A STUDY OF SOME NATURAL RADIOELEMENTS OF LOW SPECIFIC ACTIVITY.

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P R E F A C E

This thesis contains the results of work carried out in the Department of Natural Philosophy of the University of Glasgow during the three academic sessions from October, 1951, until October, 1954. The field of research was proposed by Dr. S.C. Curran, F.R.S. and it was carried through under his general supervision. It consisted of an investigation of the radio-active properties of some naturally occurring isotopes, lying in the lower part of the periodic classification of the elements. The isotopes chosen for study were suspected to be radio-active either from nuclear stability rules or from information from earlier reports.

The introductory chapter of the thesis sets out the peculiar position occupied by these long-lived nuclei in nuclear theory and in certain branches of geology. The slow rates of disintegration of the isotopes pose difficult problems in measuring techniques. The development of suitable apparatus capable of giving the accuracy required in the work is described in the second chapter. This development was already under way when the author began his period of research and he assisted Mr. D. Dixon in the construction and testing of the counter and associated electrical equipment. No claim of originality is made by the author for the apparatus described in Chapter 2 apart from the design of the improved type of counter. For this reason the description of the apparatus is brief and only included for completeness. Similarly, no account has been taken of the electronic circuitry, so necessary an adjunct to proportional

counters, since the designs were borrowed from units which had already proved their worth in proportional counter spectrometry.

The next three chapters describe separate studies of the properties of particular isotopes. This work was done in collaboration with Mr. Dixon. The results of the investigations, published at various times during the period of research, have been brought up to date by the author for the purposes of this thesis. Often they incorporate new information, both from outside sources and from later work done by the author. Because of this, many of the conclusions reached and suggestions made for improvement in experimental technique are the author's own. Where possible, these new conclusions are specifically pointed out in the text. A few of the preliminary measurements, with scintillation counters, of the energies of the gamma rays emitted in the decay of Lutetium-176 (described in Chapter 5) were made with the assistance of Mr. (now Dr.) A.T.G.Ferguson.

Chapters 6 and 7 describe original work of the author in which he was assisted in the experimental work by Mr. R.N. Glover. The investigation of the decay of Potassium-40, an important natural radio-isotope, was undertaken following the suggestion made by Sir Edward C. Bullard to Dr. Curran that a re-evaluation of the branching ratio was extremely desirable to help in settling discrepancies in the geological dating of potassium-bearing minerals. The experience of Dr. H.W. Wilson was available to the author in this investigation.

The final chapter contains a general summing-up of the work and some comments on what the author feels are the important points left

unsolved.

Each isotope or group of isotopes pose problems peculiar to themselves, and the previous knowledge about the behaviour of the isotopes varies considerably. Because of this, each chapter is almost complete in itself and for ease in making reference to the work of others, lists of references quoted in each chapter are given separately at the end of the book.

ACKNOWLEDGEMENTS.

The author would like to take this opportunity to thank Professor P.I. Dee, C.B.E., F.R.S., for his interest and encouragement and also Dr. S.C. Curran, F.R.S., for much helpful comment and discussion throughout the course of this work.

Thanks are also due to Mr. J.T. Lloyd for assistance and advice in the manufacture of some of the apparatus, and to the staff of the electronics laboratory and mechanical workshop for similar services.

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A B S T R A C T

A description is given of a low background proportional counter system suitable for the examination of long-lived radio-active isotopes occurring in nature, or of other artificially produced isotopes with a low specific disintegration rate.

An investigation of neodymium shows no evidence for the reported beta-activity and it is concluded that Nd^{150} is stable against single beta decay. A slight alpha-activity, which may be characteristic of Nd^{144} is observed.

No experimental proof of instability in either of the 'neighbouring' isobars Re^{187} and Os^{187} is found. Since recent geophysical work suggests that Re^{187} is beta-active, it is concluded from the results of the study that the energy of the electrons released in the transition is very small, of the order of 1 Kev or less. Assuming a first-forbidden transition this suggests that the half-life of Re^{187} is 10^{11} years or greater.

The emission of a low intensity L x-radiation from heavy elements, under bombardment by cosmic or other background radiation, is noticed. The effects on the ultimate sensitivity of the proportional counter and some methods of mitigating the interference are considered.

The radio-active transitions undergone by Lu^{176} are studied in detail. The beta decay is found to have a half-life of

$(4.13 \pm 0.20) \times 10^{10}$ years and seems most likely to correspond to a third-forbidden transition with a spin change of 4 units. The maximum energy of the beta spectrum is 425 ± 15 Kev. The energies of the three excited states of Hf^{176}, occupied in succession by the decaying nucleus, confirm predictions from the Bohr-Mottelson rotational model of the nucleus. Strong electron peaks from the internal conversion of the 89 Kev gamma ray of Hf^{176} and internal conversion electrons from 190 ± 10 Kev and 310 ± 10 Kev gamma rays permit classification of the transitions as E.Q. The intensity of the L x-ray emission from Lutetium is consistent with the presence of an electron capture mode of decay, amounting to (9 ± 1) % of the beta transitions. Some evidence for the emission of gamma rays following electron capture in Lu^{176} is demonstrated.

The ratio of the rates of emission of gamma rays and beta rays from K^{40} is re-measured. Two separate investigations give values of 0.124 ± 0.002 and 0.121 ± 0.004 respectively, confirming the discrepancy between recent physical and geological work on the K^{40} radio-active transition. The half-life of the isotope is estimated to be $(1.28 \pm 0.02) \times 10^{9}$ years. The energy of the gamma ray is confirmed to be 1.46 ± 0.01 Kev. New coincidence experiments to check the accepted decay scheme of K^{40} are described.

Experiments to determine the amount of backscattering of electrons and positrons from thick sheets of steel and aluminium into 2π solid angle are also treated.

LIST OF CONTENTS

1. THE NATURAL RADIO-ACTIVE ELEMENTS IN NUCLEAR PHYSICS AND GEOLOGY.

This thesis describes and interprets some new investigations of a number of naturally occurring radio-active elements. The work was a contribution to a general study, which is being made at Glasgow, of the radio-active properties of those natural radio-elements with atomic number less than 80. Interest in these elements has been growing in recent years, not only because of their significance for fundamental nuclear theory, but also for their increasing importance in geophysical work.

THE NATURAL RADIO-ELEMENTS.

The process of beta-decay involves a nuclear transformation between two 'neighbouring' isobars. A consideration of the energy released in the beta transition led Mattauch (1) to formulate his 'isobar rule'. This shows that where two neighbouring isobars are known to exist in nature, one of the pair must be unstable with respect to the other. There are four known examples of three neighbouring isobars occurring naturally. These isobaric triplets are found

at	A	=	40	A	, K	, Ca	(Z = 18, 19, 20)
at	A	=	50	Ti	, V	, Cr	(Z = 22, 23, 24)
at	A	=	138	Ba	, La	, Ce	(Z = 56, 57, 58)

at A = 176 Yb , Lu , Hf (Z = 70, 71, 72)

There are also a few cases where pairs of adjacent isobars exist, as

at A = 87 Rb , Sr , (Z = 37, 38)

at A = 113 Cd , In (Z = 48, 49)

at A = 115 In , Sn (Z = 49, 50)

at A = 123 Sb , Te (Z = 51, 52)

at A = 187 Re , Os (Z = 75, 76)

By Mattauch's rule, at least one of each of the first group and one of each of the second group of nuclei must be unstable. In the first group ^{40}K and ^{176}Lu are known to be unstable and are discussed later. Radio-activity has been found in ^{138}La (2-4) but none has yet been detected in the A = 50 group (5-7). In the second group of nuclei ^{87}Rb is known (8-13) to decay by negatron emission to ^{87}Sr, the transition going from ground state to ground state with a spin change of 3 units and parity change. ^{113}In has been reported radio active by one experimenter (14) but this has been denied by another (7). A phenomenon observed in the present work suggests that the evidence for the instability of ^{113}In might have been caused by other factors. This point is discussed in Chapter 4. No activity has been detected in ^{113}Cd (15) nor in the A = 123 group (15, 7). ^{115}In has been reported (14, 16) to show negatron activity with a long half-life. The final group, ^{187}Re and ^{187}Os is the subject of experimental work

which is described in Chapter 4, and the whole question of the mode of decay is discussed there.

The possible beta-activity of Nd^{150} which has been reported (17) raises the question of the occurrence in nature of an isotope of the 'missing' element, promethium. Neodymium has also been investigated in this work and is treated in a more adequate manner in Chapter 3.

THE FUNDAMENTALS OF BETA-RADIOACTIVITY.

Following on the introduction of the neutrino hypothesis by Pauli to explain apparent inconsistencies in the momentum and energy relations of the particles involved in the beta transformation, Fermi (18) formulated his theory of beta decay. The nucleus is treated as a composition of neutrons and protons only, and the electron and neutrino which are emitted during the transformation are presumed to be created at the moment of emission, in a manner analogous to the creation of a photon at the moment of its emission from an atom. By introducing a new interaction between the nucleon and the two light particles (the electron and neutrino) taking part in a beta transition Fermi was able to calculate the probability of the process. Five interactions are possible, scalar (s), polar vector (v), tensor (T), axial vector (A) and pseudoscalar (P). Any one, or linear combinations of any or all of these interactions

satisfy the theoretical conditions, so that Fermi's mathematical formulation is not completely unique. Any further narrowing of the choice of interactions is beyond the scope of pure beta decay theory, and the main effort of recent experimental work has been aimed at deciding which combinations of interaction do, in fact, occur.

The probability P (W) of a beta particle, with energy between W and W+dW, being emitted is of the form

$$P(W)dW = G^2 |M|^2 F(Z,W).(W_o-W)^2 (W^2-1)^{\frac{1}{2}} W dW$$

where W_o is the maximum energy of the electrons which can be emitted in any transition. Both W and W_o include the rest energy of the electron and are expressed in units of $m_o c^2$. The factor $W(W_o-W)^2(W^2-1)^{\frac{1}{2}}$ is purely statistical, while F(Z,W) is the Fermi function correcting for the effect of the nuclear Coulomb field on the energy of the emitted particles. The quantity G^2 includes a factor representing the strength of the coupling giving rise to the transition, and is regarded as a constant or nearly so. M represents a nuclear matrix element involving the wave-function of the initial and final nuclear states and an operator which produces the transformation from one state to the other. The type of operator depends on the particular form of the interaction. The ambiguity is therefore introduced into the expression

by the factor $|M|^2$. If the form of $|M|^2$ is known, then the energy spectrum of the beta rays is uniquely defined by the above equation.

THE ALLOWED TRANSITIONS.

Transitions between the initial and final states of a decaying nucleus are defined by the change in spin and parity which takes place. For certain transitions the nuclear matrix elements become independent of energy and the shape of the beta spectrum of such 'allowed' transitions is controlled mainly by the statistical factors. These allowed transitions are defined by special 'selection rules' which vary, depending on which form of interaction is assumed. The allowed selection rules are:-

(a) for S interaction, $\Delta I = 0$, no (i.e. no change in parity)
(b) for V interaction, $\Delta I = 0$, no
(c) for T interaction, $\Delta I = 0, 1$, no ($0 \rightarrow 0$ forbidden)
(d) for A interaction, $\Delta I = 0, 1$, no ($0 \rightarrow 0$ forbidden)
(e) for P interaction, $\Delta I = 0$, yes.

The rules (a) and (b) were favoured by Fermi in his original presentation of the theory. The rules (c) and (d) were introduced by Gamow and Teller (19), and are known as the G-T selection rules.

Since the nuclear matrix elements are energy-invariant in allowed transitions the shape of the allowed spectra

can give no direct information about the form of the interaction generating the decay, although Fierz (20) has pointed out that the 'statistical' shape of allowed beta-spectra may be modified if both S and V or if both T and A interactions are present, since the allowed radiations they generate can interfere with each other.

FORBIDDEN BETA TRANSITIONS.

In some transitions which are energetically possible, the selection rules for allowed beta decay do not hold, As is to be expected in quantum theory, such transitions can occur, but with a much reduced probability. The matrix element defined by the first order forbidden selection rules is expected to be a factor of 10 less than the matrix element for allowed transitions, so that the decay probability is depressed by a factor of about 10^{2}. More important, the new matrix elements have a definite dependence on the energy of the emitted electrons. The exact form of the energy dependence is a function of the type of interaction which is assumed. If neither allowed nor first forbidden selection rules are obeyed, matrix elements and selection rules characteristic of second, third and higher order forbidden transitions may be obtained. The matrix elements of all-order forbidden transitions are

expected to vary with energy in a manner depending on the specific form of interaction occurring in the beta process, and also to show a progressive decrease in intensity as the order of forbiddenness increases. Since the matrix element now depends on the type of interaction, the shape of the beta spectra of forbidden decays will also depend on which interaction generates the transition. Experimental and theoretical investigation of the shapes of forbidden spectra may therefore elucidate the interaction or combination of interactions which generate beta radio-activity.

Unfortunately, in many cases, theoretical calculations do not predict unique spectrum shapes and the results for several combinations of interactions may be adjusted to fit experimental spectra. To narrow the choice it is important to obtain as much empirical information about the spectra of forbidden transitions as possible. This is where a systematic study of the naturally occurring radio-active elements becomes of importance, because most of the natural beta-active nuclei outside the uranium-lead section of the periodic table are expected to undergo highly forbidden transitions. Even when the spin change which occurs in a natural transition is unknown, it is almost necessary to assume a highly forbidden decay to explain the persistence of the unstable nucleus in nature to the

present day. It is, of course, possible that the energy available for a certain beta transition may be so small that the disintegration rate is depressed sufficiently to explain the survival of detectable amounts of the isotope, without requiring a high spin change to slow the transition. It now seems that this may be true for rhenium - 187 (see Chapter 4) but, in general, a study of the beta activity of the natural radio-elements implies the study of highly forbidden transitions.

THEORY AND EXPERIMENT IN BETA DECAY.

A systematic interpretation of the experimental findings in terms of beta decay theory was hampered by the lack of a suitable nuclear model capable of providing information about the spin and parity changes involved in the transitions. The introduction, in 1949, by Mayer (21,22) and, independently, by Haxel, Jensen and Suess (23,24) of the highly successful spin-orbit interaction theory of nuclear shall structure, based on a hypothetical picture of the nucleus as an agglomeration of non-interacting particles in a potential well, changed the situation. By 1952, Wu (25) was able to show that the shapes of the allowed spectra argued against any appreciable Fierz interference effects, and that both Fermi and G-T interactions were

necessary to explain the experimental results. This means that either S or V with either T or A interactions generate beta transitions, with the presence or absence of P interaction not proved.

At this time the only third-forbidden beta decays that had been investigated for spectrum shape were ^{40}K and ^{87}Rb, although the 'comparative half-life' or ft values of ^{176}Lu, ^{187}Re and possibly also ^{150}Nd (12) suggested that these nuclei, too, might undergo third forbidden transitions. ^{40}K in its decay to ^{40}Ca suffers a spin change of 4 units and, according to the shell model, a change of parity. It is therefore a parity favoured transition, third forbidden only on G-T selection rules, and has been shown (26-29) to yield the spectrum shape predicted for such transitions. The decay of ^{40}K therefore gives strong support to the necessary presence of G-T interactions (T or A) but the selection rules obeyed by the transition so reduce the probability of any contribution from S, V or P interactions that nothing can be said about their presence.

On the other hand, the ^{87}Rb decay corresponds to a third forbidden beta decay with a spin change of 3 units and parity change. This type of transition is third forbidden on both Fermi and G-T selection rules. The observed shape of the beta spectrum (12, 13) can be explained by third forbidden

tensor or polar vector interaction, but by no other (30). This decay therefore suggests the presence of either or both of T and V interactions.

In conclusion, it may be stated that the latest evidence at the time of writing seems to require a linear combination of scalar and tensor interactions to generate beta transitions (31). The evidence suggests that each interaction contributes about equally to transitions which can be generated by both.

THE NATURAL RADIO-ELEMENTS IN GEOLOGY.

Until recent years the determination of the age of mineral deposits by radio-active means depended on methods based on the uranium and thorium series of natural radio-elements. Fairly successful extensions of the techniques have now been made to include the use of other natural radio-active elements. Of the natural radio-nuclides listed at the beginning of this chapter, five, ^{40}K, ^{87}Rb, ^{138}La, ^{176}Lu and ^{187}Re have at various times been considered as possible dating elements. ^{40}K and ^{87}Rb have received most attention, and the wide terrestrial distribution of potassium minerals in particular will permit future age determinations to be made over a much greater variety of deposits than has been possible in the past.

The decay of ^{138}La is now considered to be too slow for the element to be useful geologically, after the failure of attempts by Saito et al. (32) to detect excess ^{138}Ba in old rare earth minerals. ^{176}Lu is considered by Arnold (33) to have a short enough half-life to be of possible use, especially as the daughter nuclide ^{176}Hf has a low relative isotopic abundance. The recent discovery by Hintenberger et al. (34) of radiogenic ^{187}Os in rhenium-bearing molybdenite opens the way to the use of the ^{187}Re radio-active transition in geological work.

METHODS BASED ON ^{87}Rb.

The earlier strontium age determinations were based only on chemical analysis of the rubidium and strontium contents of the minerals, and by 1949 Ahrens and collaborators (35-38) were using spectrochemical analysis to obtain, for a number of lepidolites, ages which were believed to be close to the true values in most cases. Mass spectrometric analysis has also been used to determine the radiogenic ^{87}Sr content of the strontium extracted from the minerals (38,39) and recently (40-42), the introduction of isotopic dilution measurements of the rubidium and strontium contents of samples has made the method sensitive enough to apply to commoner types of minerals, in which the content of non-radiogenic strontium is

appreciable. However, at present, data is available mainly for the rarer lepidolites, and, because of the slowness of the $Rb^{87} \longrightarrow Sr^{87}$ transition, the method is useful more particularly for older minerals.

It is interesting to note that, on the accepted value of about 6×10^{10} years for the half-life of Rb^{87} (10-13), the strontium method of geochronometry gives the greatest ages so far discovered (43). There is some evidence that strontium ages are consistently higher than those given by other radio-active methods, so much so, that Holmes (44) considers that there is a systematic error in the technique. Kohman (43) suggests that this could be explained if the accepted half-life of Rb^{87} is too large. Although it is possible, because of the large number of low energy electrons in the beta spectrum of Rb^{87} (12,13) that errors in the experimental determination of the half-life may be sufficient to explain the discrepancies, Kohman (45) has proposed that the transition may be speeded by the occurrence of a proportion of unobserved 'bound' beta disintegrations. It is postulated that in these events the electron is created directly into an atomic orbit of the disintegrating atom, and that the excess energy is carried away by the neutrino. This point is raised again later.

POTASSIUM GEOCHRONOLOGY.

The isotope K^{40} has alternative modes of decay, either to A^{40} by electron capture or to Ca^{40} by beta emission (46). The transition to A^{40} has been the focus of a considerable amount of recent research. As the basis of a means of dating minerals, the electron capture branch of the decay is important in several respects. The parent and daughter nuclei are quite distinct chemically and physically offering, in principle, easy methods of separation. Also, it is unlikely, since argon is an inert gas, that there will be any appreciable amount of non-radiogenic argon trapped in the samples.

Although considerable experimental difficulties are experienced in the extraction and measurements of small quantities of argon, most of these now seem to have been overcome (47), and precise argon age determinations have been held up mainly because the decay period of the $K^{40} \longrightarrow A^{40}$ transition has not been known with sufficient accuracy. An exact determination of the branching of the decay has proved difficult to obtain. The most reliable of the recent measurements of the branching ratio (the ratio of the rate of decay to A^{40} to the rate of decay to Ca^{40}), using radiation counting methods, give values around 0.12. The branching

ratio has also been measured by geophysical methods using minerals of known age. Recent values obtained in this way tend to be about 0.09. The discrepancy is sufficiently wide to lead to possible serious errors in age determinations. A reason for the geophysical measurements giving a lower value may be sought in incomplete argon extraction, but the possibility of argon diffusion from the minerals cannot be discounted.

On the other hand, physical determinations of the branching ratio are difficult, and the results show sufficient variation to leave the correct value in some doubt. Thus the probability of errors in the previous determinations seemed to be high enough to warrant a re-investigation of the problem using the most sensitive apparatus available. This is all the more necessary because present indications are that, provided the remaining difficulties can be overcome, the potassium-argon method of determining ages will become very useful in future. More detailed discussion of the problems involved in the elucidation of the potassium decay and the potassium-argon dating technique is given in the introduction to Chapter 6.

The remainder of this report discusses investigations of the radio-active properties of the naturally occurring

nuclei K^{40}, Nd^{150}, Lu^{176} and the isobaric pair Re^{187} and Os^{187}. The decay of K^{40} is treated entirely from the point of view of its geophysical utility, because the beta decay of K^{40} has been adequately examined by others. The other nuclei are of interest for the possible high forbiddenness of their beta transitions. Lu^{176} also proves to be useful as a test of theories of nuclear structure, a topic which has been touched but briefly in this introduction. Lu^{176} may also be useful geologically, and, since it is the central member of a naturally occurring triad of isobars and may have alternative modes of decay open to it, the branching ratio of the decay and its half-life are of prime importance.

The problem of predicting the energy available for beta-decay has not been treated in the foregoing sections but the author has previously considered the question in an unpublished review, following the method of Kohman (48). Kohman's treatment is purely empirical, and he plots stability limits for beta decay from experimental data. Unfortunately, the curves which are obtained in this way are not sufficiently unambiguous to permit better than qualitative predictions, and in certain cases, especially where the energy for beta decay is small, even qualitative predictions are not possible. This proved to be particularly the case for the Re^{187} - Os^{187} isobaric pair, but this is

discussed later. The energetics of beta decay is not discussed further here, although reference to the detailed nuclear stability charts produced by the author for the above-mentioned review is made from time to time in later sections.

2. APPARATUS FOR THE EXAMINATION OF WEAK ACTIVITIES.

Introduction.

The statistical nature of the background counting rate of any type of nuclear counter tends to mask the presence of an additional weak activity. It is therefore essential to obtain as large a source/background counting rate ratio as possible. This aim may be approached in two different ways, either by increasing the amount of material examined, or by reducing the background. The amount of sample cannot be increased indefinitely because of the serious absorption of beta rays in thick sources. One method which has been used to obtain a reasonably large sample in a thin layer is to deposit the source on the inner surface of the wall of a cylindrical counter. This technique was employed by Libby and others (1-3) in Geiger counters and has recently been extended successfully by Curran et al. (4) to proportional counters. The use of a wall-mounted source in a proportional counter is attended by difficulties which do not arise in Geiger tubes, because in proportional spectrometers it is essential that the beta rays be totally absorbed within the sensitive region. In usual methods of proportional counter spectrometry it is frequently possible to apply a magnetic field to contain the particles within the counting volume (5)

but this is of no value when wall mounted sources are used. Even for moderate beta ray energies a large counter under pressure is essential.

A slight improvement in the counting rate as a function of source thickness may also be obtained by careful choice of chemical compound for the sample, or where possible, by the use of the pure element. A large improvement can sometimes be made if materials enriched in the radio-isotope are tested, but this is an expensive procedure and was not employed as a normal method of approach in the present work.

