was then undertaken. Negative results were obtained consistent with a minimum half-life of 9.4×10^{15}yr for the emission of β-rays of energy >50 kev and 4.2×10^{14}yr for β-rays of energy >10 kev.

CHAPTER SEVEN

CONCLUSION.

The geological importance of the branching ratio of K^{40} has already been indicated. Accumulation and re-evaluation of geological data by several investigators has demonstrated argon leakage from some minerals, and geologists are now prepared to accept the value of the branching ratio found in the present measurement and confirmed by recent independent measurements of the specific activity of each branch. The study of La^{138} has resulted in the detection of a previously unobserved β-spectrum. A completely new decay scheme has been established which confirms energy levels previously reported for Ba^{138} and Ce^{138} from studies of artificially radioactive isotopes decaying thereto. Otherwise negative results have been obtained with the exception of a possible γ-activity associated with electron capture in V^{50}. The sensitivity of the apparatus has, however, made it possible to quote longer minimum half-lives than those of previous investigators. It is obvious that the positive results are of some importance. The negative results have provided useful information on which to base

a reconsideration of the original query "Are the natural radioelements with Z < 80 worth further investigation?"

The negative results indicate that equipment of even greater sensitivity would be required in any future investigations. This could be achieved by using materials considerably enriched in the appropriate isotope and by a further reduction in the background. Continued use of unenriched sources is undesirable, not merely because it appears unlikely that improvement in the background alone would be sufficient to produce significant results, but also because of the difficulty of obtaining materials of the necessary purity. In the study of La^{138} considerable α and γ-activity was detected due to a minute trace of thoria still present in spectrographically pure lanthanum oxide. This result led to the establishment of a routine check on the α-activity of all sources used. Due to this precaution it was concluded that a β-spectrum previously attributed to In^{115} was probably due to radioactive impurity and that a measurement of the specific β-activity of Lu^{176} carried out by the author was of no significance. However the experiments on Te^{123} indicate that contamination is not necessarily associated with an α-activity greater than the minimum value of approximately 2 cpm observed from the

presumably cleanest materials. It is therefore fortunate that the only β-activity detected in these studies, that of La^{138}, could be amply verified since the 0.81 Mev γ-ray was calculated to have an intensity equal to that of the β-rays, and also since the β-emission from clean and contaminated lanthanum oxide was observed to be identical. It can be seen that results obtained using unenriched material require to be treated with such great caution as to render its use undesirable. In contrast, material isotopically enriched by means of the mass spectrometer should be free of radioactive contamination. Let us now consider in detail possible methods of increasing the sensitivity of the equipment. The use of enriched isotopes is obvious and requires no further comment. By remounting the NaI crystal in copper or steel instead of aluminium and by using a photomultiplier whose envelope is made of potassium-free glass, as yet not available, the sensitivity of the scintillation spectrometer in its present underground site could be increased roughly by a factor of three. However almost all the remaining problems require the use of the proportional counter. Most of its present background is believed to be due to electrons produced by the conversion of γ-rays, both from the cosmic ray flux and from

surrounding radioactive materials, in the counter wall. It is therefore extremely improbable that a large reduction in background can be achieved in the conventional proportional counter. Moljk and Drever (1956) have developed for carbon dating a large proportional counter, whose volume is defined by a cylinder of equally spaced wires running the length of the counter, surrounded by a concentric ring of proportional counters connected in parallel operating in anticoincidence with it, the ring counters and the central counter all being within the same cylindrical tube. By this method the effect of γ-ray conversion in the walls, and also of any impurity in the walls, is eliminated since an electron produced at the wall, if it penetrates to the centre counter, will have already triggered the anti-coincidence ring. The background on the surface with shielding of 2 in. lead and 1 in. steel was found to be 3 cpm, an improvement by roughly a factor of ten on the background in the mine. However this method obviously cannot be applied to a wall-mounted source. It might be possible to replace the wires defining the volume of the centre counter by a very thin support of a low Z material on which the enriched source could be mounted. The conversion of γ-rays in the source would still occur and might be

expected to increase the background slightly. However there is a much more important objection to this idea. The present studies of the isobaric pairs have shown that the energy available for each transition must be very low. (Since the log ft value of 3rd forbidden transitions is known to be approximately 19.0, the longer minimum half-lives proposed on the basis of the present experiments mean a reduction in the available energy.) The fact that the half-life of Rb^{87} found from physical measurements seems longer than that suggested by geological measurements indicates that further investigation must be made in the very low energy region because of the spectrum shape. Confirmation of the La^{138} β-spectrum shape also requires examination in the same energy range. The β-spectrum of Re^{187} was reported by Suttle and Libby (1954) to have a maximum energy of 8 kev, but its existence was not confirmed by Dixon and McNair (1954). Thus practically all the remaining problems involve investigations in the very low energy region. However the experiment designed to detect the Auger electrons of energy approximately 2.8 kev following electron capture in K^{40} was unsuccessful. Similarly Drever and Moljk (1955) failed to detect Auger electrons from an extremely thin solid source of Cl^{36}, although the