To reduce the background, the counter was operated in a heavily shielded underground room, with extra shielding of four inches of lead and additional steel. The portion of the cosmic radiation which could still manage to penetrate the shielding was largely removed by an array of Geiger counters operated in anti-coicidence with the main (proportional) counter. The separate units of the apparatus will now be described more fully.

THE PROPORTIONAL COUNTER.

This is shown diagrammatically in Fig. 1. The counter, made of copper tubing, with brass end-plates, is 14cm. in diameter and 28 cm. in active counting length, the total

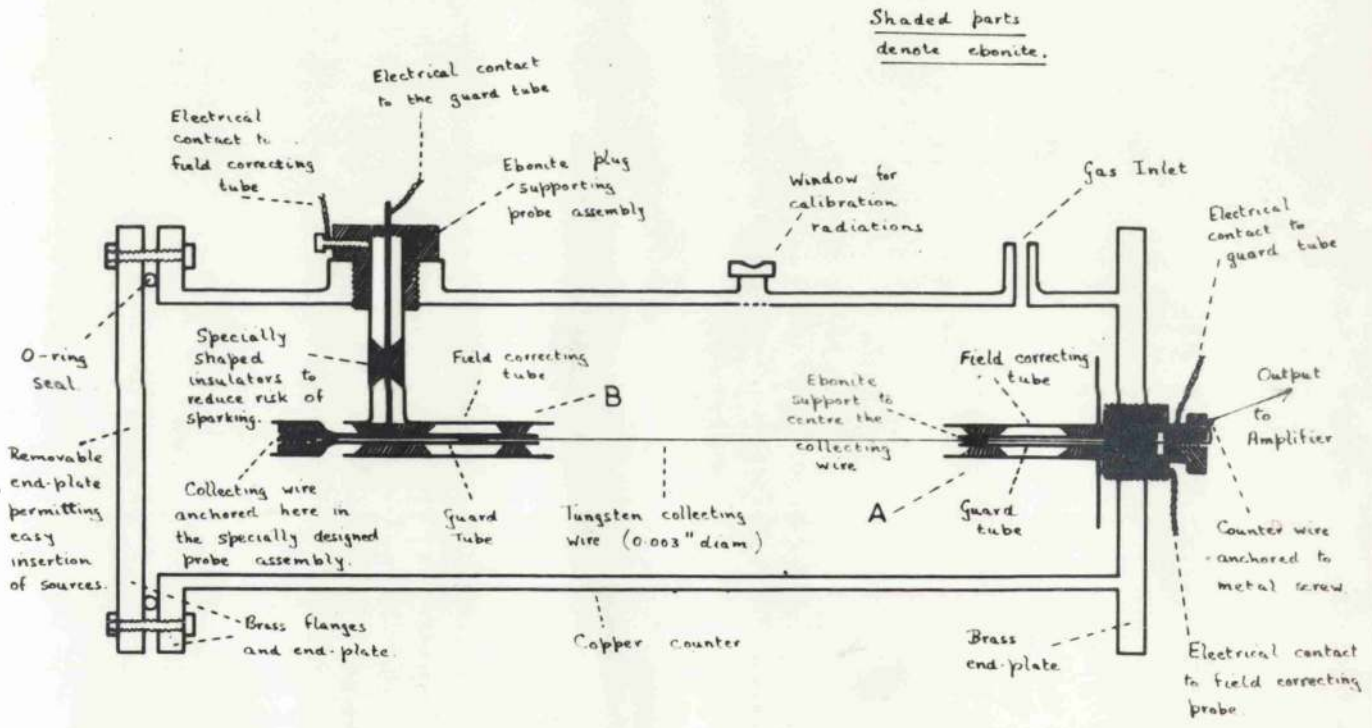

FIG. 1. DIAGRAM OF PROPORTIONAL COUNTER (NOT TO SCALE) SHOWING SPECIAL METHOD OF ANCHORING THE COLLECTING WIRE.

length being 55 cm. The tungsten collecting wire, lying along the axis of the cylindrical counter, is 0.003" in diameter, and is supported in the probe assemblies A and B. It is anchored and held taut by a small spring in B, and passes through the probe A directly to the grid of the first amplifying valve. A high negative voltage is applied to the outer case of the counter and the wire is near earth potential. In this way a high voltage D.C. blocking condenser between the counter and amplifier is unnecessary.

The probe assemblies are each essentially two co-axial brass cylinders imbedded in ebonite insulators. The outer cylinder is held at the potential appropriate to its position in the electric field inside the counter, and acts as the field correcting tube (6) defining the counting volume accurately. The inner cylinder carries the collecting wire, and is earthed to protect the wire from leakage currents from the field correcting tube and the counter.

To allow the counter to be opened for insertion of metal linings carrying the radio-active samples without breaking the collecting wire, the probe assembly B is of special design. The details are brought out in Fig. 1. but, essentially, the probe is supported in ebonite at the cylindrical wall of the counter, instead of being on the end-plate in the

normal arrangement. This design has the merit of leaving the source completely open to the sensitive region of the counter, unlike the arrangement used by Mulholland and Kohman (7) in which the wire was supported at end B by three rods stretching across the counter from end A. The rods screened some of the source and must also have had a disturbing effect on the electric field, and hence on the gas gain in the counter.

In practice, the probe B was easily disturbed mechanically and its electrical properties were not all that could be desired. In particular, there was a tendency for sparking to take place across the surface of the insulators, probably initiated by dust. An improved performance was obtained after the insulators were shaped to have slanting faces, and so presented a longer path length for a spark to traverse, but the performance was never completely satisfactory. For some of the later work the probe B was redesigned by the author to form a more robust arrangement. The new counter is shown in Fig. 2. In this, the special probe is supported in a rigid metal post fixed to the wall of the counter. The external electrical contacts are made through cylindrical springs fitted in ebonite plugs in the end-plate. When the counter is opened the springs come away with the end-plate. When it is closed they press tightly against two flat metal

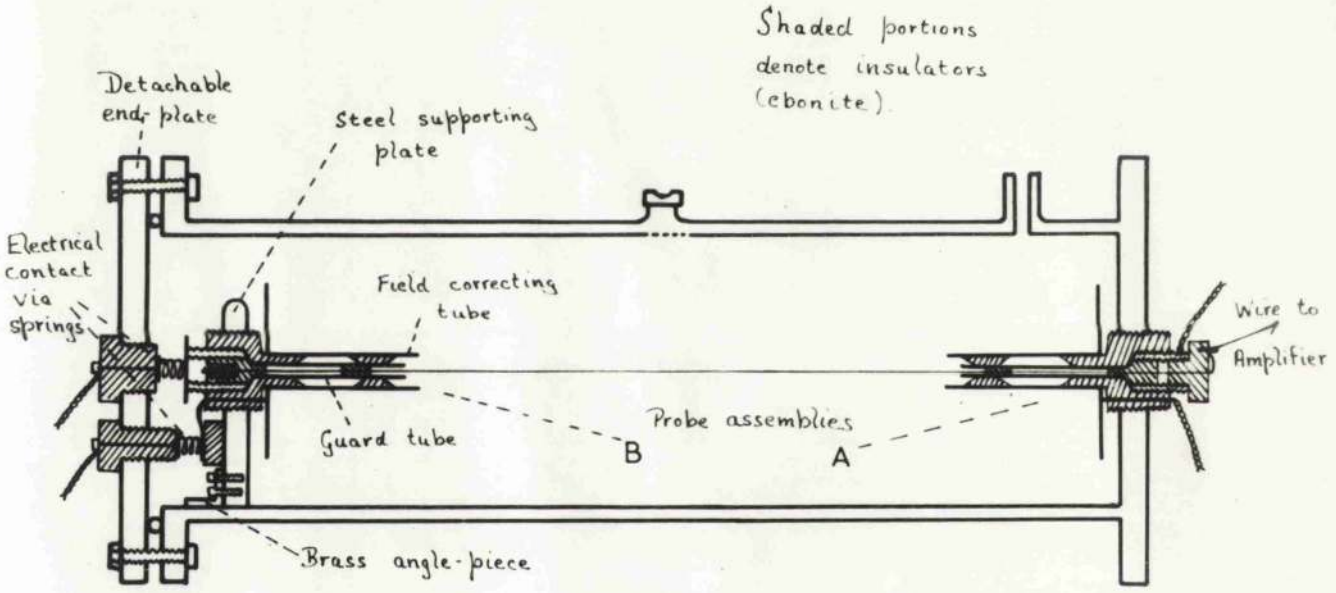

FIG. 2. DESIGN OF IMPROVED VERSION OF PROPORTIONAL COUNTER (DIAGRAM NOT TO SCALE).

plates on the probe assembly, giving the necessary connections. This design gives an assembly which is mechanically rigid and, in so far as limited experience of it under working conditions can tell, electrically sound as well.

RESOLUTION AND STABILITY OF THE COUNTER.

For a given expenditure of energy in the counter, the pulse finally arriving at the amplifier depends on statistical fluctuations in the number of electrons liberated in the primary ionising event, on statistical fluctuations in the number of electrons reaching the region of electron multiplication near the wire, and on variations in the multiplication process itself. Of these, the only effect likely to be more serious in a large counter is the second, because many of the electrons have a comparatively long distance to travel in regions of low electric field strength before they reach the high field region close to the wire. This increases the time that the free electrons spend in the gas and increases the probability of formation of heavy ions. Since the drift velocity of gaseous ions may be a thousand times smaller than the drift velocity of free electrons the rise time of the collection pulse is lengthened and the height that the pulse finally attains is reduced. To prevent this as much as possible, gases with a high electron affinity must be excluded from the counter. In practice, mixtures

of argon and methane of commercial quality were found to be suitable for the size of the counter used in the present work. Even so, extreme care had to be taken to exclude traces of organic vapours which could be present in the pumping systems or in the source, if organic materials were used in its preparation. Because of the immense area presented by a wall-mounted source to the counter even slight emission of organic molecules may 'poison' the counter. In particular, xylene was found to have disastrous effects on the operation of a proportional counter of this size, although the same gas mixture in a small (1" diameter) counter permitted satisfactory operation.

Calibration of the counter was usually made with the unfiltered fluorescence x-rays of silver or copper, excited in a beam of white x-radiation. In normal operation x-ray lines with 6% half-width at half maximum height were usual.

In many of the experiments the extremely low counting rates which were obtained required the counter to be operated continuously over a period of 12 hours, in order to obtain results of statistical significance. In general, the energy calibration of the counter remained steady to well within 5% over this length of time.

THE BACKGROUND OF THE COUNTER.

The background of the counter was just over 1500 c.p.m. when the counter was unshielded, and reduced to 380 c.p.m. when the instrument was operated in a 4" thick lead castle in a well shielded laboratory. When the counter was surrounded by an array of Geiger tubes operating in anti-coincidence with the main counter, the background was only 160 c.p.m. Further investigation showed that the bulk of the remaining background originated in the aluminium sheet lining the counter. This slight contamination appears to be a general property of aluminium and has recently been confirmed in independent work by Crathorn (8). Several substitute metals were tested and it was found that stainless steel gave the best performance, with backgrounds which were only 22 c.p.m. with lead and anti-coincidence shielding. Presumably some of the residual background must still come from the counter materials but this was not investigated further. Some, at least, of the background is known to arise from gamma radiation originating outside of the counter. Evidence for this is discussed in Chapter 4.

The final arrangement of the counters is shown in the photograph of Fig. 3. The proportional counter lies inside a ring of 22 Geiger counters of glass, with copper cathodes.

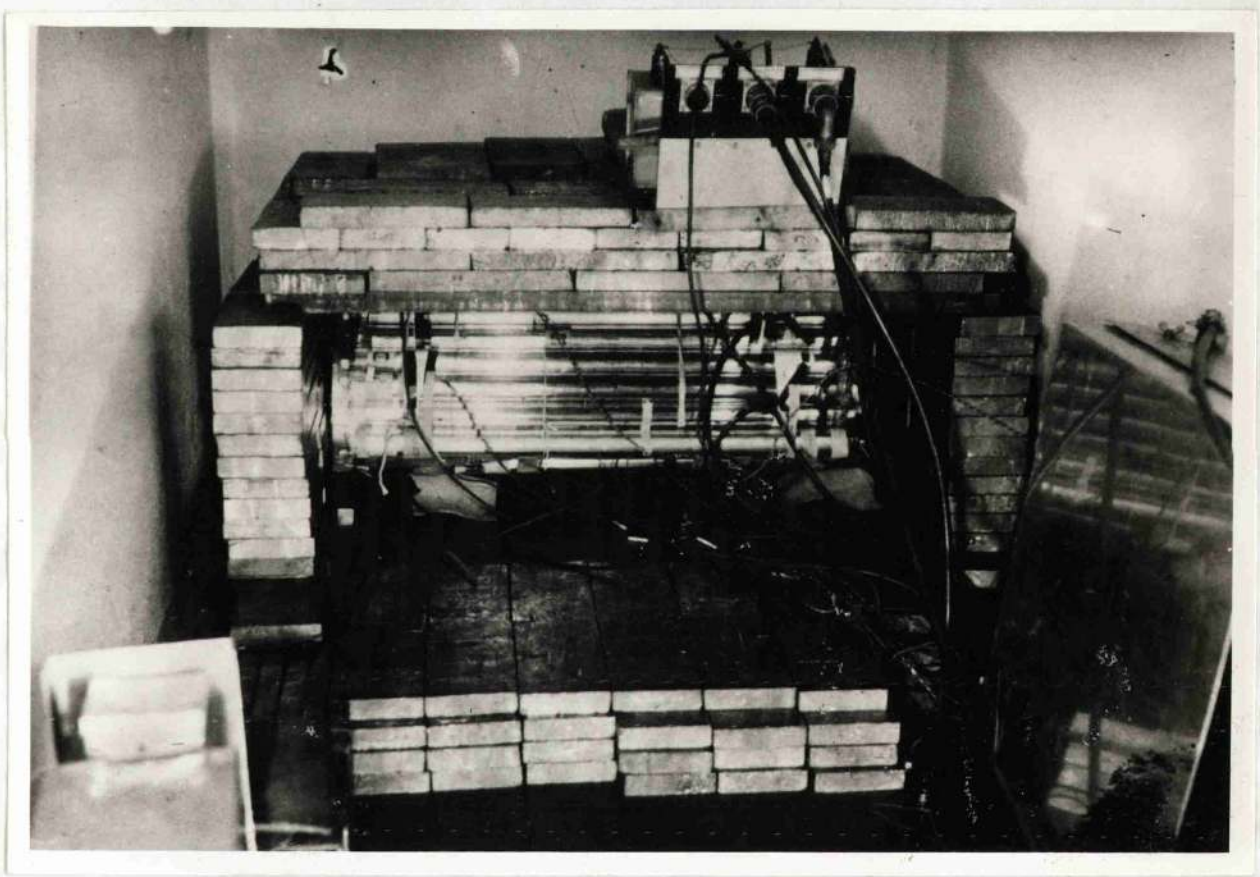

FIG.3. THE PROPORTIONAL COUNTER APPARATUS IN SITU.

A single layer of Geiger tubes completely surrounds the proportional counter. A double layer was shown to make no appreciable difference to the background. The counters considerably overlap the sensitive volume of the sample counter and appear to give very complete shielding from cosmic rays, so much so that the addition of Geiger counters at each end of the proportional counter was also found to have no appreciable effect on the background.

The final background consistently obtained with the apparatus was a total background (over all energies) of 22 c.p.m. This is comparable to that obtained by Kulp and Tryon (3) with a stainless steel Geiger counter, when the difference in available source area is taken into account. It is somewhat better than the final background of about 30. cp.m. attained by Crathorn for a 3 litre counter. (The counter used in this work has a sensitive volume of over 4 litres).

Kulp and Tryon find that the background can be further reduced by more than 50% by surrounding the counter with a 1" thickness of highly purified mercury. This was not attempted with the proportional counter used in this work because of the size of the counter. On the other hand, as pointed out by Curran et al. (4), the

effective background of a proportional counter may be very much less than its total background. This is because the action of a proportional counter in spreading out the background in a spectrum reduces the effective background if the source radiations cover only a limited portion of the total energy range. This point is brought out very clearly in work on osmium (Chapter 4) where L x-rays were examined. The part of the energy range of interest, 6-13 Kev (Fig.7) is a very small fraction of the total background energy range and the effective background is only 2.1 c.p.m., a factor of 10 better than an equivalent Geiger counter in which the counts are integrated over all energies. The same consideration applies in scintillation counters.

SUBSIDIARY APPARATUS.

The counters were operated from commercial high voltage stabilised power units, with extra smoothing added. The output from the proportional counter was amplified in a low noise linear amplifier, built from an unpublished design due to A.L. Cockcroft. The output from the amplifier was applied to the X-deflection plates of a cathode ray tube with no time-base, and the display recorded by a ciné-camera, in which the film moved

vertically past the screen. Pulse height analysis was subsequently carried out in a special micro-film reader. This photographic technique (9) makes it possible to record all the information at once, thus reducing the time required for an experiment. The introduction of the Hutchinson-Scarrott type of pulse analyser (10) made it possible to analyse a complete spectrum electronically and this was done in some of the later work on potassium.

The anti-coincidence arrangement also employed a photographic technique. The output of the Geiger counters, all connected in parallel, was shaped in a monostable multivibrator circuit and fed to the Y - deflection plates of the cathode ray tube. Even with no delay in the proportional counter circuit, when coincident events occurred in the two counter systems the Geiger array always registered first, deflecting the trace vertically on the screen to a position behind an opaque covering which hid the corresponding proportional counter pulse from the camera. When events occurred in the proportional counter alone, the spot was deflected across the centre of the screen, in full view of the camera. This proved to be a very simple and rugged anti-coincidence system and worked well in practice, and gave all the benefits of a photographic technique.

This concludes the brief description of the low background proportional counter apparatus developed for the examination of some of the natural radio-elements. It was the basic energy measuring device used in the experiments described in the succeeding chapters, although in most of the investigations scintillation counters were also employed. These, and any variations of proportional counter technique which were necessary in particular experiments, are described in the appropriate sections. Further investigation of the nature of the residual background of the proportional counter is discussed in Chapter 4, and possible improvements in the techniques are indicated.

3. A SEARCH FOR NATURAL RADIO-ACTIVITY IN NEODYMIUM.

Introduction.

Neodymium has been investigated a number of times (1-7) and most workers either failed to find beta activity or attributed it to common radio-active impurities. By far the most sensitive apparatus in the earlier work was used by Libby, who examined the element as an internally mounted source in a screen-wall geiger counter (5). A beta radiation with a maximum energy of about 11 Kev, and a half-life for the element of 5×10^{10} years was obtained. This appeared to be confirmed by work of Jha (8). Recently, Curran et al. (9) pointed out that the log ft value, assuming Libby's value for the half-life, would group with the other natural radio-elements if the maximum energy were 250 Kev. Preliminary measurements did not appear to rule out the possibility of radiations of this energy. At about the same time, Mulholland and Kohman (10) failed to find any activity, and put the maximum specific beta activity at 0.003 betas/sec/gm. of neodymium.

Neodymium occupies an interesting position in the periodic classification, next to the 'missing' element promethium ($Z = 61$), and the question of natural beta activity in one or other of the isotopes of neodymium

became of importance in connection with the occurrence in nature of possible stable isotopes of promethium (11). Nuclear stability rules, in general, limit the stable isotopes of odd Z elements to those of odd mass number. In particular, stability charts suggest ^{145}Pm and ^{147}Pm as possible stable isotopes. Of the corresponding neodymium isobars, ^{147}Nd does not exist in nature and is known to be radio-active with short half-life (12), and the recent discovery of the decay of ^{145}Pm to ^{145}Nd by electron capture (13) implies the stability of ^{145}Nd against decay to ^{145}Pm.

Consideration of beta stability limits led Kohman (14) to propose possible instability in ^{140}Nd or ^{150}Nd, both of which lie close to the limits of stability against beta decay, for even - A, even - Z nuclei. ^{140}Nd has subsequently been shown to have a 3.3 day half-life (15), but ^{150}Nd occurs in nature with 5.6% relative abundance in the natural mixture of isotopes of neodymium. ^{150}Pm lies well out-with the stability limits for even - A, odd - Z nuclei (16) but ^{150}Sm is apparently stable. If ^{150}Nd were radio-active, the-product would be ^{150}Sm, with ^{150}Pm existing naturally in transient equilibrium with ^{150}Nd.

The possibility of alpha-activity in some natural isotopes of even - Z elements has been suggested (17). Since ^{144}Nd contains two neutrons in excess of the closed shell

at 82, it may be expected to have tendencies in this direction. Several searches have been made (18-20) but only very recently has positive evidence for alpha-decay been reported (21), giving a specific emission of 0.015 alphas/sec./gm. neodymium.

EXPERIMENTAL PROCEDURE.

The apparatus described in the previous chapter was used. The output of the counter with, and without a source of neodymium was filmed and analysed to give the energy spectra shown in Fig.4. Background spectra are shown by the broken line histograms, background plus source spectra by the full lines. Two spectra, over ranges of high and low energy were taken. The same steel lining was used in all the experiments so that no errors could be introduced by contamination in the steel. A gas pressure of 40 lb./sq.in. was employed.

The source was of $NdCl_3$ prepared from 1.18 gm. of pure neodymium metal. In this, as in all the following experiments chemical reagents were tested in the counter for radio-active purity before the sources were prepared. The average source thickness was 2.1 m.gm./cm.^{2}

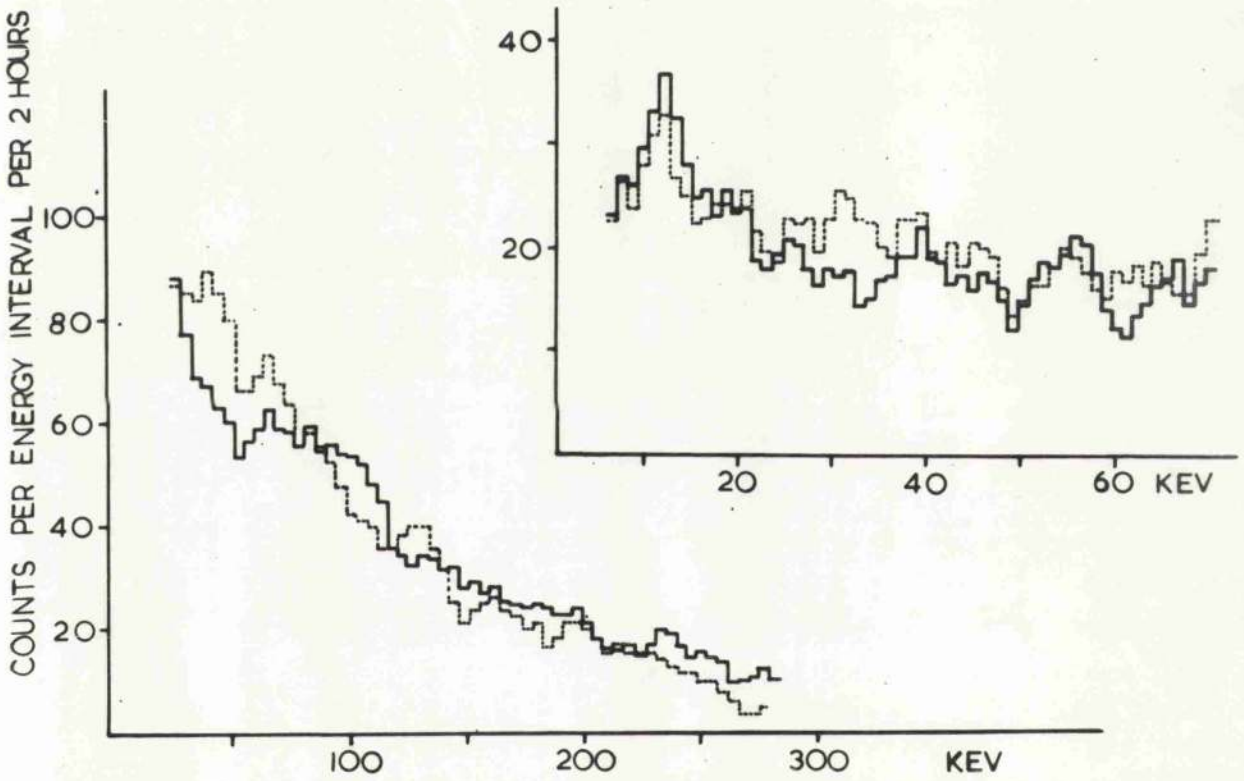

FIG. 4. SPECTRA OF BACKGROUND (BROKEN LINE) AND BACKGROUND + SAMPLE CONSISTING OF 1·18GM. NEODYMIUM (FULL LINE HISTOGRAM).

RESULTS AND DISCUSSION.

An examination of the spectra of Fig.4. shows that the background plus source spectrum has no trend away from the background spectrum at any energy above 6 Kev. This fact, and counting rate measurements at lower energies show that, if Nd^{150} is radio-active, its half-life cannot be less than 10^{15} years. There was, indeed, no evidence for beta activity, and this is in accord with the latest available information on mass measurements at the time of writing (late 1955). Since the above experimental work on neodymium was done, mass spectrographic measurements of the Nd^{150} - Sm^{150} mass difference by Hogg and Duckworth (22) have given a value of 4.6 ± 0.8 Mev for the energy difference between the ground states of the two nuclei. Recent measurements (23) of the energy release in the beta decay of Pm^{150} to Sm^{150} show that the ground states of Pm^{150} and Sm^{150} differ by 5.3 ± 0.15 Mev. On these figures the beta decay of Nd^{150} might be possible by about 250 Kev, taking the upper limit of one measurement, and the lower limit of the other. Very recently, however, Hogg as re-examined the question of the Nd^{150} -Sm^{150} mass difference (24) and concludes that his previous value was too high, and that the difference is about 4.0 Mev. With this value the

single beta decay of Nd^{150} is energetically impossible.

The presence of the source was observed to cause an increase in the number of high energy pulses, amounting to not more than 1.5 counts/minute. This was at first attirbuted to radio-active impurities, but following a private communication from T.P. Kohman concerning work on the alpha activity of Nd^{144}, an account of which has since been published (21), the source was re-examined. While the results confirmed the presence of high energy particles, the unfavourable nature of the source did not permit any more accurate measurements. The counting rate of 1.5 per minute obtained in the previous examination corresponds to a specific alpha -activity of about 0.02 alphas/sec.gm. neodymium, which is the same as that obtained by Mulholland and Kohman (10) using a proportional counter, and similar to the value of 0.015 suggested by experiments with nuclear emulsions (21). It is not known whether this alpha-activity is truly characteristic of neodymium.

CONCLUSIONS.

The experiment produced no evidence for beta-activity in Nd^{150} with a half-life of less than 10^{15} years. The absence of beta activity is in accord with recent mass measurements on the nuclei involved in a possible

transition. Some evidence was found for alpha activity in the source. If characteristic of Nd^{144}, the observed alpha emission rate corresponds to a half-life of not less than 2×10^{15} years for the isotope.

4. A SEARCH FOR NATURAL RADIO-ACTIVITY IN RHENIUM AND OSMIUM.

Introduction.

Rhenium and osmium each contain an isotope of nominal mass 187 in the natural mixture of isotopes. Since the elements are neighbouring species in the nuclear table ($Z = 75$ and 76 respectively) stability rules suggest that either ^{187}Re or ^{187}Os should be radio-active.

Some of the earlier studies (1-3) suggested that ^{187}Os was radio-active by K electron capture, but no such activity could be detected by Seliger and Bradt (4) or by Naldrett and Libby (5). Their failure to detect K capture in ^{187}Os led the latter experimenters to examine rhenium as a solid source on the wall of a Geiger counter, and evidence for beta decay in rhenium was claimed. The half-life of ^{187}Re was given as $(4 \pm 1) \times 10^{12}$ years, and by absorption in aluminium the maximum energy of emission of beta rays was found to be 43 Kev. These results were, in the main, confirmed by Sugarman and Richter (6) who examined purified rhenium in the same apparatus. Recent work by Gauthé and Blum (7) appeared to confirm that the end-point of the beta ray spectrum occurred at about 40 Kev.

On the basis of these results the log ft value is about 17.7, which suggests a third-forbidden transition. The spin of Re^{187} has been found (8) to be $\frac{5}{2}$, so that from the position of Re^{187} in the order of nuclear shell filling, the odd proton must inhabit a $d_{\frac{5}{2}}$ level (9). Thus the ground state of Re^{187} has even parity. The spin of Os^{187} was unknown when the work described in this chapter was done, but on shell theory ideas (9) it seemed possible that the last odd neutron in Os^{187} might occupy an $h_{\frac{9}{2}}$ or even an $i_{\frac{13}{2}}$ level. This would make the decay either third order *l*-forbidden, or fourth forbidden with $\Delta I=4$ (no).

Preliminary experiments here, using a proportional counter lined with aluminium, suggested that rhenium was indeed beta-active, but with a transition energy of 400 Kev. These results were subsequently proved to be erroneous, because the apparent beta-activity was shown to originate in the aluminium foil carrying the rhenium sample. The element has since been given a more thorough examination in the steel-lined counter.

EXPERIMENTAL PROCEDURE.

The same apparatus and techniques were employed as in the examination of neodymium. Sources were prepared from spectrographically pure rhenium metal. The metal, in the

form of a very fine dust, was shaken up with amyl acetate which contained a trace of nylon to cause the metal to adhere to the wall of the counter. Two sources, containing 0.617 gm. and 0.3805 gm. of rhenium respectively, were prepared and spread over 900 $cm.^2$ of counter wall, and examined. Background measurements were made using the same steel cathodes in order to eliminate the effects of any slight radio-active contamination within the steel. The energy spectra obtained with, and without, the source sample are shown in Fig.5. as full-, and broken-line histograms respectively. The two sets of spectra cover the energy range from 3.3 to 340 Kev.

RESULTS AND DISCUSSION.

An examination of the high energy spectra of Fig. 5 shows that there is no significant departure of the source + background spectrum from the background. The total number of pulses collected in this energy range confirms the absence of any radiations from the source. These are:-

Background, per 12 hours	=	13,030 ± 114 counts
Background + source, per 12 hours.	=	13,043 ± 114 counts
increase in counting rate per 12 hours	=	13 ± 161 counts.

Hence any increase in counting rate caused by the insertion

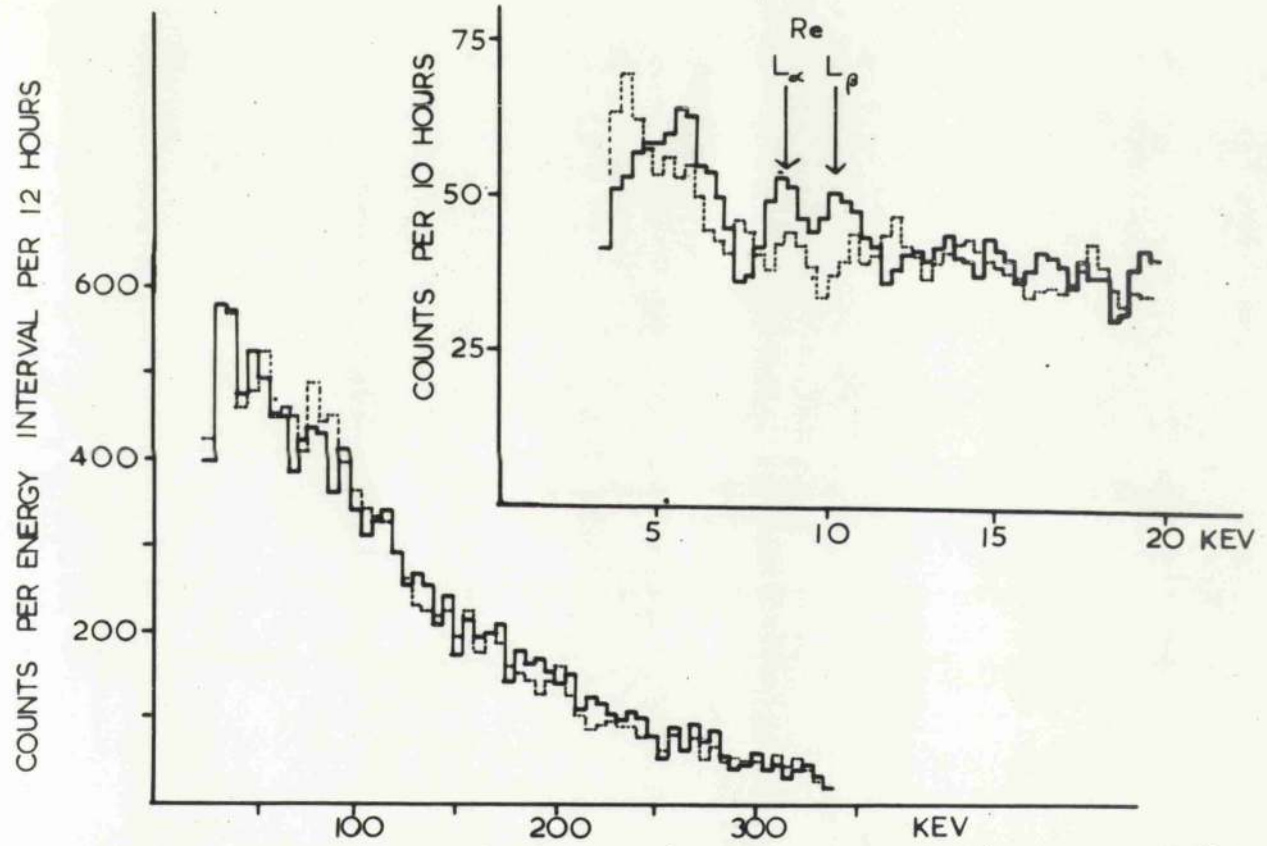

FIG. 5. SPECTRA OF BACKGROUND (DOTTED HISTOGRAM) AND BACKGROUND + SAMPLES OF RHENIUM (FULL LINE HISTOGRAM).

of the source into the counter is zero, within the limits imposed by the statistical nature of the events.

The low energy (3.3 - 20 Kev range) spectra show a slight divergence between 8 and 11 Kev, but otherwise the two curves follow each other closely. The integrated counting rates from 5 to 20 Kev give:-

Background, per 10 hours	=	1842 ± 43 counts
Background + source, per 10 hours	=	2112 ± 46 counts
increase in counting rate, per 10 hours	=	270 ± 63 counts

This increase was very small compared with approximately 8×10^4 counts, a large proportion of them falling in the 5-20 Kev range, which had been expected from published data on the radio-activity of rhenium.

The slight peaking between 8 and 11 Kev in the spectrum of Fig.5. corresponds well, in energy, with L x-rays of rhenium (or osmium). The emission of x-rays is not expected from Re^{187} which, if radio-active, can decay only by negatron emission. There is no evidence for the presence of a continuous beta-ray spectrum, and counting rate measurements were extended in energy down as far as 1 Kev without showing any beta-emission. The lack of experimental evidence for the beta decay of Re^{187} led to the search for instability in Os^{187} which is described in the

succeeding sections.

Later work showed that the slight L x-ray emission from rhenium and some, if not all, of the accompanying rise in counting rate could be attributed to background effects. In anticipation of these results, a lower limit of about 10^{16} years can be placed on the half-life of Re^{187} for beta-emission, unless the energy of the transition prove to be very low.

A SEARCH FOR RADIO-ACTIVITY IN OSMIUM.

A sample of osmium was first checked for positron or K x-ray emission in the shielded proportional counter system. A solid source was prepared from osmium tetroxide of more than 99% purity by reducing the tetroxide in amyl acetate, containing a little nylon as an adhesive. The first source contained 1.5 gm. osmium, and the spectra obtained with, and without the source are shown in Fig.6, on which the energies of the rhenium K x-rays are indicated. The results obtained with this source were not conclusive, so the spectrum obtained with a thicker source, containing 7.5 gm. osmium, is also displayed in Fig.6. It was obvious that K x-radiation, if present, was of low intensity, even allowing for the low efficiency of the

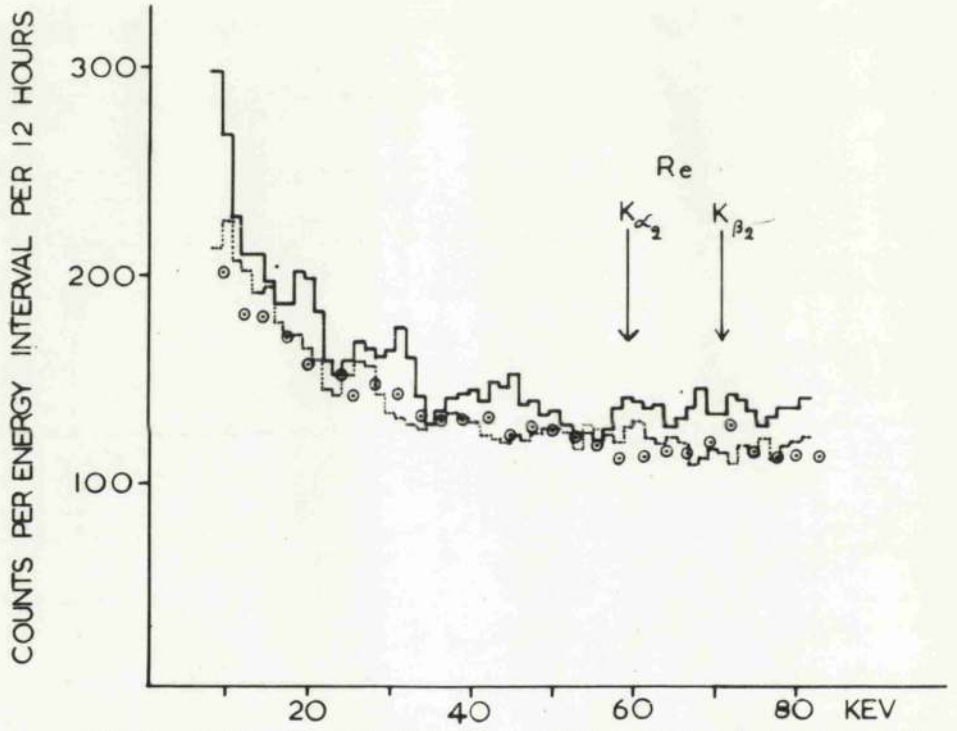

FIG. 6. SPECTRA OF COUNTER BACKGROUND (MARKED ⊙), BACKGROUND + 1·5 GM. OSMIUM (DOTTED HISTOGRAM) AND BACKGROUND + 7·5 GM. OSMIUM (FULL LINE HISTOGRAM).

counter (4%) for electromagnetic radiation of this energy. Nor was there evidence for a high energy positron spectrum, but an increase in the height of the spectrum at low energies was noted when osmium was present in the counter.

The low energy end of the spectrum was examined in more detail with increased gas gain in the counter and the results for two different samples of osmium are displayed in Fig. 7. The spectra show very definite L x-ray lines which were at first presumed to be L x-rays of rhenium following electron capture in Os^{187}, but the resolution of the counter was not sufficient to distinguish between L x-rays of rhenium and osmium. On the experimental evidence it seemed that Os^{187} might decay by L capture alone, perhaps because the difference in energy between the ground states of Re^{187} and Os^{187} is so small that K-capture is energetically impossible. That such an interpretation is not unreasonable is shown by nuclear stability charts (10) on which Os^{187} and Re^{187} both lie so close to the beta-stability limits that it is impossible to say in which direction the transition may go. However, it first remained to prove conclusively the absence of K x-ray emission from osmium. This required the high detection efficiency of a sodium iodide scintillation counter, and, as it happened, proved to be a vital experiment.

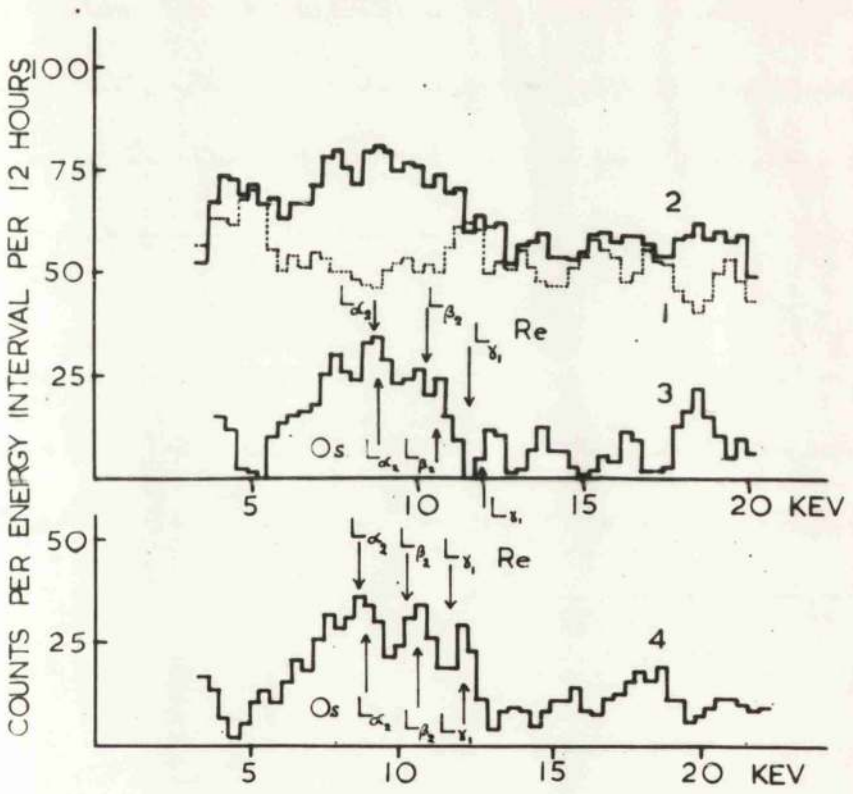

FIG. 7. SPECTRA OF OSMIUM AT LOW ENERGY.
HISTOGRAM (1) SHOWS BACKGROUND, (2) BACKGROUND + SOURCE OF 7·5 GM. OF OSMIUM, (3) THE DIFFERENCE BETWEEN 1 AND 2. HISTOGRAM (4) REPRESENTS THE DIFFERENCE SPECTRUM OF ANOTHER 7·5 GM. SAMPLE OF OSMIUM. RHENIUM AND OSMIUM L X-RAY ENERGIES ARE MARKED.

A SEARCH FOR K X-RAY EMISSION FROM OSMIUM.

The sodium iodide crystal was a cylinder, 0.7" in diameter and 0.7" high, packed in a thin perspex container which was lined with alumium foil to reflect the light on to the cathode of a photomultiplier. The output pulses were amplified, lengthened, and then displayed on a cathode ray tube. The trace was photographically recorded and analysed as before. Provision was made for packing samples of the source round the crystal container giving a geometrical efficiency approaching 50%. The apparatus was heavily shielded with lead. Energy calibrations were made with the 46.7 Kev gamma line of RaD.

Spectra obtained in this experiment are shown in Fig. 8. The background has a definite peak at about 75 Kev. When osmium (about 10 gm.) was packed tightly round the crystal the spectrum showed a distinct change. A peak appeared at an energy equal to the energy of osmium K x-rays, while the former peak in the background disappeared, and the background + source spectrum lay markedly below background in the 70-100 Kev region. Repeat experiments confirmed the differences between the background and source + background spectra. A control experiment using tungsten instead of osmium provided a spectrum which followed the osmium +

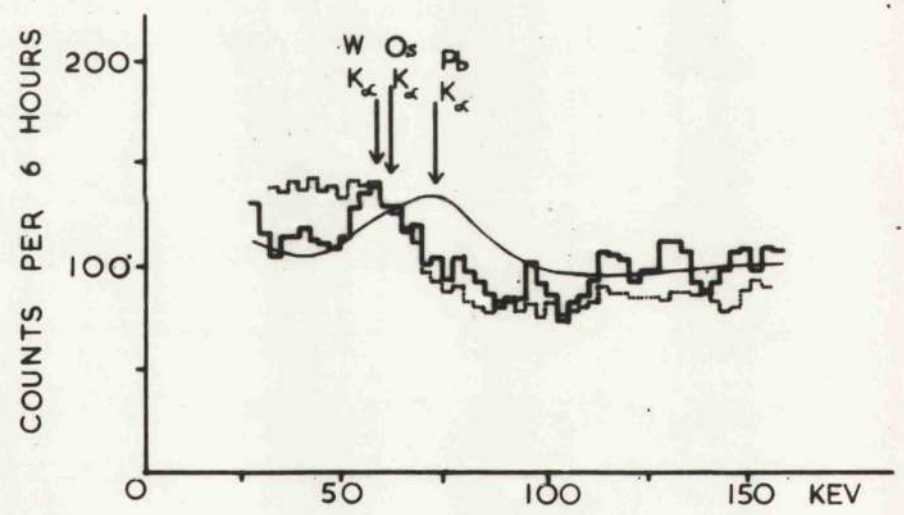

FIG. 8. SPECTRA OBTAINED FROM A SCINTILLATION COUNTER SURROUNDED BY OSMIUM (FULL LINE HISTOGRAM) AND BY TUNGSTEN (DOTTED HISTOGRAM). THE SMOOTH CURVE IS THE BACKGROUND SPECTRUM OF THE CRYSTAL UNDER A SHIELD OF LEAD.

background spectrum well.

As shown on Fig. 8, the position of the peaks in the osmium and tungsten spectra is the expected position for K x-rays of the two elements. The only other heavy metal near the crystal was the lead shield, and the position of the peak in the background spectrum suggests that it is caused by K x-rays from lead. Presumably the x-ray emission arises from the interaction of cosmic particles or gamma radiation with the lead in the shield. It seems probable that the change in the shape of the spectrum which was observed when either osmium or tungsten surrounded the crystal can best be explained by assuming that the metals filtered the lead x-radiation out of the background. The absorption of the background x-rays in osmium or tungsten would excite characteristic K x-radiation of the two metals, producing the peaks observed in their spectra.

The scintillation counter experiments, therefore, gave two important results. The first was that there is no K x-ray emission from osmium which can be attributed to K capture. This follows from the equality of the x-ray peaks of osmium and tungsten in Fig. 8. Although the mass of tungsten packed round the crystal was twice

the mass of osmium, the intensity of x-ray emission from the two samples was expected to be approximately equal because the number of lead x-rays in the background would be the same in both experiments. This is borne out by the experimental results, leaving no room for x-ray emission from other causes. The second point brought out by the experiment was that the background in a lead-shielded system contains a small amount of characteristic K x-rays of lead. The same presumably holds for any shielding element.