experiment was successful using a gas source. The two possible explanations are either that the proportional counter, for some unspecified reason, fails to detect low energy radiation which produces ionisation close to the wall, or that, probably due to scattering, low energy electrons seldom emerge from even extremely thin solid sources. Both explanations, irrespective of which is correct, although the latter, in the author's opinion, seems rather more probable, preclude the use of a solid source in the proportional counter. Gaseous sources would be ideal, but can only have a limited application, if any, to the investigation of very weak activities. For most of the elements requiring further study it would be extremely difficult to find a stable gas let alone one which would not poison the proportional counter when introduced at the appreciable pressure which would be required, even with enriched sources, to improve markedly on the sensitivity of the present equipment. Since the operation of proportional counters at temperatures up to 900°C has been proved feasible by Moljk et al. (1955), perhaps a large high temperature proportional counter could be developed. It may be possible to find suitable compounds, which, although solid or liquid at room temperature, exist as a vapour at temperatures at present

attainable with these counters and could be introduced into them at an appreciable pressure. It appears to the author that to increase the sensitivity of the equipment by a factor large enough to make further investigation worthwhile will be both extremely difficult and extremely expensive. In the author's opinion it is doubtful if the importance of the results which might be obtained is sufficient to justify the trouble and expense of further investigation.

A P P E N D I X

APPENDIX

SATURATION BACKSCATTERING OF β-RAYS FROM STEEL AND ALUMINIUM.

1. INTRODUCTION.

Radioactive sources are often deposited on a very thin film of a substance of low atomic number e.g. nylon, to minimise the scattering of electrons in the source support. This approach is clearly impossible in experiments involving very low specific activities since extremely large source areas are necessary to obtain reasonable counting rates. The present investigation is concerned only with saturation backscattering i.e. backscattering from backing materials of sufficient thickness ($\sim$ the range of the β-ray in the material) to produce maximum scattering of the β-rays. It is not intended to be an exhaustive study of the problem, being undertaken originally because the measurement of the K^{40} branching ratio by the first method required accurate values of the backscattering factors from steel for the Co^{60} and K^{40} β-rays, values which could not be obtained from the published data due to its inconsistency. The results have,

however, been applied in several other investigations to calculate specific β-activities.

The techniques which have been used in the study of backscattering are of two types, both involving Geiger counters. In the first method, thin end-window Geiger counters were used with the sources mounted externally. This method has been applied extensively by Zumwalt (1950), Burtt (1949, 1950), Yaffe and Justus (1949), Yaffe (1950), Glendenin and Solomon (1950) and Seliger (1950). In the second method the source was mounted internally. Christian et al. (1952) used a gas flow Geiger counter with the source mounted at one end. Only Charpak and Suzor (1952) and Seliger (1952) using 4π Geiger counters have studied backscattering in 2π geometry. There seems to be general agreement that the intensity of backscattering increases with increasing atomic number of the backing material. However there is considerable doubt concerning the variation, if any, of the intensity of backscattering with the maximum β-ray energy. The backscattering factor in the range 0.2 - 1.5 Mev was claimed by Zumwalt and by Seliger (1950) to be constant, whereas Yaffe and Burtt found that it increased with energy up to $\sim$0.6 Mev (see Fig.19) but Suzor and Charpak and Christian et al. indicated that it tended

to decrease with increasing energy. It is reasonably certain that the variation below ~0.6 Mev was due to the geometry used since it was shown by several investigators that the angular distribution of the backscattered β-rays was anisotropic. In particular, Seliger (1952) found that backscattering factors determined in 2π geometry and in less than 2π geometry did not correspond, the former being higher for low Z backings and lower for high Z. However even in investigations which agreed on the variation of backscattering with energy, the actual values of the backscattering factor for a given backing were not very consistent.