THE BACKGROUND OF THE PROPORTIONAL COUNTER.

The proportional counter system was also shielded with lead, although about half of the inner surface of the shield was hidden from the counter by an inch thickness of iron. Since the effective surface area of the proportional tube was about 1000 sq. cm., while that of the sodium iodide crystal was only 12.4 sq.cm., the lead x-radiation incident on the walls of the proportional counter would be about 80 times as intense as that on the crystal. Allowing for the steel covering half of the lead, this figure is reduced to 40. Much of the radiation would be absorbed in the walls of the counter, but about 2% could penetrate to the counting volume. This means that about the same intensity of lead x-rays enter the proportional

counter as entered the scintillation counter.

Under normal circumstances no trouble would arise because of the low efficiency of the counter at these energies. If, however, a thin layer of a heavy element covered the wall of the counter, some of the x-rays would be absorbed, causing the emission of low-energy photo-electrons and the characteristic x-radiation of the element. Once again the K x-rays would be detected but rarely, but L x-rays following the K emission would have a high chance of being detected. It seemed just possible that the L x-rays previously observed to be emitted from osmium might be caused by this mechanism rather than by L electron capture in Os^{187}. Further experiments were designed to check this possibility.

A SEARCH FOR EMISSION OF L X-RADIATION FROM HEAVY ELEMENTS.

Two metals, platinum and tungsten, were used for this investigation. Platinum foils, 54.6 m.gm./cm.^{2} thick, were placed round the inside of the counter and spectra were taken in the usual way. The tungsten sample was examined as a layer of metallic powder mixed with adhesive and approximately 10 m.gm./cm.^{2} thick. The spectra are shown in Fig. 9, with a background spectrum for comparison. L x-ray peaks are prominent in the spectra of the two elements. Since there is no reason for suspecting any natural electron-capture activity in these elements there can be little

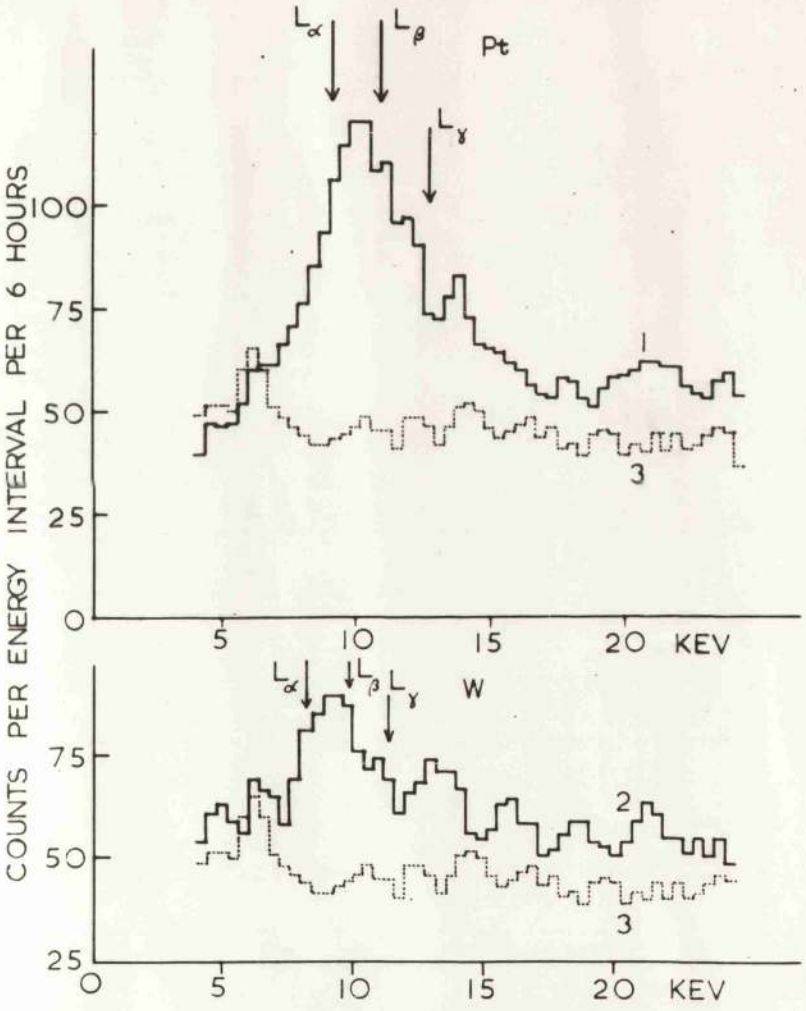

FIG. 9. SPECTRA OF RADIATIONS OBTAINED FROM SAMPLES OF PLATINUM (1) AND TUNGSTEN (2) COMPARED WITH BACKGROUND (3).

doubt that the x-ray emission is a background effect. The intensity of the radiation from the three samples:-

Osmium	(8.3 mgm./cm.^{2} thick)	38 counts/hour
tungsten	(10.2 mgm./cm.^{2} thick)	43 counts/hour
platinum	(54.6 mgm./cm.^{2} thick)	80 counts/hour

suggests that all of the x-ray emission from osmium can be accounted for this way. It is concluded that if Os^{187} is radio-active its half-life must be greater than 10^{15} years to explain the failure to detect any electron capture activity. If, as further evidence discussed in the next section suggests, Re^{187} is the unstable isotope of the pair then Os^{187} is completely stable against electron capture decay.

Interaction of the background x-rays with the sample can similarly explain the slight x-ray peak noted in the rhenium spectrum (Fig.5). The occurrence of L x-rays in such circumstances has not, so far as the author is aware, been demonstrated before, although it is worth pointing out that the L x-ray emission from In^{113} observed by Cohen (11), and attributed to L electron capture, might have originated in this way.

DISCUSSION. (i) THE PROBLEM OF INSTABILITY IN THE ^{187}Re - ^{187}Os GROUP

Since preliminary reports of the negative results of the present search for radio-activity in ^{187}Re appeared in publication (12) further work has been reported by others. Libby's group has revised (13) earlier estimates of the maximum energy release in the decay of ^{187}Re from 43 Kev to 8 Kev and the half-life from 4×10^{12} years to less than 10^{11} years, using the same apparatus as in the earlier studies (4,5). The report quoted a private communication from M.G. Mayer who suggested that, on shell model considerations, the beta decay of ^{187}Re should be only first forbidden with a spin change of 2. A very recent measurement (14) of the spin of ^{187}Os confirms that the spin is $\frac{1}{2}$. Assuming that ^{187}Re is beta-active and that its transition is first forbidden, the decay will obey G-T selection rules so that

$$\text{Log } (W_o^2 - 1) \text{ ft} = 10 \text{ approximately.}$$

where W_o is the total energy release in the transition. This equation can be solved graphically for different values of the half-life to give the maximum energy of decay. From isotopic abundance considerations ~~that~~ the half-life is unlikely to be less than 10^{10} years, which shows that the maximum possible energy available for the

transition will not exceed 5 Kev. In the experiments of the present study no beta rays were observed with an energy greater than 1 Kev. This would suggest that the half-life must be of the order of 10^{11} years or greater.

Undoubtedly the physical nature of the source examined in the proportional counter was not really suitable if the energy of the beta rays is very low, although Libby and co-workers used similar sources. Recent experiments by Drever and Moljk (15) show that the action of a proportional counter with low energy radiations from an internally mounted solid source leaves much to be desired, and it may be that a Geiger counter as used by Libby will show greater sensitivity under these conditions. Since it now appears that the energy release is very small any future investigation will probably require the element in a suitable gaseous form.

(ii) RHENIUM IN GEOPHYSICS.

As pointed out by Suttle and Libby (13), if Re^{187} decays with a half-life of less than 10^{11} years its decay product Os^{187} may be detectable in ancient rhenium-bearing minerals. The presence of radiogenic Os^{187} in samples of rhenium-bearing molybdenite has since been reported (16) and the half-life period for the beta decay of Re^{187} placed

in the range 5×10^{9} to 2.5×10^{11} years from measurements of the amount of Os^{187} in the samples (17).

There is little doubt that the discovery of radiogenic Os^{187} in mineral deposits is the most satisfying demonstration of the instability of Re^{187} which has yet been given. The absence of beta rays above 1 Kev suggests that the half-life is probably near the upper limit given by the geological work. An accurate estimate of the half-life by both geological and counter methods may yet prove important in connection with the possible occurrence of 'bound' beta decay in some natural radio-elements. This process, if at all important, may be expected to be especially so in the decay of Re^{187} because of the high atomic number of the nucleus and the low energy available for the beta transition. The problem of bound beta decay in relation to the natural radio-elements will be taken up again in Chapter 8.

(iii). THE ULTIMATE SENSITIVITY OF THE COUNTING APPARATUS.

The generation of L x-radiation by the background will impose one of the final limitations on the sensitivity of the counter arrangement. There appear to be a few ways in which it may be possible to mitigate the effects. If the cause of the trouble is mainly lead x-rays and not soft

gamma radiation, it will be possible to filter out the x-rays by lining the inside of the shield with ~~the~~ thick steel. X-rays generated in the steel have too low an energy to penetrate the walls of the proportional tube. Alternatively, it may be preferable to dispense entirely with lead and use only steel to shield the counters. This is advocated by Putman (18) who considers that heavy elements are undesirable since some of the secondary gamma radiation is produced by the decay of cosmic mesons, preferentially stopped in high Z materials. In the counter arrangement the shield is required only to cut out as much gamma radiation as possible, the anti-coincidence ring may be relied upon to remove ionising particles.

Apart from attempting to remove the agent causing the L x-radiation it may be possible, to some extent, to remove its effects by control experiments. These would involve measuring background spectra, not with a clean steel lining in the counter, but with a sample of a heavy element on the lining to simulate the source as closely as possible. It would appear best to use a neighbouring element, but a danger arises since the background contains a proportion of mono-energetic radiation. The swift variation in the photo-electric absorption near an absorption edge may introduce appreciable errors even between

neighbouring elements. The most sensitive method of all would be to use for the background determination a sample of the element in which activity is suspected. The source could then be a sample of equal mass and similar chemical composition, but enriched in the unstable isotope.

CONCLUSIONS.

No direct proof of the existence of instability in either ^{187}Re or ^{187}Os could be obtained in the investigation.

Sufficient experimental evidence was obtained to show that if, as the latest evidence suggests, ^{187}Re decays by negatron emission then the energy of the transition must be very low, less than 1 Kev instead of 40 Kev as proposed formerly. This suggests a half-life of 10^{11} years or more, for a first forbidden transition.

An x-radiation of low intensity, generated by cosmic ray background radiations, was shown to be emitted by heavy elements. This emission can mask a weak radio-activity unless special precations are taken to allow for its presence.

5. THE NATURAL RADIO-ACTIVITY OF LUTETIUM.

Introduction.

Natural lutetium contains 2.6% of an isotope Lu^{176} which is the central member of a triad of naturally occurring isobars Yb^{176}, Lu^{176} and Hf^{176}. The existence of a beta transition to Hf^{176} is well established (1-6) with a maximum energy of about 400 Kev and half-life greater than 10^{10} years. The log ft value of about 18.9 classifies the decay as at least third-forbidden. Flammersfeld (4) holds that about 67% of the total decay rate of Lu^{176} is contributed by an electron capture branch to Yb^{176}, but Scharff-Goldhaber (5) could find no evidence of K-capture.

By measurement, the spin of the ground state of Lu^{176} is 7 or greater (7) and, on theoretical reasoning (8) probably 10 ± 1. Such a high spin can be compatible with a third forbidden beta transition to an even-even nucleus only if the transition proceeds first to an excited state of the daughter. A gamma ray of energy about 400 Kev has been observed by Flammersfeld (4) but was attributed to the K-capture branch. A complex gamma spectrum detected by Scharff-Goldhaber (5) was found to contain gamma rays of 270 Kev, 180 Kev and 90 KeV in about equal intensities, the latter being strongly converted. This 90 Kev radiation is considered by Goldhaber and Hill (9) to be

identical to the 89 Kev gamma ray known to follow beta decay from the 3.75 hr. isomeric state of Lu^{176}. Identification of the 89 Kev gamma ray as arising in an E2 transition was made by McGowan (10) from a measurement of the K/gamma ratio. Goldhaber and Hill propose a decay scheme, shown in Fig. 11, in which the beta transition proceeds to an excited state of Hf^{176} which then decays to the ground state emitting three gamma rays in cascade, the last of which is the 89 Kev transition from a 2+ state to the 0+ ground state. Arnold and Sugihara (11) confirm the main points of the decay scheme by showing coincidence between the two high energy gamma rays and also between the gamma and beta rays.

Good agreement is observed between the gamma ray energies and the predictions of the Bohr-Mottelson rotational model of the nucleus (12) suggesting that the first three excited states of Hf^{176} have spins and parities, in order of increasing excitation energy, of 2+, 4+ and 6+.

Thus, although a considerable amount of work had been done on the decay of Lu^{176}, the decay scheme, especially as regarded a K-capture branch, had not been completely confirmed nor had beta and gamma ray spectra been published. It was with the main intention of providing the spectra and proving the decay scheme that a study of the element was included in this series.

Recently, Arnold (21) has re-examined the gamma ray

emission following the decay of Lu^{176} and has shown three gamma rays of energy 306, 203 and 89 Kev in cascade. No evidence for K electron capture was observed and an upper limit of 10% was put on the K-capture/beta decay branching ratio. This information was not available until after the completion of the present study.

THE PROPORTIONAL COUNTER EXPERIMENTS.

Two sources, prepared from lutetium oxide of the highest spectrographic purity, were used. One, of lutetium chloride, was 0.49 m.gm./$cm.^2$ thick and produced 480 c.p.m., the other consisted of lutetium oxide and was only 0.149 m.gm/$cm.^2$ thick. Both sources were deposited on steel, but kept well within the confines of the counting volume to prevent distortion of the spectrum which can be caused by escape of particles at the ends of the sensitive region of the counter. Full anti-coincidence shielding was used and the output photographed and analysed in the normal way. The spectrum observed from the thin source is shown in Fig.10. The results of subsidiary experiments have been fitted to the low energy end of the main histogram to give a complete display from 5 Kev to 500 Kev.

The figure shows a number of more or less distinct lines superimposed on a continuous spectrum, which has an

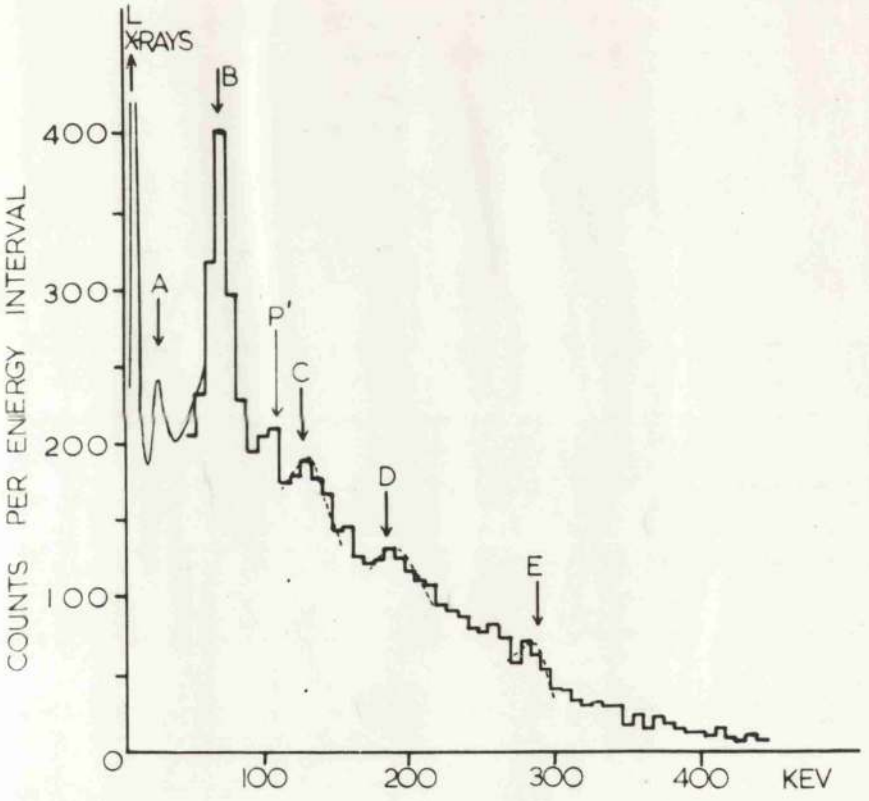

FIG.10. THE SPECTRUM OF Lu^{176} USING A 0·149 MG./CM.^{2} SOURCE. THE SYMBOLS ARE EXPLAINED IN THE TEXT.

end-point near 500 Kev. An L x-ray line is prominent between 5 and 10 Kev. Two peaks A and B (25 and 78 KeV respectively appear in the correct positions to be attributed to K and L internal conversion lines of the 89 Kev gamma ray which is known to occur. Three smaller peaks C (125 Kev), D (180 Kev) and E (290 Kev) are present. C and D can be due to K and L conversion electrons from a gamma ray near 190 Kev and E to L conversion electrons from a 300 Kev gamma ray. These gamma rays were observed alone in later experiments and their energies determined more exactly.

A Fermi plot of the beta spectrum is shown in Fig.11 The plot approximates closely to two straight lines which cross the energy axis at 425 Kev and 510 Kev. The difference between the crossing points is almost the same as the energy of the L internal conversion line of the 89 Kev gamma, and the complex nature of the Fermi plot can be attributed to the superposition of one beta spectrum extending from zero energy to a maximum at 425 Kev and a spectrum carried out along the energy scale by the coincident detection of L conversion electrons and beta rays. This is always so when a complex decay is examined in conditions of 2π solid angle counting geometry. The energy of the beta transition is therefore 425 ± 15 Kev, in good agreement

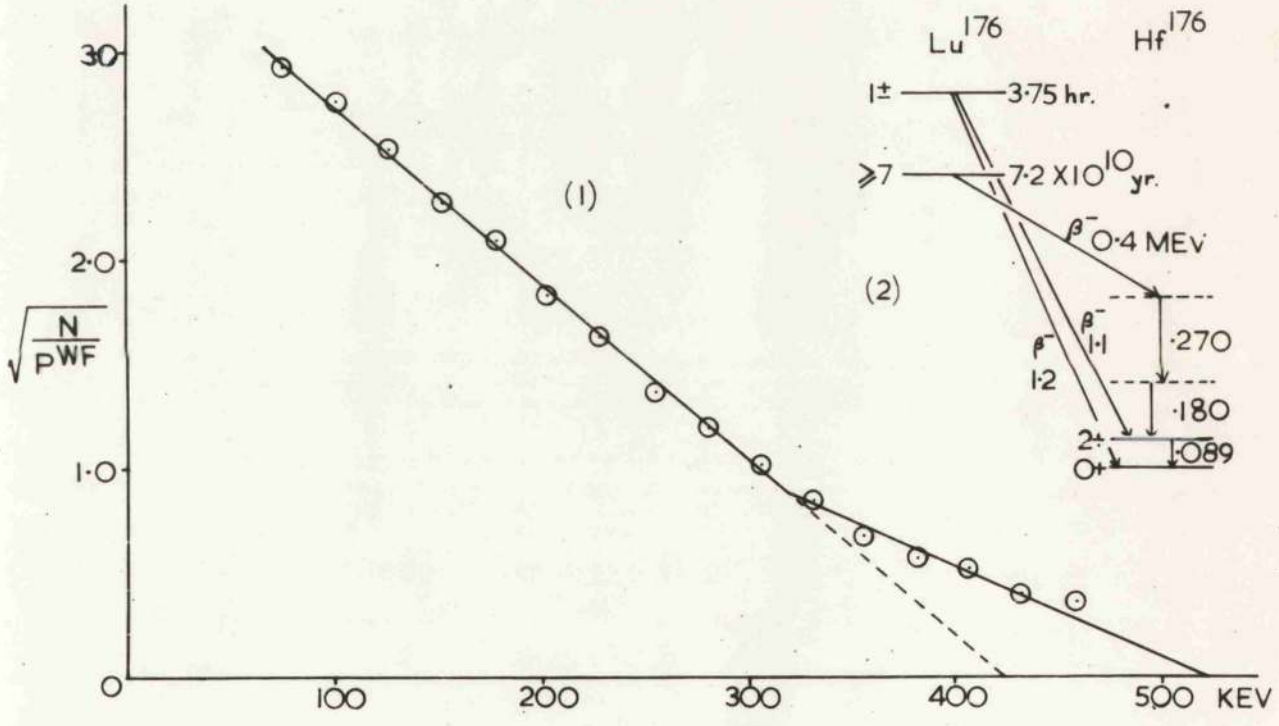

FIG. 11. (1). FERMI PLOT OF THE Lu^{176} BETA SPECTRUM.
(2). THE DECAY SCHEME OF Lu^{176} ACCORDING TO GOLDHABER AND HILL.

with previous results (3,4). The straight line character of the Fermi plot would suggest that the spectrum is of allowed shape but it is possible that the complex nature of the spectrum with the addition of electron lines on the true beta spectrum would mask any slight deviations from the allowed shape.

The counting rate observed from a source containing 0.2141 gm. of lutetium oxide was 480 ± 10 c.p.m. Allowing for absorption in the source, the counting geometry, backscattering of electrons from the steel lining of the counter and the mode of decay, this corresponds to a half-life of

$$(4.13 \pm 0.20) \times 10^{10} \text{ years.}$$

for the Lu^{176} isotope. The log ft value, 18.77 for a 425 Kev transition, appears to classify the decay as at least third-forbidden. These values are slightly different from those given in the published account of this work (13), this being due to the use of an increased figure for the backscattering of electrons from steel. The new backscattering factor, 0.45, is derived from the author's own work which is discussed in Chapter 7. This figure is considered more reliable because the measurements were made in a counting geometry chosen to simulate the experimental conditions as closely as possible. The

new value is sufficiently different to have appreciable effects on some derived values and on the conclusions drawn from the lutetium experiments, especially as regards a possible K-capture branch of the decay. However, supporting evidence for the revised branching ratio is good, but these points will be discussed later.

THE INTERNAL CONVERSION ELECTRONS.

The internal conversion lines from the 89 Kev gamma ray are shown in detail in Fig. 12. The calculated positions of the expected groups are marked. Because of the necessary source thickness, complete resolution of the lines was not obtained but the approximate relative intensities of the groups agree with the measurements made by Mihelich and Church (14) on the artificial isomer. Since Mihelich and Church showed that the empirical values of the K and L internal conversion coefficients classed the 89 Kev transition as E2, the 89 Kev gamma observed to follow the beta decay of the long-lived ground state of Lu^{176} may be taken as arising in a transition between the same two states in Hf^{176} in accordance with the decay scheme of Fig.11.

Calculations based on relative internal conversion coefficients of Mihelich and Church and on the K/gamma

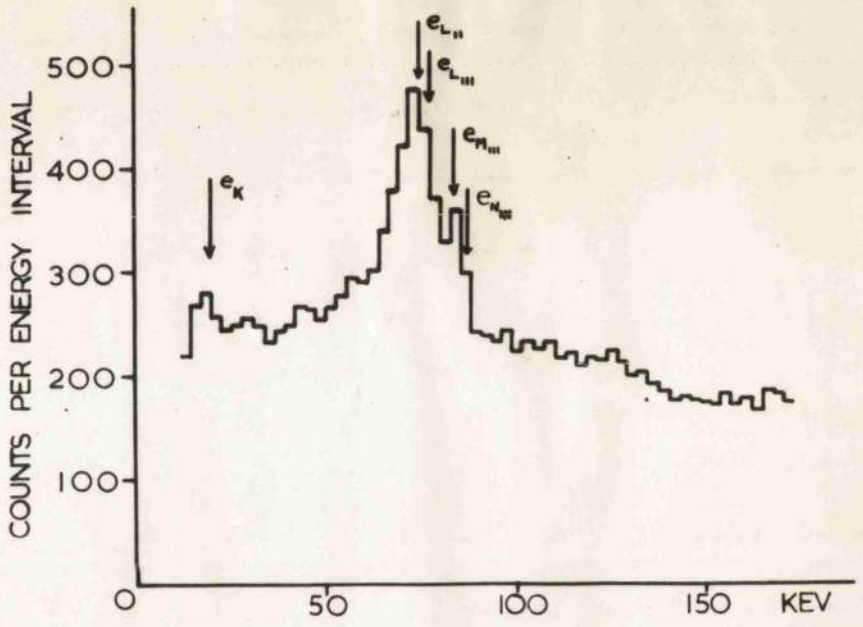

FIG.12. INTERNAL CONVERSION LINES FROM THE 89KEV GAMMA RAY TRANSITION.

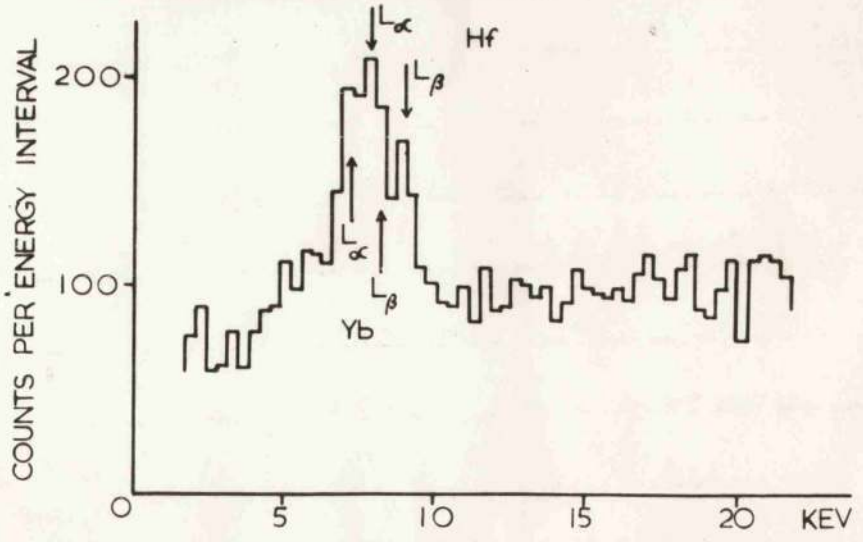

FIG.13. L X-RAYS OBSERVED IN THE DECAY OF NATURAL LUTETIUM-176.

ratio obtained by Mc.Gowan (10) for the artificial isomer show that gamma rays will be emitted in some 6% of the 89 Kev transitions. The proportional counter was used to search for this expected gamma radiation. The source was covered with a 500 m.gm./cm.^{2} sheet of steel to absorb all particle radiation and the spectrum is shown in Fig.14. The expected 89 Kev line is present and the 53 Kev K x-ray peak is evident. Allowing for changes in the amount of absorption in the steel and the differing efficiency of the counter for the x-rays and the gamma rays, the observed K/gamma ratio is 3.9, compared with Mc.Gowan's value of 1.3 for the artificial source. Allowing for contributions to the x-ray peak from internal conversion of the higher energy gamma rays, assuming E.Q. transitions, the ratio is expected to be 4.4. This agrees well with the observed value, considering the very low efficiency of the proportional counter for electromagnetic radiation at these energies.

In anticipation of experiments showing the presence of gamma rays of 190 Kev and 310 Kev energy it was suggested that the peaks C,D and E in Fig.10 were internal conversion electrons from these transitions. From the size of the peaks estimates, necessarily rough, were made of the amount of internal conversion undergone by the gamma rays. Comparison with the theoretical values of Rose et al. (15)

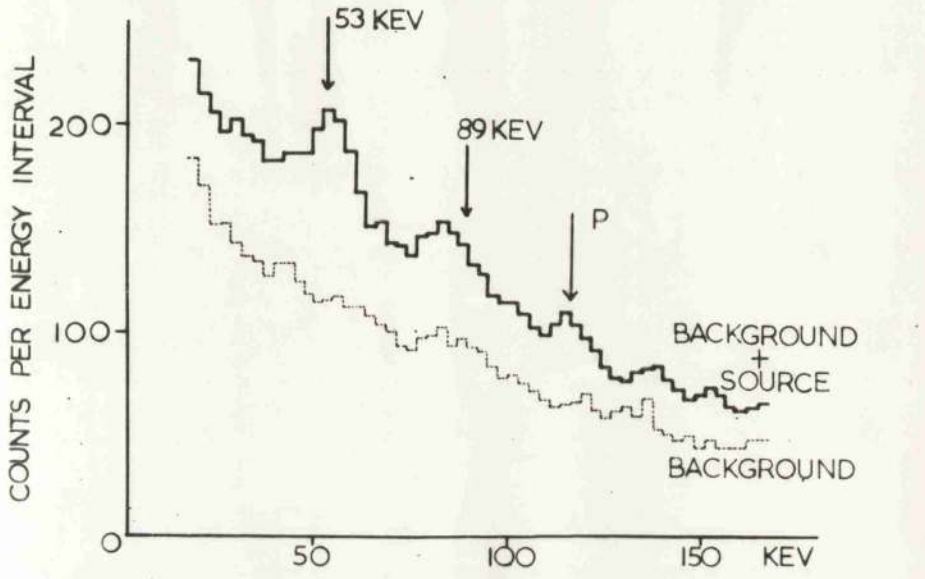

FIG.14. GAMMA RAY SPECTRUM OF LUTETIUM OBTAINED WITH PROPORTIONAL COUNTER.

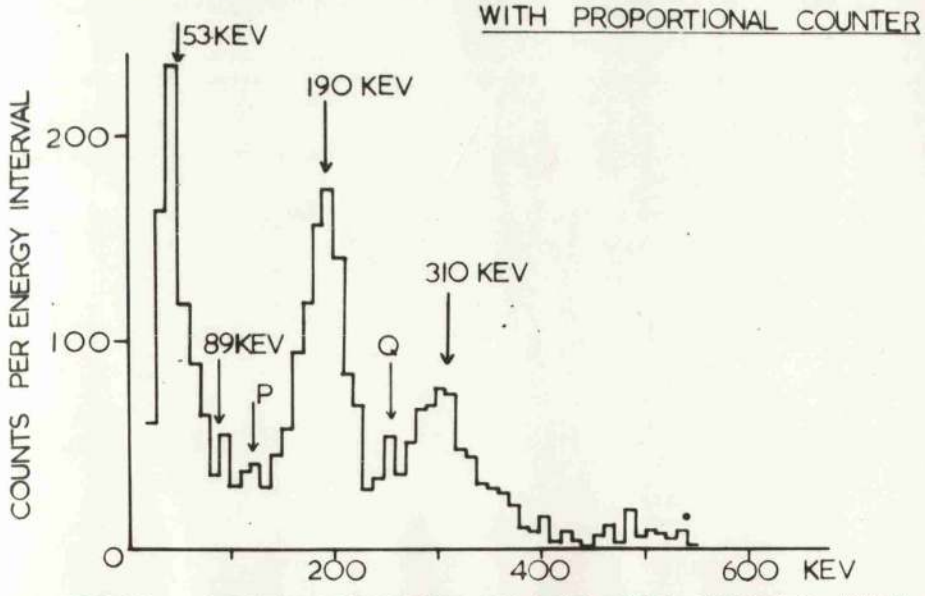

FIG.15. GAMMA SPECTRUM OF LUTETIUM OBTAINED WITH SCINTILLATION COUNTER.

showed that the evidence seemed to favour electric quadrupole transitions, which agrees with the above calculations on the K/gamma ratio.

THE POSSIBILITY OF ELECTRON CAPTURE IN ^{176}LU.

A K-capture branch of the decay may be expected to show up by emission of L x-rays following K-capture. As already shown (Fig.10), L x-rays were observed, and Fig.13 shows the L x-ray peak in more detail. Some of these x-rays must arise in K and L conversion of 89 Kev gamma rays and of the other gamma rays following the beta transition. On making allowance for the 2π geometry, backscattering, and the detection of beta rays and conversion electrons in coincidence with the x-rays, it can be shown that some 6% of the L x-rays arising in the beta decay branch can appear alone in the counter and contribute to the L x-ray peak. On the other hand the counter should 'see' 50% of the x-rays following K or L capture. This state of affairs, so favourable to the detection of electron capture, was used to put a limit on the amount of K-capture which could be present. It was found that only about 46% of the L x-rays actually observed in the proportional counter could be accounted for by the beta decay branch of the transition, assuming a fluorescence yield of 0.20 for Hf L x-rays (16). If the remaining 54% originate in an

electron capture branch the results suggest that electron capture transitions amount to 7% of those in the beta decay branch. This value had to be modified to take account of L x-rays arising in the internal conversion of gamma rays following electron capture, as shown later.

Unfortunately, the resolution of the counter was not sufficient to separate the x-ray groups. However, the expected positions of L x-ray lines of hafnium and ytterbium marked on Fig. 13 show that the shape of the spectrum is not inconsistent with the presence of x-ray groups of both elements.

THE GAMMA RAYS EMITTED IN THE DECAY OF ^{176}LU.

Further information about the gamma rays emitted in the decay of ^{176}Lu was sought by the use of sodium iodide scintillation counters. Thallium activated crystals, $\frac{3}{4}$" long and $\frac{3}{4}$" in diameter were used. The source, of lutetium oxide, was mounted in a well defined position at the end of the crystal, with sufficient absorber to shield the crystals from beta rays. The gamma lines were compared with the 279 Kev gamma line and the K x-ray line of ^{203}Hg for energy determination.

The gamma ray spectrum obtained from the source, examined by a single crystal, is shown in Fig.15. Lines

at 310 ± 10 Kev and 190 ± 10 Kev are distinct. A strong K x-ray line is also present at about 53 Kev. Calculations showed that many of the x-rays arose from the absorption of gamma rays in the source which was 0.45 gm./cm.^{2} thick. The almost complete absence of 89 Kev gamma rays confirms their high degree of internal conversion. The relative intensities of the 190 Kev and 310 Kev lines, taking into consideration the efficiency of the detector and source absorption, are almost equal. The rate of emission of each gamma ray was found to be approximately equal to the calculated beta decay rate of the source. Evidence derived from the gamma ray spectrum is therefore quantitatively in favour of the decay scheme of Fig. 11.

Qualitative evidence for the expected gamma ray cascade was obtained from gamma-gamma coincidence experiments. The source was mounted beteeen two identical sodium iodide crystals. The pulses from each photomultiplier tube after amplification were lengthened and fed, one set to the X-, the other set to the Y- deflection plates of a cathode ray tube. A brightening pulse was fed to the grid of the tube when coincidences were obtained between pulses in the separate channels. Thus the display was a set of spots on the face of the cathode ray tube, the abscissa of the spot being proportional to the energy of the gamma ray detected in

one crystal, and the ordinate proportional to the energy of the gamma ray observed, in coincidence, in the other crystal. The resolving time of the equipment was about 5 microseconds, but the counting rate was very low so that this rather large resolving time was of no consequence.

Sections of the coincidence display are shown in Figs.16. The figures show the number of gamma rays, as a function of energy, which were observed to be in coincidence with gamma rays lying in certain bands of energy, the bands being chosen to include one or other of the three gamma rays. Graph 1 shows gamma rays in coincidence with gamma rays lying between 300 and 320 Kev, graph 2 those in coincidence with 180-200 Kev gamma rays and group 3 with 80-100 Kev gamma rays. It is obvious from Fig. 16 that the 310 Kev and the 190 Kev rays are in coincidence, and that each are in coincidence with K-rays. Since many of the x-rays arise in internal and external K-conversion of the 89 Kev gamma rays in the thick source, this is tantamount to saying that the three gamma rays, 89 Kev, 190 Kev and 310 Kev are in cascade. The results are entirely in agreement with the decay scheme of Goldhaber and Hill.

DISCUSSION.

The excited states of Hf^{176} have been used in support of the Bohr-Mottelson rotational model of the nucleus (12). In

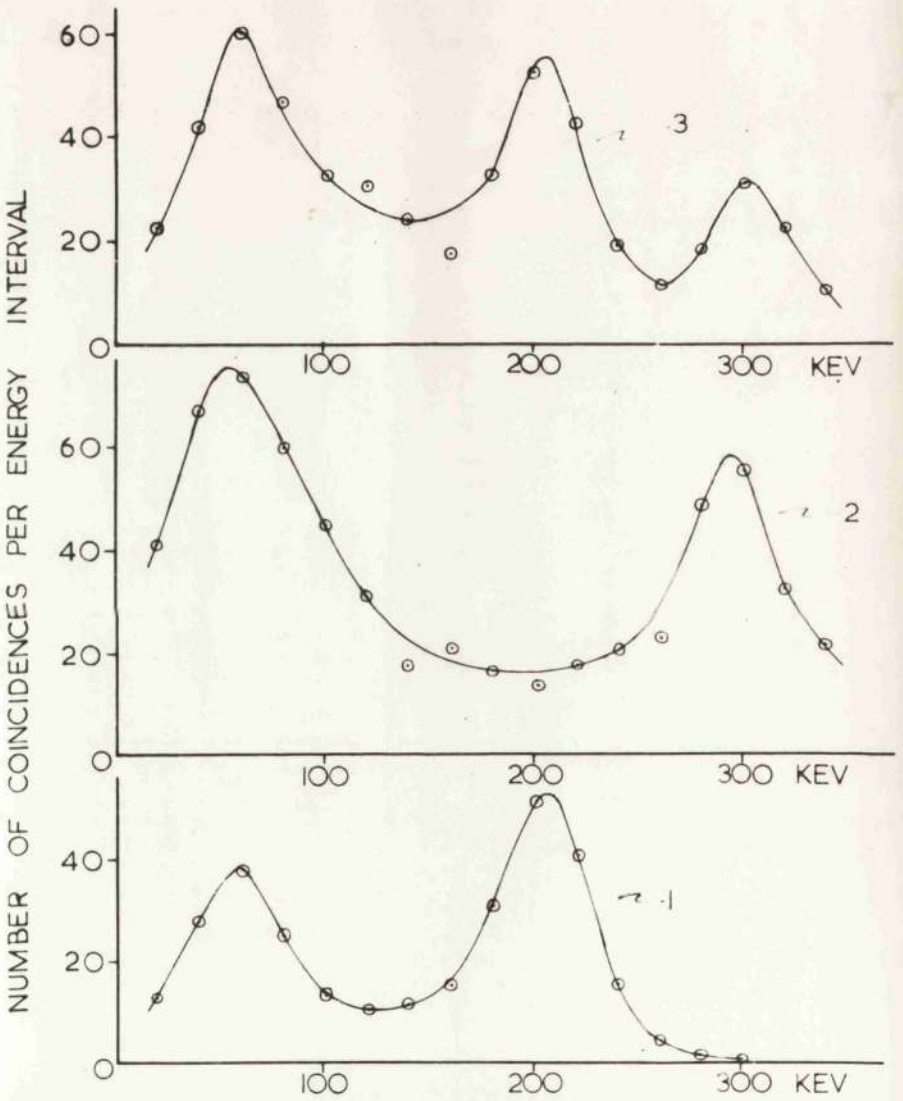

FIG.16. GAMMA-GAMMA COINCIDENCE SPECTRA OF LUTETIUM-176. THE SIGNIFICANCE OF EACH IS EXPLAINED IN THE TEXT.

this theory, low-lying rotational states of the nucleus are associated with the large deformations to be expected in nuclei where many particles lie outside closed shells. The energies of the low-lying states are expected to be given closely by the formula.

$$E = \frac{\hbar^2}{2\mathcal{J}} I(I+1), \quad I = 0,2,4,6, \text{ even parity}$$

The following table compares the observed energies of the states with theoretical values given by the above expression, normalised to give an energy of 89 Kev for the first excited state.

Excited state of Hf^{176}	E.2	E.4	E.6
Experimental energy	89 Kev	279±10 Kev	589±20 Kev
Theoretical value	(89 Kev)	297 Kev	623 Kev

The agreement is very satisfactory, and this evidence, in addition to the amount of internal K-conversion undergone by the three gamma rays, agrees with the assignment of spins and parities of 2+, 4+ and 6+ to the first three excited states of Hf^{176}. The assignment of 2+ and 4+ to the first two states is also in accord with empirical rules derived by Scharff-Goldhaber (17) for the first and second excited states of even-even nuclei.

The evidence for an electron capture branch of the decay was shown to depend on excess L x-rays. If electron capture occurs with detectable frequency it is certain that it will proceed via excited states of Yb^{176} because of the large spin change involved in a ground state to ground state transition. In this region of the nuclear table, the energy of the first excited state of even-even nuclei varies little from nucleus to nucleus (17), being about 100 Kev. With this knowledge it is interesting to examine the experimental spectra more closely for further evidence of the electron capture branch. This is forth-coming in the presence of two tiny peaks marked P in the gamma ray spectra of Figs. 14 and 15, and a peak P' in the spectrum of particle radiations shown in Fig. 10. The gamma ray peaks P lie close to 115 Kev in both Figs. 14 and 15 and P' corresponds to particles with an energy of about 105 Kev, being the correct position for L-conversion electrons from a 115 Kev gamma ray. Thus these three peaks, separately almost insignificant, taken together offer strong proof for the presence of a hitherto unsuspected gamma ray. The normal rules for the first excited states of an even-even nucleus (17) show that the spin and parity of the first excited state of Yb^{176} are almost certainly $2+$. The presence of appreciable L-conversio
n

makes the assignment of the 115 Kev gamma ray to a transition from a 2+ state to the 0+ ground state of ^{176}Yb entirely plausible.

In addition to the peak P in Fig.15 another small peak Q may be discerned at an energy of 250±10 Kev. It is possible that this may be evidence of another gamma ray in the electron capture branch, but the peak could also arise by the coincident detection of a 190 Kev gamma ray and a K x-ray in the crystal, although this process might be expected to give a broader peak. The origin of the peak is therefore open to some doubt, but it is worth noticing that the energy is approximately what might be expected from a 4+ to 2+ transition on the Bohr-Mottelson theory, assuming a 2+ to 0+ transition energy of 115 Kev.

Thus it appears that the experimental results are in favour of an electron capture branch in the decay of natural ^{176}Lu. There is strong evidence for the emission of a 115±10 Kev gamma ray following the electron capture, with the possibility, also, of the emission of a 250 Kev gamma. These proposals are incorporated in the decay scheme of Fig. 17 which was drawn up on the results of the present study.

If gamma rays follow electron capture, the L x-rays

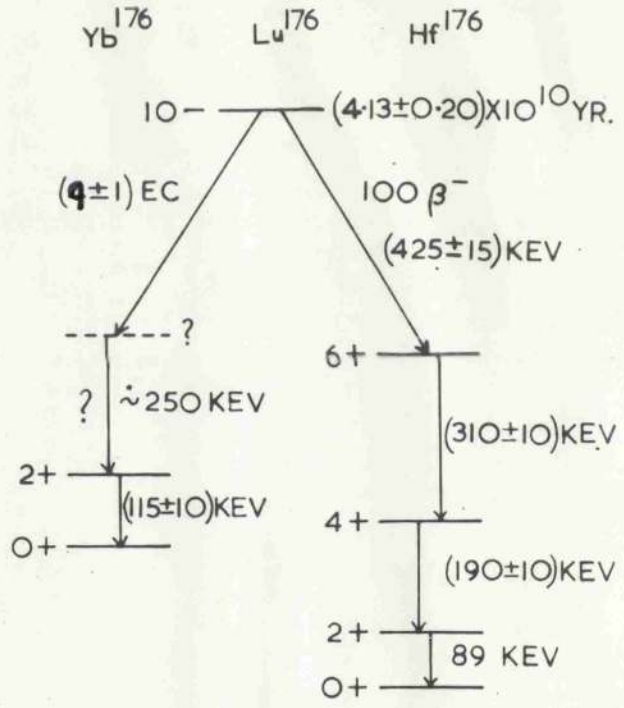

FIG. 17. PROPOSED DECAY SCHEME OF THE LONG-LIVED GROUND STATE OF LUTETIUM-176.

and electrons from internal conversion of these gamma rays will alter the proportion of the electron capture transitions which contribute to the Lx-ray peak. Calculations based on the K-conversion coefficients of Rose et al. and $\frac{K}{L}$ values of Gellman et al. (22) show that the intensity of electron capture must be changed from the original estimate of 7% of the beta transitions to 9%. Further evidence for a branching ratio of this magnitude is provided by the observed ratio of the intensities of the 115 Kev gamma ray peak and the 89 Kev gamma peak in the proportional counter spectrum of Fig.14. This agrees with the calculated value to within 5%. Good agreement is also obtained between theory and experiment for the ratio of the number of L internal conversion electrons from the 115 Kev gamma ray and the L,M and N conversion electrons from the 89 Kev gamma ray.

The foregoing interpretation of the electron capture branch of the decay is of recent origin and based on the author's re-investigation of the experimental data using a new value for the backscattering of electrons from steel. In the published account of this work the use of a different backscattering factor made the electron capture branch seem of low intensity and almost completely masked by the beta decay branch. Thus the extra gamma ray peaks were overlooked. Unfortunately, the author has now no

opportunity to check the predictions experimentally, but it would appear that an experiment based on a 4π counting geometry in a scintillation counter would separate the two branches of the decay very effectively. By integration in the crystal, the products of the beta decay branch would produce spectra with beta spectrum characteristics, in other words, the spectrum would be continuous over a band of energies. However, the products of the electron capture branch, x-and gamma rays, would produce discontinuous line spectra, and should be easily detectable above a predominant beta mode of decay.

The log ft value of 18.77 for the beta decay suggests a third or possibly a fourth forbidden transition. The minimum spin change is thus $\Delta I = 3$ and the spin of the ground state of Lu^{176} at least 9 units. The single particle arbitals which can couple to give a spin of this order are $h_{\frac{11}{2}}$ and $i_{\frac{13}{2}}$ (Reference 18, page 98). This implies that the parity of the ground state of Lu^{176} is odd. If this is so, then a fourth forbidden beta transition to the 6+ level of Hf^{176} is ruled out, and only a third forbidden transition can explain the observed log ft value. Hence $\Delta I = 3$ or 4 (yes). The shape of the Fermi plot seems to rule out the possibility $\Delta I = 3$ (yes) which was found to apply in the case of Rb^{87} (19), whose Fermi plot departed markedly from the

allowed shape and required the third forbidden correction (20). Thus it appears that the ground state of Lu^{176} may have a spin of 10 units and odd parity.