Because of these inconsistencies revealed by the literature it was felt desirable to make our own measurements on backscattering using a geometry simulating as closely as possible the wall-mounted source in the proportional counter. It was also claimed by Yaffe and Christian et al. that the presence of a thin non-conducting film, on which the source was mounted, between the source and the backing material considerably reduced the amount of backscattering. This criticism, if correct, would invalidate almost all the reported measurements on backscattering. Consequently methods were evolved by which it could be tested. Finally

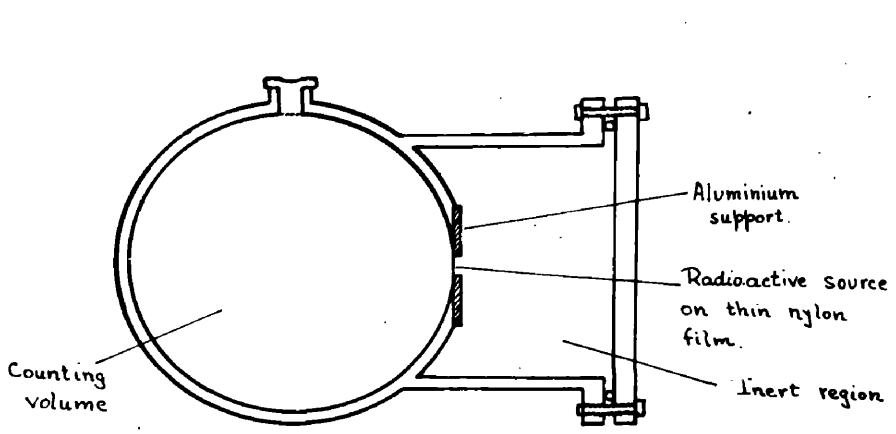

Fig.18. Diagram of the proportional counter used for the backscattering experiments.

the measurements were extended to investigate the report by Seliger (1952) that backscattering factors for positrons were about 30% lower than for electrons.

2. EXPERIMENTAL ARRANGEMENT AND PROCEDURE.

FIRST METHOD.

The counter, which is sketched in Fig.18, was very similar to that used by Balfour (1954), who investigated the scattering of electrons of energy < 100 kev by using the conversion lines of Te^{125m}. The proportional counter was of diameter 14 cm. with a 2.5 cm. diameter hole opening into a large side chamber. Over this hole could be screwed a thin aluminium sheet with a 1.3 cm. diameter hole across which was stretched a nylon film, of thickness < 10 microgm/cm^2, carrying the radioactive source. The source was thus effectively on the cylindrical counter wall i.e. in 2π geometry. Metal plates could be screwed tightly against the source to act as backscatterers. This method was considered better than the mechanical device for changing the backing metal used by Balfour (1954) since it ensured good contact between source and backscatterer. Access was obtained via the side chamber to change sources and

backscatterers. The counter was filled with 15 cm. pressure of methane, low pressure and a gas of low atomic number being used to render negligible scattering of β-rays from sources mounted on nylon back into the sensitive volume of the counter by the gas in the side chamber. It was checked experimentally that no correction was required for this effect or for the possibility of β-rays entering the counting volume after scattering at the walls of the side chamber. The energy resolution of the methane-filled counter was rather poor. However this was of no great importance since the counter was being used merely as a detector of β-rays and not as a spectrometer. Sufficiently good energy calibration was obtained by using the edge of the 8 kev fluorescence X-ray line of copper. Tests with the same source showed that counting rates were accurately reproducible after refilling the counter. To meet the suggestion that the backscattering factor was altered by the presence of a thin non-conducting film between the source and the backscatterer the source was reversed so that it was in direct contact with the backscattering material i.e. with the nylon film between the source and the sensitive volume of the counter. It was shown that no appreciable backscattering or absorption was caused by the nylon, the

change in counting rate on reversal of the source being always $<1\%$.

Counting rates were taken with and without thick metal backings, all pulses of energy >200 ev being recorded, and calculation was made of the saturation backscattering factor, defined as

$$\frac{\text{increase in counting rate with backscatterer.}}{\text{counting rate without backscatterer.}}$$

Steel and aluminium backings were used with sources of the β^--emitters Co^{60} (0.306 Mev), Na^{24} (1.39 Mev), Sr^{89}(1.46 Mev) and P^{32} (1.7 Mev), and with the β^+-emitter Na^{22} (0.542 Mev). The figures in brackets are the maximum energies of the β-spectra.

SECOND METHOD.

This procedure was devised to enable backscattering factors to be measured when the radioactive source was deposited directly on the backing. β-emitters, whose decay scheme contained a high energy γ-ray, were used. If the energy of the γ-ray was >100 kev, the detection efficiency of the proportional counter at the pressure and with the gas used was negligible, and the counting rate was

due only to the β-rays. Sources of approximately equal strength were prepared on nylon, steel and aluminium. The β-ray counting rates were measured in the proportional counter. The relative strengths of the sources were determined by placing each in turn in a fixed position relative to a NaI scintillation counter biassed at approximately 100 kev. To keep the experimental conditions identical during the γ-ray measurements the source mounted on nylon was backed by a sheet of the backscatterer under examination of the same thickness as that on which the source had been directly deposited.