Arnold (21) has suggested that the half-life of Lu^{176} may be short enough to permit its use in dating experiments, especially as the daughter nucleus Hf^{176} has a low isotopic abundance. It seems unlikely that the electron capture branch of the decay will be useful in this way, in view of its low transition probability and the comparatively high relative isotopic abundance of Yb^{176}.

CONCLUSIONS.

Natural Lu^{176} has been shown to undergo a beta (negatron) transition with a half-life of $(4.13 \pm 0.20) \times 10^{10}$ years, followed by three gamma rays in cascade. The observed excitation energies of the first three excited states of Hf^{176} provide good agreement with predictions from the Bohr-Mottelson rotational model of the nucleus, and are accordingly assigned spins and parities of 2+, 4+ and 6+, confirming earlier proposals. This assignment permits beta decay, with significant probability, from the ground state of Lu^{176} ($I = 10-$) only to the 6+ state of Hf^{176} in accordance with experiment.

The presence of electron capture to an excited state

of Yb^{176} seems well established. Fairly conclusive evidence has been obtained for a gamma ray (energy, 115±10 Kev) in this branch of the decay. It seems possible that a second gamma ray (energy about 250 Kev) may also be emitted. The branching ratio of the decay (electron capture transition probability/beta decay probability) is (9±1)%. The proposed decay scheme for natural Lu^{176} is shown in Fig. 17.

6. THE BRANCHING RATIO OF POTASSIUM-40.

Introduction.

Although the emission of beta rays from potassium was first discovered (1) in 1905 and conclusively proved two years later (2), it was not until after 1943, when experimental evidence for K-electron capture was obtained (3), that intensive investigation of the decay was begun. The reason for much of the work was to place on a firm basis the use of the potassium-argon transition as a means of deriving the ages of potassium-bearing minerals.

The potassium-argon dating technique offers distinct advantages over methods making use of other radio-elements, and, in spite of initial difficulties, seems to have great potentialities. Apart from the obvious advantage afforded by the widespread terrestrial distribution of potassium-bearing minerals the main point of difference between the $^{40}_{K} - ^{40}_{A}$ transition and other natural transitions is that the end-product is a chemically inert gas. This not only permits tiny quantities of the decay product to be separated from the mass of accompanying material with a fair degree of ease and certainty, but also ensures that the argon is not likely to be appreciably contaminated with argon of other origin, since atmospheric argon could not easily

be trapped in the mineral at the time of its formation. In this respect the potassium-argon technique scores heavily over what would otherwise be a more suitable method, one involving the K^{40} - Ca^{40} transition which is faster. Calcium, however, is a common constituent of rocks and the high Ca^{40} content (97%) of ordinary calcium effectively masks the presence of small amounts of radiogenic Ca^{40} in all but the rarest cases. In spite of the advantages of an inert gas as a decay product the separation process requires considerable care and some of the earlier measurements (4,5) either failed to find argon in dated potassium minerals, or could show no relationship between the argon and the potassium content of the mineral and its age. Such a relationship was first demonstrated by Aldrich and Nier (6).

Any method of dating which involves the use of a radio-active isotope relies for its accuracy on a precise knowledge of the decay contents of the isotope. Much of the recent experimental work on the K^{40} decay has been aimed at obtaining a correct value for the rate of decay to A^{40}. Since the beta decay rate is more easily determined it has become usual to relate the rate of decay by electron capture to the beta transition rate by a branching ratio defined as $\frac{\lambda_{EC}}{\lambda_{\beta}}$ where the symbols represent the appropriate decay contents.

Comparison of the number of x-rays and Auger electrons emitted from the source with the beta decay rate gives the branching ratio directly. A number of investigators (3,7-11) have attempted direct measurements of the branching ratio, with results which vary from as high as 3 or 4 K-captures per beta decay to less than 0.07 per beta ray. The wide variation is mainly due to the very low energy (about 2.95 Kev) of the argon K x-ray which follows K-capture in K^{40}, a fact which makes quantitative work with solid sources extremely difficult. So far, no attempt has been made to examine K^{40} for K-capture in the gaseous state, owing to the difficulty of finding a suitable compound. However, recent work (12) on the operation of proportional counters at high temperatures may make such an investigation possible in the future. Of the direct determinations of the branching ratio the most notable experiment was that of Sawyer and Wiedenbeck (10) who differentiated between beta rays and Auger electrons in a specially designed screen-wall Geiger tube. The outcome of this experiment was a branching ratio of 0.135, subject to a possible error of up to 30%.

Happily, there is another way in which the branching ratio may be found by counter methods. This relies on the association of the high energy gamma ray emitted in the K^{40} decay with the electron capture branch, according to the

decay scheme on Fig.18. This scheme was first suggested in a slightly modified form by Suess (13) and supported on theoretical grounds by Morrison (14). Assuming that the decay scheme is valid, the branching ratio of the K^{40} transition may equally well be given by the ratio of the gamma and beta emission rates. A number of determinations of the gamma-beta ratio have been made, but onee again the results show considerable variation. Recent values (10,15-18) range from 0.05 to 0.127. Birch (19) considers that Sawyer and Wiedenbeck's figure of 0.127 is probably close to the correct value, and this is also selected by Burch (18) as best. It is close to the branching ratio determined by the same group from observations of Auger electrons (10). Thus, although the spread in empirical values of the branching ratio is considerable, counter experiments suggest that the ratio probably lies between 0.12 and 0.13.

In addition to the foregoing 'physical' methods of determining the branching ratio of K^{40}, two geological methods are available. The first is simply the reverse of the potassium-argon dating technique. The amount of K^{40} and of radiogenic A^{40} in a sample of a mineral of known age is determined. Then, assuming the rate of decay of K^{40} to Ca^{40}, the branching ratio may be found. The method is not completely independent of 'physical' determinations because it requires the beta decay constant of K^{40}. The

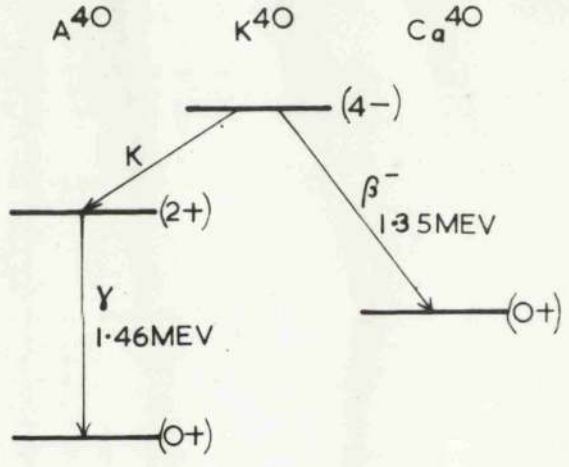

FIG.18. DECAY SCHEME OF K^{40} (AFTER SUESS AND OTHERS).

second more direct, but less universally applicable method obtains the branching ratio directly from a determination of the ratio of radiogenic A^{40} and Ca^{40} in the mineral. In this experiment no assumptions are required about the age of the sample or the rate of decay of K^{40} to Ca^{40}.

Aldrich and Nier (6), using the mass spectrometer to analyse the argon extracted from various mineral samples, found that the branching ratio appeared to lie between 0.02 and 0.09. Other values near 0.06 followed (20,21) and it began to look as if the physical and geological experiments gave irreconcilable results. This was the state of affairs at the time when the author's own work was begun. It seemed possible that the physical determinations of the branching ratio could be in error by an appreciable amount. In view of the sensitive apparatus which had been developed for examining beta emitters of low specific activity and steadily improving techniques in scintillation counting it seemed worth while re-determining the gamma-beta ratio of K^{40}.

Before proceeding to an account of original work it is of interest to consider more recent geophysical experiments. The early low branching ratios of 0.06 were shown to be the result of incomplete argon extraction

(22,23), new measurements giving about 0.09. A paper by Shillibeer and Russell (24) gives convincing evidence in the form of a series of age determinations of mineral samples with widely varying ages, for a branching ratio of 0.089. The calculations give excellent agreement with ages found by the lead-ratio and other methods. The same report discusses a nunber of investigations which serve to put the whole potassium-argon dating technique on a firm practical basis. It is shown that accidental loss of argon in the extraction process is unlikely. Similar conclusions are reached by Carr and Kulp (25) from experiments involving the use of tracer A^{37}. There is also claimed to be little or no diffusion of argon from lepidolites or feldspars during geological time. Gentner et al. (26), however, find evidence for diffusion loss from sylvites. Thus the latest measurements of A^{40} and K^{40} in potassium minerals suggest a branching ratio of 0.09 which is still smaller than that given by physical methods.

Only one determination of the branching ratio by direct comparison of radiogenic A^{40} and Ca^{40} in a sample of sylvite has been reported (27), giving a branching ratio of 0.126 ± 0.003, in close agreement with physical measurements. However, the possibility of argon diffusion

from sylvites (26) raises some doubts about the validity of the result.

DETERMINATION OF THE GAMMA-BETA RATIO OF K^{40}. (First Method).

The intensity of the gamma ray emission from a source of potassium was measured in the apparatus sketched in Fig.19. The gamma radiation was detected in a thallium activated sodium iodide crystal, cylindrical in shape, 2" long and ¾" in diameter. The crystal was examined by an E.M.I. (Type 6262) Photomultiplier whose output was amplified and fed through a pulse height discriminator to an electronic scaling unit, where the pulses were counted. The results were obtained in the form of integral bias curves by varying the setting of the discriminator. The bias curves were usually obtained by counting from voltage levels corresponding to the liberation of about 100 Kev of energy in the crystal and downwards until tube noise became noticeable (between 20 and 10 Kev). The total counting rate could then be obtained by extrapolating the curves to zero energy. This procedure was considered to be more accurate than counting at a fixed bias level because gamma rays of different energies had to be counted, for reasons that will be explained presently. Energy calibration for the bias curves was made using the 46.7 Kev line of RaD. Since the experiment was basically

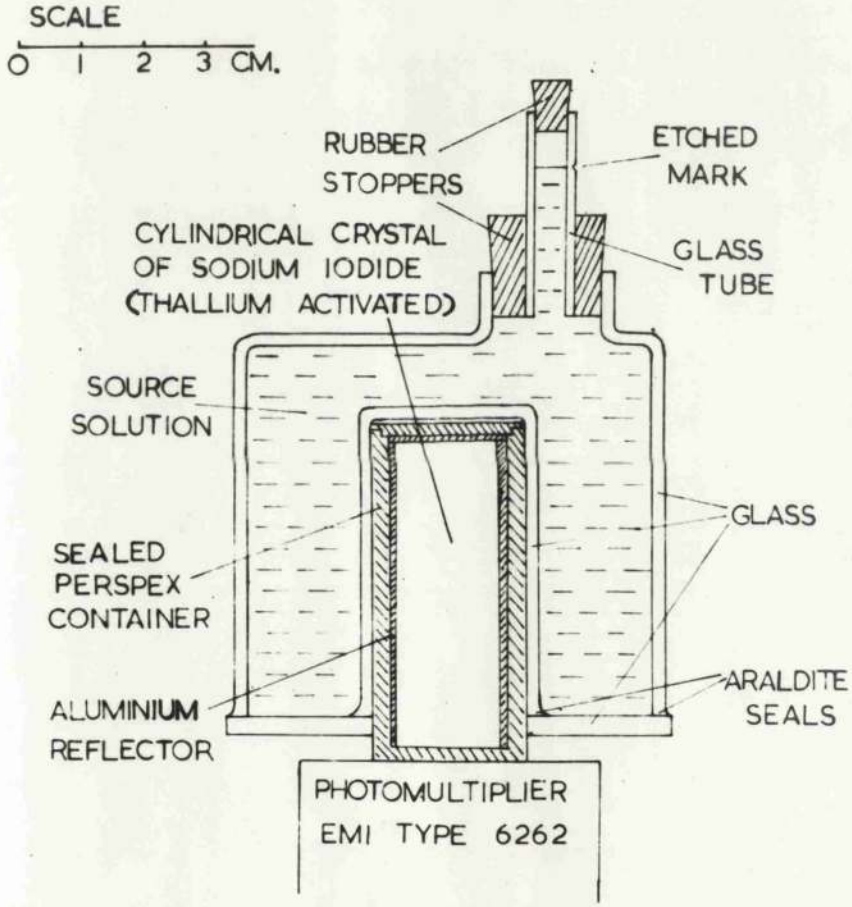

FIG. 19. DIAGRAM OF THE APPARATUS USED FOR MEASURING GAMMA RAY INTENSITIES IN THE FIRST POTASSIUM-40 BRANCHING RATIO EXPERIMENT.

the comparison of the rate of gamma emission from two sources in identical geometrical conditions, the best way of ensuring similar conditions was considered to be the use of solutions of the radio-active materials. A cylindrical glass container for the solutions was set over the crystal as in the sketch, so ensuring that the sources always had the same configuration relative to the detector. Since the sources completely surrounded the crystal this arrangement should also give the most favourable source to background counting rate ratio. This was further improved by use of a 4" shield of lead to reduce background. Sufficient absorber was built into the apparatus to ensure that the most energetic beta rays emitted by any material used in the experiments could not be detected in the sodium iodide crystal.

To obtain the maximum stability of operation of the apparatus arrangements were made for emptying, rinsing and filling the source holder without in any way disturbing the position of the crystal on the face of the photomultiplier or exposing the tube to light. Between experiments the container was thoroughly rinsed with acid, water, distilled water and acetone in that order. Air was then blown through the bottle to remove the last traces of moisture and 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. Even when solutions of ^{60}Co were used it was found to be an easy matter to remove every trace of contamination.

The potassium source solution was chosen to give the maximum amount of potassium in as small a volume as possible. The most suitable appeared to be a near-saturated aqueous solution of potassium hydroxide. The solution, in distilled water, contained 493 gm. potassium per litre. When the source holder was filled to a fixed mark with 186c.c. of this solution a counting rate of some 700 c.p.m. was obtained above a background of 87 c.p.m. under lead shielding. This difference was sufficient to ensure accurate determination of the counting rate due to the source.

Since the source solution had a high density a series of tests were made to see if the background of the apparatus depended on the contents of the source holder. Measurements of the backgroumd were made with the bottle empty, and also when it contained such non-radioactive liquids as distilled water, salt solution, calcium chloride solution and carbon tetrachloride (chosen for its chlorine content to simulate the presence of potassium in the source). It was found that the background was independent of the liquid contained in the holder, but that the background taken when the bottle was empty was consistently a few counts per minute low.

Hence all background measurements were made when the bottle contained distilled water.

To avoid the uncertainty of relying on a calculation of the efficiency of the gamma ray detector the experiment was based on the comparison of the gamma-beta ratio of ^{40}K with the gamma-beta ratio of a suitable control material in the same experimental conditions. The most suitable source to use as a control appeared to be ^{60}Co whose gamma transitions (1.17 and 1.33 Mev) are fairly close, in energy, to the 1.46 Mev gamma ray liberated in the ^{40}K decay, and whose gamma-beta ratio is known to be 2. Had the gamma rays from ^{60}Co and ^{40}K been of equal energy it is easy to show that the gamma-beta ratio of ^{40}K, R say, is given by:-

$$R = 2\frac{\gamma_K}{\gamma_{Co}} \cdot \frac{\beta_{Co}}{\beta_K} \quad \ldots\ldots\ldots\ldots\ldots\ldots (i).$$

Where γ_K and β_K are the observed gamma and beta counting rates of a source of potassium and γ_{Co} and β_{Co} the corresponding rates for a source of ^{60}Co, determined under similar conditions. Various corrections which are discussed later, had to be applied to the measurements because neither the beta nor gamma ray spectra of the two sources are identical.

The rates of beta emission from the two sources were

determined in a well-shielded proportional counter arrangement similar to that described in an earlier chapter. As with the gamma ray measurements, the intensity of beta emission was derived from bias curves which were extended down to an energy of about 200 ev. The curves were very flat at low energies and no difficulty was experienced in determining the relative beta emission from the two sources. Hence, ideally the experiment consisted simply of finding the intensity of gamma emission from equal volumes of solutions of potassium and Co^{60} in the apparatus which has just been described. Thereafter, accurately known volumes of the two solutions were separately spread over the inner wall of the porportional counter to give the relative beta emission rates. Then the branching ratio could be found from equation (i).

POSSIBLE SOURCES OF ERROR IN THE EXPERIMENT.

A number of possible sources of inaccuracy were considered likely to introduce appreciable errors into the experiment. These are now considered and corrections derived where necessary.

(i) In order to obtain large source areas in the beta ray experiments the sources were mounted on the inner surface of the wall of the proportional counter. Thus the sources were

deposited on what was, in effect, an infinitely thick backing material and the counting rate would be considerably influenced by electrons scattered from the wall into the sensitive volume. If the amount of backscattering were different for the two sources then observed counting rates would not be in the same ratio as the true rates of beta emission. The absolute magnitude of the backscattering was not important, only the variation of the backscattering factor with energy, but the literature on the subject was often contradictory. To clear up the point some subsidiary experiments were made and these are described in the next chapter (Ch.7). For present purposes it is enough to anticipate the findings and say that no appreciable dependence of the intensity of backscattering on the energy of the electrons was found. Hence the relative rates of beta emission from the two sources are obtained directly by measurement.

(ii) Full anti-coincidence shielding was used when obtaining the rate of beta emission from potassium, but the detection of gamma rays from ^{60}Co in the anti-coincidence array caused the loss of some 4% of the accompanying beta rays. Hence the rate of beta emission from ^{60}Co was determined without use of the Geiger array.

(iii) A correction had to be made to the potassium gamma ray count to allow for the detection in the crystal of bremsstrahlung excited by the beta rays of K^{40} in their passage through the dense hydroxide solution. The size of the correction was determined experimentally by observing the increase in the gamma rate when P^{32}, a pure beta emitter whose spectrum covers much the same energy range as the beta spectrum of K^{40}, was added to the potassium hydroxide solution. The activity of the P^{32} was monitored in the proportional counter to give a direct estimate of the correction required. The correction is shown graphically, applied to the K^{40} gamma ray bias curve in Fig. 20.

No such correction was made to the Co^{60} gamma rate because,

(a) the intensity of Co^{60} beta rays relative to the gamma rays is a factor of about 16 down on the relative beta-gamma intensity from K^{40} ~~and~~ Co^{60}

(b) the energy of the Co^{60} beta transition is considerably less than that of K^{40} and

(c) the Co^{60} solution, mainly water, did not contain the high concentration of relatively heavy atoms that existed in the potassium solution.

(iv) a slight correction was calculated to allow for the coincident arrival in the crystal of the two gamma rays from a Co^{60} decay.

(v) Empirical tests were made to determine what effect the source solutions had on the transmission of the gamma rays to the crystal. These experiments took the form of adding equal amounts of a Co^{60} solution to water and to strong potassium hydroxide solution in the gamma ray counting apparatus. Results consistently showed that the higher counting rate was obtained from the Co^{60} when it was added to the hydroxide, suggesting that increased scattering in the denser solution was more important than increased absorption, in the geometry of the experiment. Accurate measurements showed that a correction of about 6% had to be added to the gamma counting rate of Co^{60} to allow for this.

(vi) Finally, a correction was required to take account of the variation with energy of the efficiency of the sodium iodide detector. This could be obtained directly, by calculation, to give

$$\frac{I_{Co}}{I_K} = 1.015 \frac{\mu_K}{\mu_{Co}} \cdot \frac{\gamma_{Co}}{\gamma_K} \quad \ldots\ldots\ldots\ldots\ldots \text{(ii)}$$

where I_{Co}, I_K are the true numbers of gamma rays of Co^{60} and K^{40} respectively, which enter the crystal, μ_{Co} and μ_K the mass absorption coefficients of sodium iodide for the gamma rays of Co^{60} and K^{40}, and γ_{Co}, γ_K the observed gamma ray intensities of the two sources. The numerical factor arises

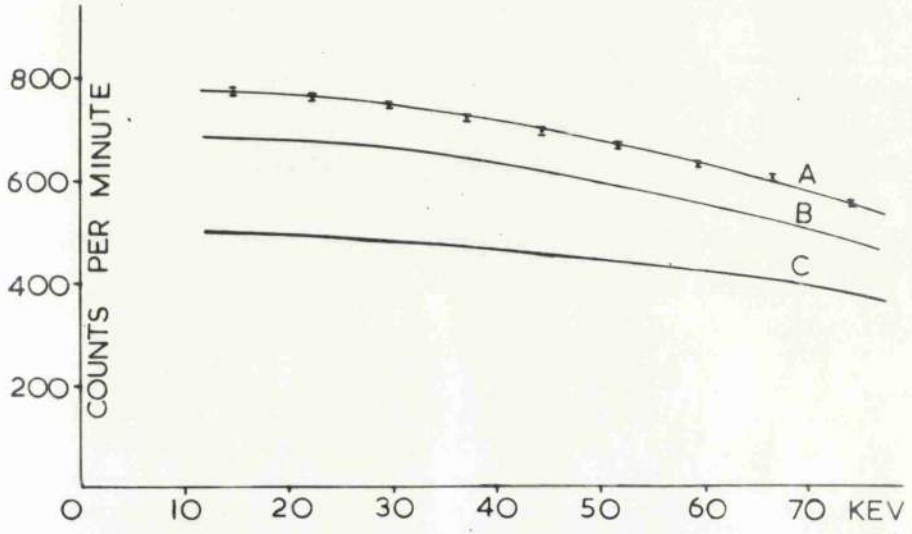

FIG. 20. POTASSIUM-40 GAMMA RAY BIAS CURVES
(A). EXPERIMENTAL (B). WITH BACKGROUND REMOVED
(C). CORRECTED FOR BREMSSTRAHLUNG.

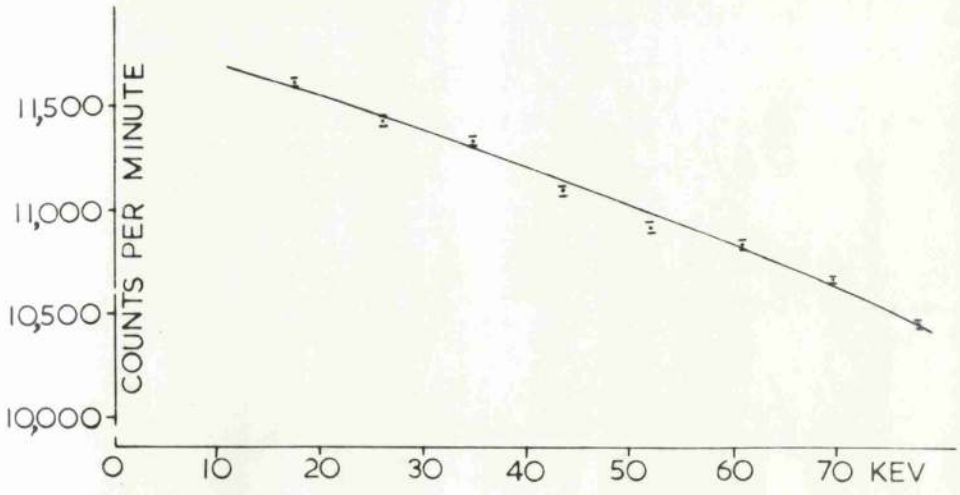

FIG. 21. CORRECTED BIAS CURVE FOR COBALT-60 GAMMA RAYS.

because the crystal cannot be considered mathematically as a thin layer of sodium iodide. The factor depends on the size of the crystal and can be estimated graphically. Hence, from equations (i) and (ii)

$$R = \frac{2}{1.015} \cdot \frac{\gamma_K}{\gamma_{Co}} \cdot \frac{\mu_{Co}}{\mu_K} \cdot \frac{\beta_{Co}}{\beta_K} \quad \text{.....(iii)}$$

and the gamma-beta ratio may be found. In this equation it is assumed that the counting rates γ_K and γ_{Co} have been corrected for all the above effects except crystal efficiency.

RESULTS.

The results of the gamma counting experiments are shown by the bias curves of Figs. 20 and 21. Fig. 20 gives the experimental bias curve for K^{40} (curve A) and with background subtracted (b) and corrected for the effect of bremsstrahlung (C). The corrected curve for Co^{60} is shown in Fig.21. The final result of the experiment was a gamma-beta ratio of

$$0.124 \pm 0.002$$

where the error is derived from the statistics of counting.

DETERMINATION OF THE GAMMA-BETA RATIO OF K^{40}. (Second Method).

A second method of finding the gamma-beta ratio of K^{40}

was devised to overcome many of the unsatisfactory features and reduce the sources of error inherent in the first. Since most of the corrections were basically due to the differemce in energy between the K^{40} transitions and the corresponding ones in Co^{60}, a new calibrator was chosen for the second method. An ideal isotope to use appeared to be Na^{24} which emits a beta ray with a maximum energy of 1.39 Mev (K^{40}, 1.36 Mev) followed by two gamma rays in cascade with energies of 2.76 Mev and 1.38 Mev, (K^{40}, 1.46 Mev), if the two gamma rays could be examined separately. The presence of the 2.76 Mev gamma ray prevented the use of the source in the first experiment because, in it, the sodium iodide crystal was suitable for use merely as a detector of gamma radiation. A 2" x 2" cylinder of sodium iodide was found to cause an appreciable amount of total absorption of the 1.38 Mev gamma ray of Na^{24}, producing a line spectrum which separated the 1.38 Mev gamma ray from the 2.76 Mev gamma. Hence, essentially, the experiment was the same as the original except that, instead of comparing total gamma count rates, the ratio of the gamma intensities of the two sources was found by obtaining gamma ray spectra and comparing the areas under the total capture peaks. This produced considerable advantages in other directions too, as will be seen.

The gamma ray spectra were obtained electronically by using a Hutchinson-Scarrot pulse analyser (28). The source,

contained in a thin circular glass vessel, 22c.c. volume, was placed close to the flat end of the crystal. The source and crystal were shielded by lead.

The procedure was to fill the source holder with potassium hydroxide solution and obtain the spectrum. A trace of ^{24}Na was added and another spectrum taken. Subtraction of the two spectra gave the ^{24}Na gamma ray spectrum alone, and subtraction of background from the first left the ^{40}K gamma ray spectrum. These are shown in Fig.22. The relative beta emission rates were determined by spreading a sample of the mixed source on the wall of the proportional counter. By using the mixed source it was ensured that no errors could by introduced in relative source strengths used for beta and gamma measurements. Counting rate measurements were then made over a period of days until the rate became constant after the ^{24}Na had completely decayed. Knowing the background of the counter it was easy then to obtain the intensity of the ^{40}K beta emission and to calculate the relative beta emission rates of the two sources at the time when the gamma spectra were being obtained. A half-life of 15.05 hrs. (29) was assumed for ^{24}Na.

The best illustration of the superiority of the new technique is obtained by considering in how far the corrections required for the first method apply to the second.

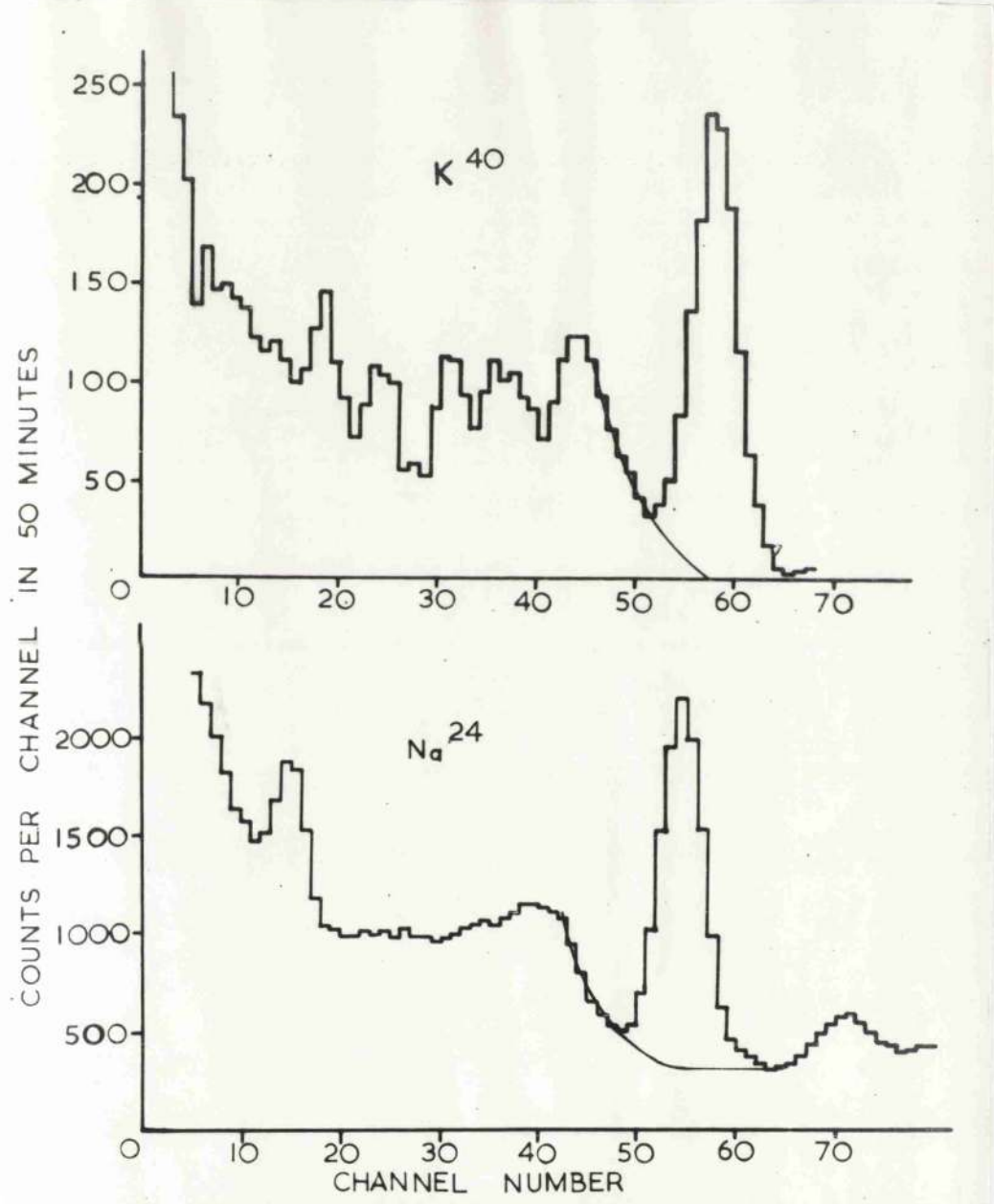

FIG.22. GAMMA RAY SPECTRA FOR THE DETERMINATION OF THE BRANCHING RATIO OF K^{40}. (SECOND METHOD)

CORRECTIONS REQUIRED FOR SECOND METHOD.

(i) Since the beta rays of ^{40}K and ^{24}Na cover approximately the same energy ranges, backscattering of electrons would not be expected to produce relative changes in the beta counting rates of the two sources. In fact it is now known that even with ^{60}Co and ^{40}K no errors were caused by backscattering but the experiments which led to this conclusion had not been completed when the present work was done. By using mixed sources, and since the beta ray energies are similar, it was also ensured that any absorption and scattering of the beta rays in the source layer would be the same for both ^{40}K and ^{24}Na.

(ii) Once again the Geiger counter anti-coincidence ring was not employed because of the danger of losing some ^{24}Na beta-ray counts by the detection in a Geiger of an associated gamma ray. Final measurements of the ^{40}K beta counting rate after the decay of the ^{24}Na were checked with full anti-coincidence shielding.

(iii) Bremsstrahlung could not cause errors in the gamma ray peaks because, for ^{40}K, the maximum energy of the beta ray is less than the energy of the gamma ray, and for ^{24}Na the two energies are so close that any contribution to the gamma ray peak would be quite negligible.

(iv) A correction for coincidences between the gamma rays of ^{24}Na had to be made. This could be calculated since the strength of the source was known.

(v) Errors originating in the transmission of the gamma rays through the source were expected to be small because (a) the source was much thinner than before, (b) the two sources were mixed, so that the gamma rays traversed the same medium, (c) the energies of the gamma rays were much closer in this experiment than when ^{60}Co was used as the calibrator.

(vi) A correction for the slightly differing efficiency of the crystal for the two gamma rays was calculable, as before. Hence, in this second experiment, corrections had to be applied for coincident detection of gamma rays and to allow for changes in the efficiency of the crystal. Both could be calculated. One new correction had to be made to allow for the dead-time of the pulse analyser, but this was small because the counting rates were low.

RESULTS.

The relative intensities of the gamma ray lines of the two sources were found by estimating the areas under the gamma ray peaks within the boundary lines shown in Figs.22. To increase the accuracy the limits were first determined for the peak in the K^{40} gamma spectrum, then transfermed to the

^{24}Na peak. The final result gave a branching ratio of

$$0.121 \pm 0.004.$$

where the limits of error refer mainly to the estimated errors in finding the areas under the gamma ray peaks.

DISCUSSION.

The two determinations of the gamma-beta ratio of ^{40}K have given consistent results. The second method was basically more accurate as well as being quicker in practice, the main doubtful point being the accuracy of the estimations of the areas under the peaks in the gamma ray spectra. The final results are in good agreement with the value of 0.123 selected by Endt and Kluyver (30) as the weighted average of all previous measurements of the ratio. If the gamma-beta ratio is a true indication of the branching ratio there can remain little doubt that the branching ratio is appreciably greater than that found in the best geological experiments to date. *

An unsuccessful attempt was also made in the present work to obtain a direct estimate of the branching ratio by observations of the K x-rays and Auger electrons from a thin solid source of potassium in a proportional counter. Potassium chloride, in which the potassium was enriched 50 times in the K^{40} isotope, was examined as a wall mounted

* See note on page 97.

source 5 micrograms/cm^2 thick in a small counter. The source thickness was only a fraction of the expected path length of an Auger electron but, in spite of this, little evidence for a K-capture branch was obtained in the spectrum. A slight peak, much less intense than expected, was observed but it appeared to be too low in the energy scale to be related to the argon X-ray emitted in the transition.

There was no obvious reason for the failure of the experiment but, recently Drever and Moljk (31) have observed a similar effect. They examined the K-capture of Cl^{36} as a gas in a proportional counter and then permitted the chlorine to condense on the wall of the counter to form a solid source with an average thickness of only 0.03 micrograms/cm^2. The K-capture peak shifted to a lower energy and reduced to about one tenth of its former intensity. It seems that for some reason the radiations do not emerge from the source with the expected energy, even when the source is very thin, or that the proportional counter does not function normally, when the primary radiations have a very low energy, causing the ionisation to take place far from the central wire. There is little doubt that the only really satisfactory way of obtaining the K^{40} branching ratio directly will involve the use of ~~the~~ a gaseous sample of the element. No simple compounds of potassium are gaseous at ordinary temperatures but the recent success of Moljk et al.

(12) in operating proportional counters at high temperatures may make possible a direct observation of the K^{40} branching ratio. Meanwhile the gamma-beta ratio experiments probably give the closest approach to the correct value of the branching ratio.

The discrepancy between the physical and geological estimates still stands. The latest geological value of 0.09 obtained by Shillibeer et al. (23) has been attacked by Kohman (32) because of the use of the $\frac{Pb207}{Pb\ 206}$ age for dating the specimens. This is likely to make the ages too large and lower the calculated branching ratio. This would be in accord with the physical determinations.

As for the physical measurements, the branching ratio rests mainly on the validity of the decay scheme assumed for K^{40}. While the scheme now seems unassailable it was thought useful to check some of the features by further experiment. These investigations and the reasons for them are discussed shortly.

THE HALF-LIFE OF K^{40}

In addition to the branching ratio, the potassium-argon dating technique requires the half-life of the K^{40} decay. This was estimated from the beta counting rate

and branching ratio obtained in the first method, assuming a backscattering factor of 0.45 ± 0.015 for K^{40} beta rays from the steel wall of the proportional counter (see Ch.7). The figure obtained for the total half-life of K^{40} was

$$(1.28 \pm 0.02) \times 10^{9} \text{ years}$$

taking Nier's value of $0.0119 \pm 0.0001\%$ for the abundance of K^{40} in ordinary potassium (33). This half-life is close to the half-life of 1.27×10^{9} years found by Sawyer and Wiedenbeck (10) in their classic investigation of the decay of K^{40} but slightly smaller than the 1.31×10^{9} years preferred by Endt and Kluyver (30) and by Burch (18).

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

It has been assumed in the preceding sections that the gamma-beta and the branching ratio of K^{40} are identical, in accordance with the decay scheme shown in Fig.18. There has not been any really satisfactory direct proof of the equivalence of the two ratios, because even the best of the direct measurements of the branching ratio, by Sawyer and Wiedenbeck (10), is subject to sizeable errors. It seemed to be just possible that the difference between the physical and geological determinations of the branching ratio could be explained by the assumption of an incorrect decay scheme. The gamma-beta ratio can differ from the

true branching ratio if

(i) an appreciable number of electron capture transitions proceed directly to the ground state of $^{40}_{A}$

(ii) the gamma ray follows the beta decay to Ca^{40}, instead of appearing in the K-capture branch.

(iii) more than one gamma ray follows electron capture. If (i) is true then the real branching ratio is greater than the gamma-beta ratio, and will increase the discrepancy between the two methods of determining the branching ratio. If (ii) is true then there is no relation between the gamma-beta ratio and the branching ratio. If (iii) is true then the gamma-beta ratio will exceed the branching ratio as appears to happen in practice. In fact, the gamma-beta ratio was almost exactly twice the earlier geological values, which were still current when the present investigation was begun. This suggested that each capture event might be followed by two gamma rays. However unlikely this appeared, it was desirable to have direct experimental proof that it did not happen.

What is probably the most satisfactory evidence for the decay scheme as it stands was provided by mass measurements of A^{40}, K^{40} and Ca^{40} by Johnson (34). He

showed that the energy available for the decay of K^{40} to A^{40} and Ca^{40} is 1.49±0.07 Mev and 1.30±0.07 Mev respectively. If this is correct then the possibility of the gamma ray occurring in the beta decay branch must be discounted, because the gamma ray energy, 1.46 Mev according to the best of the recent measurements (35,36), exceeds the energy available for beta decay. Equally, the energy available for the decay to A^{40}, according to Johnson's figures, does not permit more than one gamma ray unless the second is very soft. Nor have particle excitation experiments (30) revealed the presence of any excited state in A^{40} below the 1.46 Mev state observed in the decay of K^{40}.

Sawyer and Wiedenbeck's observation that the electron capture disintegration rate is approximately equal to the gamma emission rate shows that the probability of electron capture to the ground state of A^{40} is small. This is supported theoretically by Morrison (14). The absence of 0.511 Mev gamma rays (37,38) and of 180° coincidences (39) shows that the probability of decay by positron emission is also small. This, too, is confirmed by Morrison.

Direct evidence for the decay scheme, in the form of coincidence measurements, is more nebulous because of the difficulty of obtaining coincidences with the low disintegration rates available. Coincidences between X-rays

and Auger electrons and the gamma rays have been reported (40) but the coincidence rates were extremely low. Searches for beta-gamma coincidences have given negative results (41,42), but on the whole, it seemed advisable to repeat the coincidence work abd check the purity of the gamma radiation with more efficient apparatus.

GAMMA RAY EXPERIMENTS.

The gamma ray spectrum of K^{40} was obtained by packing 330gm. of pure KCl round a 2"x2" sodium iodide crystal with sufficient absorber to remove all beta rays. After amplification the output from the photomultiplier was analysed on a multi-channel electronic kicksorter to give the spectrum shown in Fig. 23. Comparison with the gamma rays emitted by Na^{22} (1.277 Mev) and by Na^{24} (1.38 Mev) showed that the energy of the gamma ray emitted in the decay of K^{40} is 1.46 ± 0.01 Mev. This is in excellent agreement with other recent determination s (35,36).

Further examination of the spectrum of the gamma rays emitted by the source failed to reveal the presence of any other gamma with an energy greater than 100 Kev. Experiments using a proportional counter showed no gamma ray with an energy less than 100 Kev. It is therefore concluded that no gamma rays softer than the 1.46 Mev

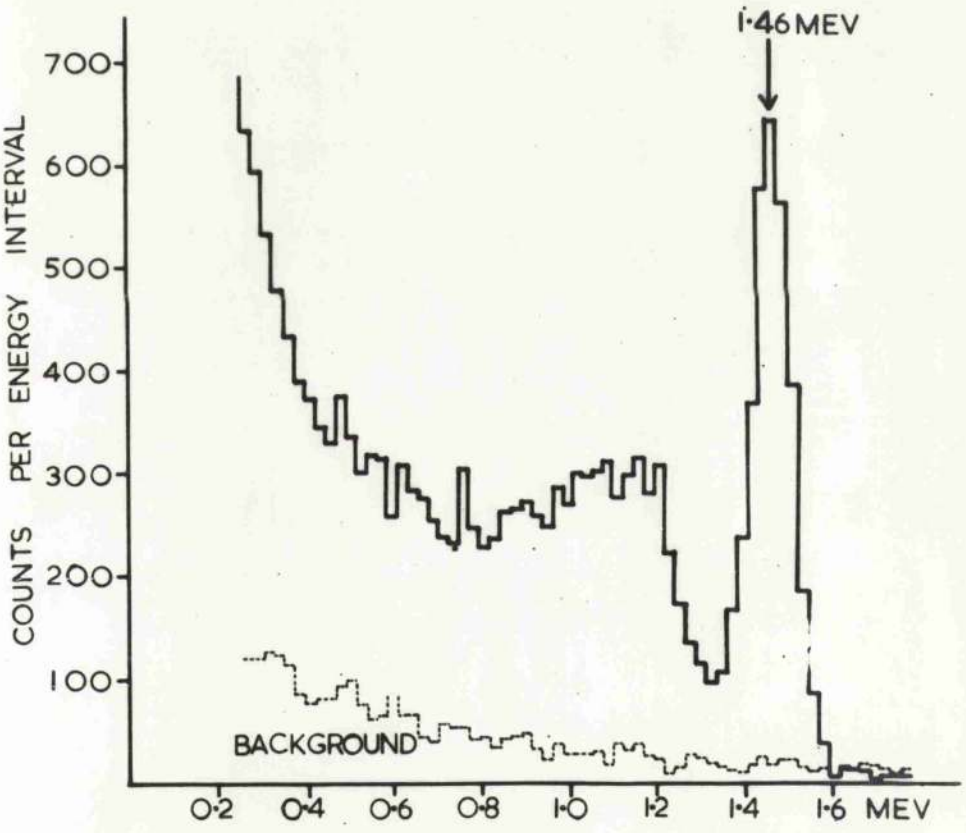

FIG.23. GAMMA RAY SPECTRUM OF POTASSIUM-40 OBTAINED WITH SODIUM IODIDE SCINTILLATION SPECTROMETER.

line are emitted in the decay of K^{40}.

The only other way in which the rate of gamma emission could exceed the rate of electron-capture would occur if two gamma rays were emitted with energies so close to 1.46 Mev that the resolution of the scintillation spectrometer was not sufficient to separate them. From mass considerations this is an extremely unlikely proposition and was ruled out by examining the region of the K^{40} gamma ray spectrum beyond 1.46 Mev. This part of the spectrum is shown in Fig.24, from which the background spectrum has been subtracted. Also shown in Fig. 24 is the same region of the spectrum of gamma rays from a Co^{60} source bearing a similar geometrical relationship to the crystal. The spectra are normalised so that the gamma ray peaks are of equal intensity and can be compared directly. In the Co^{60} spectrum there is a well defined peak A at 2.5 Mev which corresponds to the coincident detection of the 1.17 and 1.33 Mev gamma rays. The edge B is obtained when the Compton edge of one gamma ray is carried forward by the full energy of the other. On the other hand, in the K^{40} spectrum no integration is observed at 2.92 Mev and this rules out the possibility of the emission of two high energy gamma rays in coincidence. A more conventional coincidence experiment using two sodium iodide crystals also failed to show gamma-gamma coincidences. Hence it can

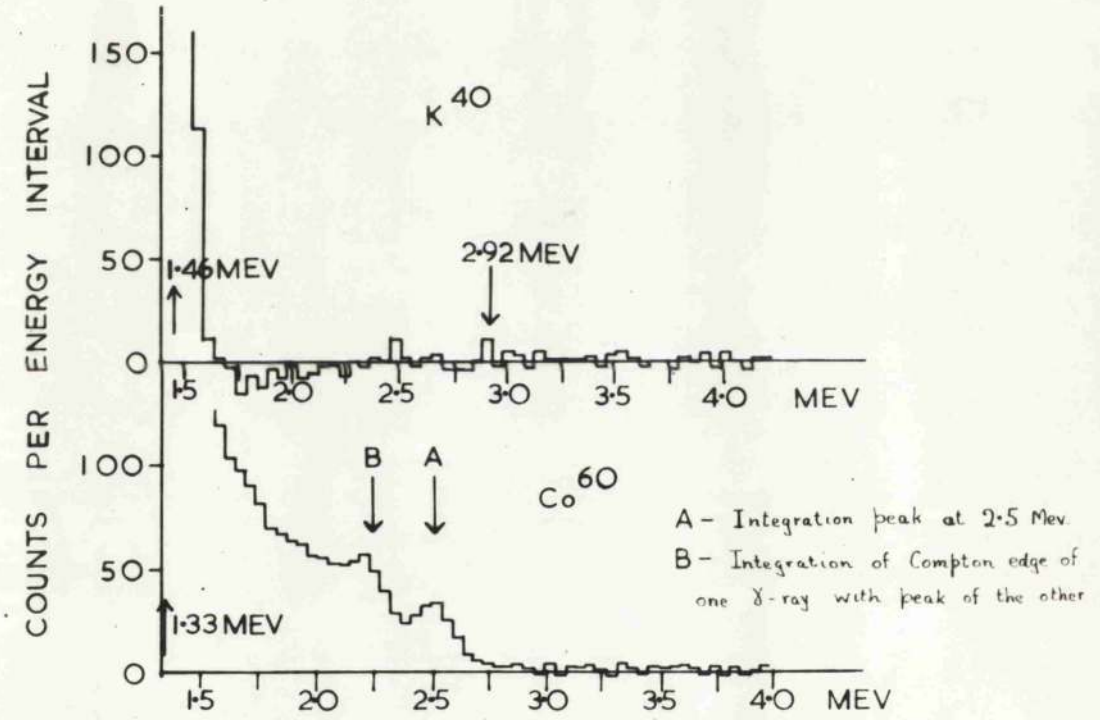

FIG.24. HIGH ENERGY REGION OF K^{40} AND Co^{60} GAMMA RAY SPECTRA.

safely be concluded that only one gamma ray is emitted in whichever branch of the K^{40} decay it arises.