Then if 'a' and 'b' are the β-rates and 'x' and 'y' the γ-rates of a source deposited on a backing and on nylon respectively, the saturation backscattering factor is given by

$$\frac{a/x - b/y}{b/y}$$

In these experiments Na^{22}, Na^{24} and Co^{60} were used, again with steel and aluminium backings.

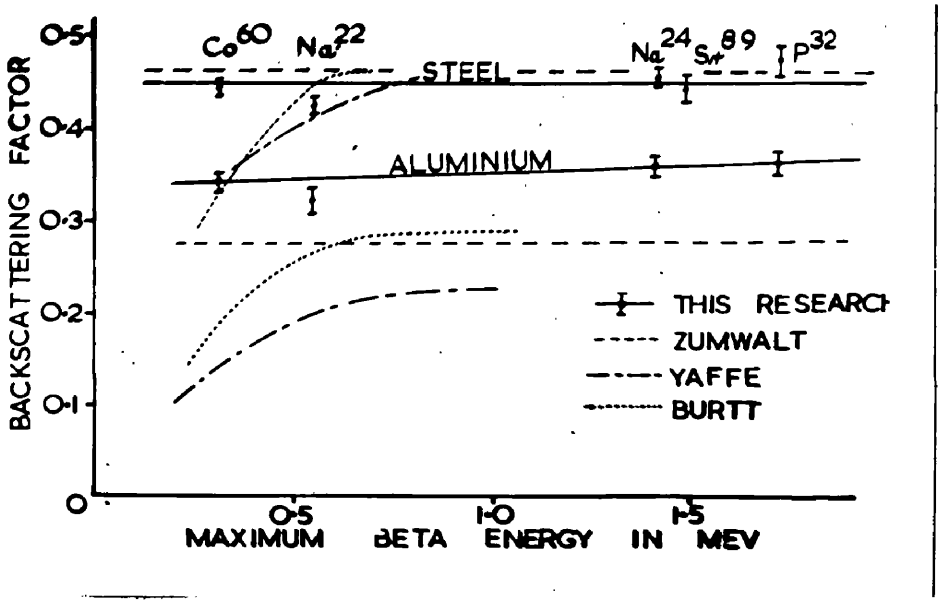

Fig.19. Variation of backscattering with energy for steel and aluminium.

3. RESULTS AND DISCUSSION.

The results obtained by these two methods were in good agreement. The amount of backscattering from steel was found to be independent of energy in the range investigated. The average of all the measurements on all the sources gave a backscattering factor of 0.450 ± 0.015 i.e. 45% of the electrons entering a thick steel support are reflected back into the counter. For aluminium there was some slight evidence of a slow increase of the amount of backscattering with β-ray energy. The average backscattering factor was 0.35 ± 0.03.

The results are shown in Fig. 19 together with, for comparison, those of some previous investigations. The results for steel show reasonable agreement except at lower energies. Since several investigators have shown that low energy electrons tend to be scattered in a direction tangential to the backing the fact that the geometry previously used was less than 2π is probably sufficient explanation. The results for aluminium show considerable variation. However practically the same explanation would suffice as it is known that backings of low atomic number give an excess of low energy β-rays in a tangential direction.

Several experiments were carried out with the nylon film between the source and the backing metal. The results obtained agreed with those of the two methods described. Hence it is concluded that the occurrence of a thin non-conducting film between source and metal does not affect the amount of backscattering.

For both steel and aluminium and in both methods, the positron emitter Na^{22} gave a backscattering factor approximately 5% lower than the average values. There was no evidence of the marked difference ($\sim$30%) between positron and electron backscattering factors reported by Seliger (1950, 1952).

In conclusion, it is felt that backscattering factors are best determined by each investigator by whom they are required, especially if the geometry is less than 2π or the source not particularly thin.

PRELIMINARY INVESTIGATIONS OF Os^{187} AND Ta^{180}.

Since β^--emission from Re^{187} was not observed by Dixon and McNair (1954), it was decided to carry out a sensitive search with the scintillation spectrometer for 60.4 kev X-rays of rhenium which would follow electron capture in Os^{187}. Preliminary results showed no evidence of electron capture.

Exploratory experiments which were undertaken on Ta^{180}, the recently discovered middle member of the triad (Hf^{180}_{72}, Ta^{180}_{73}, W^{180}_{74}) have given no evidence of the emission of either β or γ-rays or of X-rays attributable to electron capture.

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