A SEARCH FOR BETA-GAMMA COINCIDENCES IN THE K^{40} DECAY.

The beta rays were detected in a disc of plastic phosphor (1.5%1:1:4:4 tetraphenyl 1:3 butadiene dissolved in polystyrene). A 0.001" thick foil of aluminium on top of the disc reflected the light on to the cathode of the photomultiplier, and also carried the source, 60 m.gms. of KCl enriched 50 times in the K^{40} isotope. The source was covered by a thin sheet of plastic for protection, and by a thick steel foil to absorb beta rays. On the other side of the steel came the gamma ray detector, a 1" cube of sodium iodide packed in magnesium oxide reflector. The whole system was encased in 6" of lead to reduce background. The outputs from the two photomultipliers were amplified and applied to a coincidence circuit whose resolving time was 6.2×10^{-7} sec. The coincidence rate and the separate counting rates were recorded.

The presence of the potassium source was found to increase the coincidence rate by $(1.43 \pm 0.02) \times 10^{-3}$ coincidences per detected beta ray. Since part, at least, of this positive effect could be caused by coincidences between beta rays and bremsstrahlung, a control experiment

was performed using P^{32}, a pure beta emitter. This proved that $(1.04 \pm 0.016) \times 10^{-3}$ coincidences per detected beta ray could be ascribed to this cause. This did not completely account for the observed coincidence rate with K^{40} but calculations based on the efficiency of the apparatus showed that the remainder could reasonably be accounted for by scattering of the K^{40} gamma rays in the system. It is concluded that there is no possibility of beta-gamma coincidences in the decay of K^{40}.

CONCLUSIONS.

Two separate experiments have given values for the ratio of the intensities of the gamma and beta emission from K^{40} which are equal, within experimental error. The weighted mean of the two results gives for the gamma-beta ratio a value of

$$0.123 \pm 0.003$$

Assuming the validity of the commonly accepted decay scheme of K^{40}, a scheme which is given added weight by new coincidence measurements, the true electron capture-beta decay ratio for K^{40} must be greater than, or equal to (0.123 ± 0.003). This allows for the possibility of some electron capture to the ground state of A^{40}.

The energy of the gamma ray following the decay of K^{40} is confirmed to be

$$1.46 \pm 0.01 \text{ Mev.}$$

The total half-life of the K^{40} isotope has been re-determined to be

$$(1.28 \pm 0.02) \times 10^{9} \text{ years.}$$

Postscript Note:

I am informed privately by Dr. Curran that the potassium-argon dating method was discussed at the Gordon Conference on Nuclear Chemistry held recently in the U.S.A. Clear evidence of diffusion of argon in minerals has been obtained by the Pasadena team and the conclusion is that 12.1 to 12.4 % for the branching ratio of K^{40} is now perfectly compatible with mineral age work. The paper containing this information was read to the conference by G.J. Wasserburg. This result is very gratifying, confirming, as it does, the work discussed here.

7. SOME MEASUREMENTS ON THE SCATTERING OF ELECTRONS FROM ALUMINIUM AND STEEL.

Introduction.

To reduce to a minimum the scattering of electrons in the material on which a radio-active source is deposited it is often the practice to use a very thin film of a substance like nylon which contains atoms of low atomic number. This approach could not be taken in the foregoing experiments in which, to obtain the benefits of a thin source with a reasonable counting rate, the samples had to cover a relatively large area on the wall of the proportional counter. Backscattering of electrons out of the wall of the counter will affect the counting rate to an extent which must be discovered.

Published data on the backscattering of electrons is not very consistent. There is good reason for believing that this may be partly due to the different geometrical arrangements of source and counter in experiments in which external sources were used, because of the anisotropic distribution of reflected electrons (1-4). There seems to be general agreement that the intensity of backscattering increases as the atomic number of the backing material increases (1-3), 5,6) and that the angular distribution of the backscattered radiation changes. There is some doubt about the exact variation, if any, of the intensity of backscattering

with the energy of the incident beta particles. This is especially so in the intermediate energy range from 200 Kev to about 1.5 Mev which was of most importance for the work on natural radio-elements. The backscattering factor in this energy range is sometimes said to be constant (1) while others find that it increases with energy over at least part of the range (2,5) and yet others that it decreases with increasing energy of the incident electrons (7,8). In how far these variations are caused by different geometrical arrangements is not clear.

The first potassium branching ratio experiment required an exact knowledge of the difference in the amount of backscattering suffered by the beta rays from Co^{60} and K^{40} at the steel walls of the proportional counter. Since the literature did not contain convincing data for the 2π counting geometry employed in the proportional counter experiments, it was felt best to make some measurements of the backscattering phenomenon using as similar a geometrical arrangement as possible.

The simplest and quickest way of obtaining results is to deposit the source on a thin nylon film, which in itself causes negligible backscattering, and to take counting rates with, and without metal foils placed behind the source. The addition of a thin film of nylon between

the source and metal backscatterer makes the arrangement different from the practical case in which Co^{60} and K^{40} were deposited directly on the metal lining of the counter. Some experimenters (2,8) quote evidence which seems to suggest that the presence of a thin non-conducting film considerably reduces the backscattering from the metal. This is an important point which strikes at the basis of almost all previous measurements of the backscattering of electrons, but, so far as the author is aware, has never been rigorously examined. To investigate this matter some measurements of backscattering were made using the nylon film technique and repeated with the sources deposited directly on the metal scatterers. The main difficulty encountered in making scattering measurements on sources deposited directly on the metal arises from the necessity of having a comparison source on nylon with zero backscattering. To permit the amount of backscattering to be calculated, the absolute disintegration rate of each source must be known, or, at the least, the ratio of the absolute disintegration rates is required. A quick and accurate method of measuring the relative strengths of the sources was evolved and will be described later.

In no way was the investigation meant to be an exhaustive study of the problem, but was undertaken

primarily to obtain a correction to be applied to the beta counting rates in the K^{40} branching ratio experiment. With the exception of a few measurements on the backscattering of positrons only problems relating directly to a determination of the backscattering to be expected of Co^{60} and K^{40} beta rays were considered. But electron scattering is an important topic in its own right and, although the investigation is rather out of the main stream of the work covered in this thesis, the problems seemed to merit a more detailed treatment than could be given in a passing reference during the discussion of the branching ratio experiments.

Only 'saturation' backscattering from thick sheets of metal is considered, because this is the only case of interest for the proportional counter investigations of the natural radio-elements.

EXPERIMENTAL ARRANGEMENT AND PROCEDURE (First Method).

The apparatus, which is sketched in Fig. 25, was very similar to that used by Balfour (6) who investigated the scattering of rather lower energy electrons than are of interest here. It consisted of a large (14cm.diameter) proportional counter with the addition of a large side

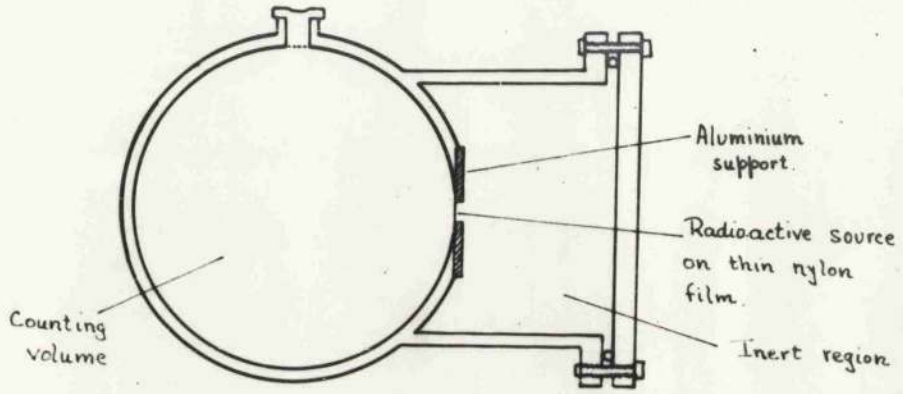

FIG. 25. PROPORTIONAL COUNTER USED FOR MEASUREMENTS ON BACKSCATTERING.

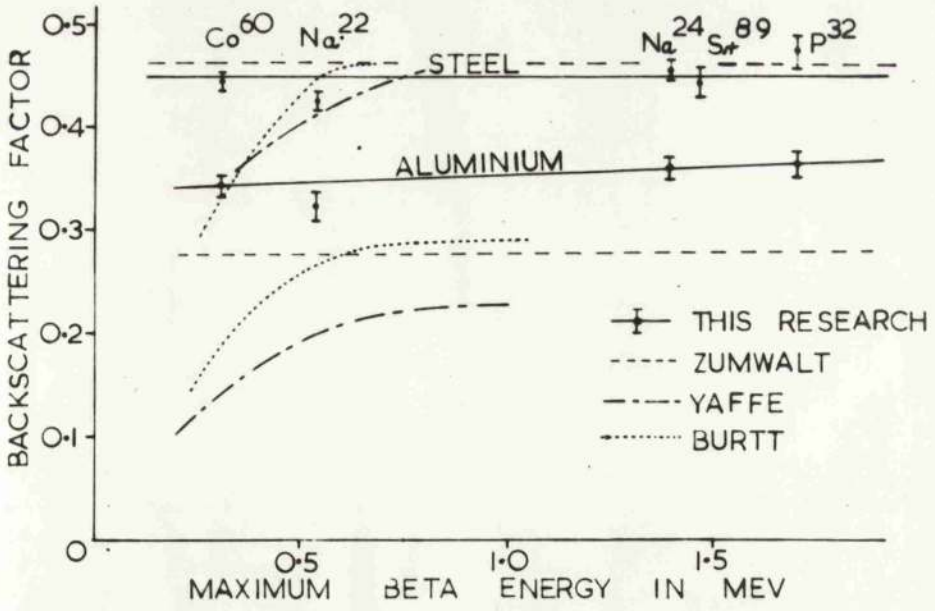

FIG. 26. EMPIRICAL BACKSCATTERING FACTORS.

chamber. A 1" diameter hole in the cylindrical wall of the tube opened into the side chamber. Over the hole in the counter could be placed a thin aluminium sheet with a ½" diameter hole across which was stretched a nylon film carrying a radio-active source. The whole arrangement was so constructed that, in so far as the sensitive volume of the main counter was concerned, the source was effectively on its cylindrical wall The side chamber was large so that electrons escaping through the nylon behind the source had negligible chance of re-entering the counter after scattering at the walls of the side-chamber. In this way it is possible to have a source examined under 2π solid angle conditions and supported on an extremely thin backing. Since the gas pressures were equal on each side of the film no stresses were imposed on the nylon. Metal plates could be placed behind the nylon to act as backscatterers. Access could be obtained via the side chamber to change sources and backscatterers. This prevents any disturbance of the counting chamber itself.

The counter was filled with 15 cm. pressure of methane, the pressure and atomic number being kept low to reduce any scattering in the gas immediately behind the source where the solid angle for re-entry of the electrons into the counter is large. The energy resolution was rather poor but since the counter was being used purely as a detector of beta rays this

was of no importance. The edge of the 8 Kev fluorescence x-ray line of copper gave a sufficiently good energy calibration to ensure that the counting measurements were always made at the same level. As in the potassium branching ratio experiments all electrons with an energy greater than 200 e.v. were recorded. This permits the findings to be applied directly to the branching ratio measurements.

The counter was opened from the side to permit changes of source or backing and refilled. This was considered better than a mechanical device for changing the backing metal because the foils could be placed hard against the nylon support, simulating 2π solid angle coverage as closely as possible. Tests with the same source showed that counting rates were reproducible from one filling of the counter to the next. Experimental proof that the nylon film on which the source was deposited did not itself cause any appreciable backscattering or absorption was obtained, by reversing the source so that the nylon lay between the source and the sensitive volume of the counter. The change in counting rate on reversal of the source was always less than 1%.

Thus this first series of experiments was carried out by preparing sources of various beta emitters on a nylon foil, less than 10μgm./cm^2 thick, and obtaining the

apparent disintegration rate with, and without thick metal backings. The backscattering factor, defined as

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

was calculated for each. Steel and aluminium scatterers were used with sources of ^{24}Na (1.39 Mev), ^{32}P (1.7 Mev), ^{60}Co (0.306 Mev), ^{89}Sr (1.46 Mev) and ^{22}Na (0.542 Mev positrons) to cover the required energy range, since ^{40}K could not itself be treated because of its low disintegration rate. End-point energies of the beta spectra of the sources are given.

EXPERIMENTAL PROCEDURE (Second Method).

This procedure was devised to enable backscattering factors to be obtained when the radio-active material was deposited directly on the metal scattering foils. A first attempt was aimed at producing sources of equal absolute disintegration rate on nylon and on the metals but this proved extremely difficult and the required accuracy could not be obtained. Instead, another technique was developed in which the relative 'true' strengths of the sources were monitored by a gamma ray count while the beta ray count gave 'apparent' strengths. From these the backscattering factor could be calculated.

Beta-emitting sources which also produced a high energy gamma ray in their decay were chosen. If the energy of the gamma ray is high, greater than 100 Kev, say, the response of the proportional counter at the pressure and with the gas used will be negligible and the counting rate will be caused by the beta rays only. Approximately equal quantities of suitable sources were deposited on nylon, aluminium and steel and the beta counting rates observed. The relative strengths of the sources were then monitored by placing each, in turn, in an absolutely fixed position near a simple sodium iodide scintillation counter. A few subsidiary experiments proved that, with the thickness of metal used in the experiment, the gamma ray intensity observed in front of the source did not depend on the metal on which the source was mounted.

Those sources, ^{22}Na ^{24}Na and ^{60}Co which were used in the first series of experiments, but which were also suitable for the second method, were tested.

RESULTS AND DISCUSSION.

The first result of the experiments was that the backscattering factors did not depend on the method of measurement, whether direct or by the gamma monitoring scheme. This conclusively proves that the insertion of a

thin organic film between the source and the metal has no effect on the backscattering.

The amount of backscattering produced by a steel plate was found to be independent of energy in the range of energy covered in these experiments. The result of averaged measurements of the backscattering factor are shown in Fig. 26 where the abscissae represent the maximum energy of the beta rays from each source. On average it was found that

$$0.450 \pm 0.015$$

of the electrons entering a thick steel support are reflected back into a 2π solid angle. This means that backscattering can be neglected in the potassium branching ratio experiments since only the ratio of the intensities of the beta emission from each source is required.

For aluminium the average backscattering factor was found to be

$$0.35 \pm 0.03$$

although in this case there is slight evidence for a slow increase in the amount of backscattering as the energy of the beta rays increases.

For both steel and aluminium and in both methods of measuring the backscattering factor, the positron emitter ^{22}Na gave a slightly lower value. The difference amounted to about a 5% reduction in the amount of backscattering.

Certainly there was no evidence to support the findings of Seliger (3,9) who reported positron backscattering factors which were about 30% less than for electrons.

Fig. 26 also shows some values obtained by other workers. In general, the agreement is fairly close for backscattering from steel, but there is a tendency for the amount of backscattering measured by others to fall off with energy, as shown. No measurements on geometries with less than 2π solid acceptance angle were attempted here.

CONCLUSIONS.

A brief investigation of backscattering showed that the intensity of backscattering from steel does not depend on the energy of the incident electrons in the energy range investigated. A slight increase with energy is possible in the amount of scattering obtained from aluminium. The introduction of a thin film of organic material between a radio-active source and the metallic scatterer does not affect the amount of backscattering from the metal into a 2π solid angle. Backscattering of positrons is apparently slightly less than for electrons.

8. SOME CONCLUDING REMARKS ON PROBLEMS ENCOUNTERED IN THE FOREGOING WORK ON NATURAL RADIO-ACTIVITY.

This concluding chapter contains a brief summary of the main results of the experiments discussed previously and some thoughts on the problems encountered in the work on the natural radio-elements.

^{150}Nd is now certain that ^{150}Nd is stable. The absence of beta rays and considerations based on recent mass measurements of ^{150}Nd and ^{150}Sm lead to this conclusion, which is not in conflict with any nuclear stability rule. Probably the activity reported in some of the earlier investigations was caused by radio-active impurities in the samples.

Beta decay in ^{176}Lu, followed by the emission of three gamma rays in cascade, has been confirmed. In addition, evidence for an electron capture branch of the decay has been observed. This is to be expected from stability criteria, because both ^{176}Hf and ^{176}Yb lie within the beta stability limits for even-A, even-Z nuclei. The suspected occurrence of gamma rays in the electron capture branch is to be expected both from beta decay theory and from the Bohr-Mottelson nuclear shell theory. Their presence is required to speed the electron capture transition because of the high spin of the ^{176}Lu ground state, and their presence

is likely because Yb^{176} lies within the group of elements to which the Bohr-Mottelson theory especially applies, and low-lying rotational states are predicted for such nuclei. The observed intensity of the electron capture transition lies within the limit proposed by Arnold (1) from his investigation of the gamma rays. In the present experiment reliance for the electron capture branch has been placed mainly on proportional counter measurements where the geometry of the source is well defined and thin sources are possible. Even so, the estimate of the branching ratio of the decay tends to be rather sensitive to specific assumptions about the intensity of backscattering, not only of the beta rays, but also of the rather low energy internal L-conversion electrons. For this reason discrimination between the two branches of the decay in a 4π counting geometry would be desirable, as proposed earlier. A similar type of experiment would probably give a better indication of the shape of the beta spectrum than could be obtained in the 2π geometry where distortion is introduced by the other products of decay.

The failure to detect radio-activity in the Re^{187} - Os^{187} group raises interesting possibilities. The recent discovery of radiogenic Os^{187} in rhenium minerals (2) confirms the stability of Os^{187}, as observed, but equally

implies instability in Re^{187}, instability which could not be found in the proportional counter. Certainly, the earlier suggestions (3-5) for a 43 Kev transition are completely ruled out. It seems possible that Libby and co-workers, who were responsible for the main work on Re^{187}, might have run into trouble with radio-active contamination in the absorbing foils which were used to measure the energy of the supposed radiation. Indeed, this much is suggested in a recent paper (6) which proposes an 8 Kev transition energy for Re^{187}. This may still be suspect on the grounds that aluminium absorbing foils were used. According to experience gained in the present work, aluminium is a dangerous metal to use in low activity work, because of the possible occurrence of a slight variable activity in the metal. The absence of any detectable beta emission from Re^{187} when examined in the proportional counter suggests a very low energy transition. Exactly how low the energy is, is difficult to say, because of the suspected poor operation of a proportional counter when required to measure low energy emissions from wall-mounted solid sources. However, it seems unlikely that the energy can exceed 1 Kev (the lower limit of the proportional counter measurements) because even if the efficiency of the counter fell off at low energies, quite a small fraction of the expected disintegration

rate should have been easily detected above background. The occurrence of radiogenic Os^{187} in geological deposits implies that the half-life of Re^{187} does not exceed, say, 10^{11} years. Since the transition is obviously of very low energy it cannot be highly forbidden to have a half-life as low as 10^{11} years. This supports the proposal, based on nuclear shell theory and confirmed by recent measurement of the spin of Os^{187}, that the transition is only first forbidden with a spin change of 2.

Recently, in order to explain why geological ages based on the Rb^{87}-Sr^{87} transition tend to be high, Kohman (7) has suggested that the Rb^{87}-Sr^{87} transition may be speeded by the unobserved process of 'bound' beta decay. This process is proposed to involve creation of the electron directly into an atomic orbital of the disintegrating atom and the removal of virtually all of the disintegration energy by the neutrino. Theoretical treatments of the process have been given (8-14) but the calculations have been made only for allowed transitions and do not give any agreement on the quantitative importance of the process. However, there is general agreement that it should be more important in forbidden transitions. Thus, if this process can be present in sufficient degree to explain the difficulties encountered in strontium geochronometry it could be the

for heavy elements and low transition energies and may be more important

predominant mode of decay in ^{187}Re where Z is high and the transition energy very low. Certainly, to investigate the matter further, special techniques will be required with ^{187}Re since it has by far the lowest energy of any known beta emitter. It does not seem likely that the decay can be adequately examined from other than a gaseous source. If bound beta transitions are truly ~~observed~~ unobservable, that is, if the neutrino carries off excess energy and no photons are emitted with high enough energy to be detected in the counter, then the decision whether or not the process is ever important may well hinge on the decay of ^{187}Re where conditions for its occurrence are most favourable. Careful geological determinations could provide an estimate of the half-life of ^{187}Re to compare with future nuclear counting experiments.

The branching ratio of ^{40}K is greater than, or equal to the gamma-beta ratio which has been found to be 0.123 ± 0.003 in the present investigation. This figure has been confirmed by other work published very recently (15) and is considerably in excess of the best geological estimates to date. The discrepancy* probably arises in the difficulties encountered in the geophysical measurements or in loss of argon by diffusion from the minerals throughout geological time. It does not, any longer, appear possible to blame

* Probably no longer exists. See note on page 97.

measurements of the gamma ray emission for the difference, since recent work is fairly consistent. In complete absence of any reason for believing that the gamma intensity should not be a good indication of the rate of decay by electron capture, we must therefore assume that the rate of transition of K^{40} to A^{40} is known. Thus, if the physical branching ratio is at fault it must be due to a wrong value of λ_β, the beta decay constant. One might consider bound beta decay reducing the observed value of λ_β below the true value and hence increasing the physically determined branching ratio, although any appreciable amount of bound beta decay in the transition to Ca^{40} seems unlikely, in view of the high energy released in the transition and the low atomic number of the decaying nucleus. However, the measured value of λ_β is also required to obtain the branching ratio in the geological experiments and it can readily be shown that if λ_β is increased (and the physical branching ratio decreased) the geological branching ratio will also decrease and at a faster rate than the physical ratio. In fact, if λ_β is to be adjusted to bring the two branching ratios into agreement it will have to be <u>decreased</u> to about a third of its determined value. In other words, about three times the true beta decay rate of K40 has always been observed

experimentally. This is inconceivable and it seems much more likely that the errors arise in the geological determinations.

Thus in the case of the decay of K^{40} the onus seems to be on the geophysicists to check the validity of their figures. On the nuclear side it would be useful to be able to measure the branching ratio directly, by observation of the products of K-capture.

In conclusion, there appears to be considerable scope for investigating some of the natural radio-elements as gases in a proportional counter in order to obtain more accurate information at low energies. This is especially the case of Re^{187} and K^{40}.

The action of a proportional counter at low energies also requires investigation. It seems possible that the difficulty may arise because, when wall-mounted sources are used, the initial ionisation takes place close to the wall of the counter for low energy radiations. Under these circumstances negative ion formation in the gas may be important. It is known from some tests made in the present work that when certain organic vapours are present in small quantity in the counter, negative ion formation causes severe distortion of the spectrum of x-ray lines, even when the energy is as high as 20 Kev and the primary ionisation takes place throughout the whole volume of the counter. It is

possible that small amounts of impurities in the normal counter gases may capture electrons readily enough to cause distortion of the output when the primary ions are few and in regions of low field strength far from the collecting wire.

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