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ALPHA RADIOACTIVITY OF NUCLIDES WITH ATOMIC NUMBERS LESS THAN 83

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By

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ALPHA RADIOACTIVITY OF NUCLIDES WITH ATOMIC NUMBERS LESS THAN 83

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ABSTRACT

In a survey for alpha activity among cyclotron-produced neutron-deficient nuclides of the elements below lead alpha activity was detected in a number of rare earth nuclides with atomic number greater than 62, that of samarium, and in a gold and a mercury nuclide. A detailed study of the alpha-active nuclides of europium, gadolinium, terbium, and dysprosium was made.

The tren ds of the rare earth alpha decay energies are discussed, particularly with relation to the abnormally decreased neutron binding energies just beyond the closed shell of 82 neutrons.

The relationship between alpha decay rates and energies in the rare earth region is examined. Experimental values of alpha decay energies are used to calculate predicted values of the rare earth alpha decay rates by extrapolation of four different alpha decay rate formulas adjusted to give agreement with alpha decay rate data of even-even type nuclides in the heavy element region. These predicted values are compared with the experimental values, and the problem of the determination of the "effective nuclear radius for alpha particles" discussed.

A brief review of various alpha decay rate treatments is made. A semi-empirical correlation of alpha decay rate data of even-even type nuclides of the heavy elements is carried out using a formula for the effective nuclear radius based on experimental alpha particle bombardment cross sections. Finally, a new many-body type alpha decay rate formula, relating decay rate to nuclear level spacing, is derived and discussed.

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I. INTRODUCTION

Radioactive decay by emission of alpha particles is a commonly observed mode of decay in the isotopes, both natural and artificial, of the elements of atomic number greater than 82, that of lead. However, with the lone exception of the alpha emitting isotope of natural samarium ($Z = 62$), discovered by Hevesy and Pahl[1] in 1933, alpha decay in isotopes of elements below bismuth ($Z = 83$) had not been reported prior to 1949.[2] At various times the problem of alpha stability in these lighter elements has been considered.[3] It had been noted from observation of the general slope of the experimental mass defect curve that most isotopes of mass number greater than about 150 are energetically unstable toward alpha decay. The fact that alpha decay had not been observed (excepting in samarium) in the naturally occurring isotopes of the medium heavy elements could be adequately explained by the quantum mechanical consideration of the rate of penetration of the coulombic potential barrier by escaping alpha particles. The decay rate formulas developed by Gamow[4] and by Gurney and Condon[5] and verified by comparison with experimental alpha decay data in the heavy elements showed the alpha decay rate to be a very sensitive exponential function of decay energy. Thus, the naturally occurring isotopes of medium heavy elements might be unstable toward alpha decay by energies up to about 2 Mev and still have unobservably long alpha decay half-lives (greater than $\sim 10^{16}$ years).

Kohman[6] has made an analysis of the binding energies for alpha particles in medium heavy elements, based on the semi-empirical mass equation of Bohr and Wheeler[7] and the experimental mass defect curve. On this basis Kohman has predicted for the medium heavy elements that those nuclides sufficiently far to the neutron-deficient side of the beta stability region might exhibit observable alpha decay. The mass defect curve is not well enough known in the regions of interest to make very exact predictions. A semiempirical formula for alpha decay energy developed by Feenberg,[8] taking into account the finite compressibility of nuclear matter, leads to the same general conclusions.

Studies[9] of the systematics of alpha decay energy among the heavy elements have shown that, in accordance with the above mentioned theoretical predictions, for a series of isotopes (constant Z) the natural trend of alpha decay energy is to increase as the number of neutrons is decreased, except for a large discontinuity at the closed shell of 126 neutrons. While alpha decay energy generally increases with decreasing neutron number, the energy available for orbital electron capture or positron decay also generally increases(aside from fluctuations due to nuclear shell effects) and, consequently, half-lives for both modes of decay decrease. Whether or not by the removal of neutrons in the medium heavy elements alpha emission would become an important mode of decay before the half-lives became inconveniently short for detection had to be tested by experiments.

The production of such highly neutron-deficient isotopes became feasible with the completion of the 184-inch cyclotron at Berkeley, for it was shown[10] that the highly excited compound nuclei formed by bombardment of

heavy and medium heavy elements with 200 to 400 Mev alpha particles, deuterons, or protons dissipated much of this excitation energy by the evaporation of the small nuclear particles, predominantly neutrons. Thus, fair yields of (d,xn), (p,xn), or (α,xn) reactions with x ranging from one to twelve or more could be reasonably expected at high bombarding energies. The competition of the fission reaction with neutron evaporation was expected to be much less serious in medium heavy than in heavy elements.[11]

It, therefore, seemed worth while to initiate a survey in which targets of various medium heavy elements would be subjected to high energy particle bombardments and then examined for alpha radioactivity.

In 1949 Thompson and co-workers[2] reported some positive results of this survey, the discovery of alpha radioactivity in neutron deficient isotopes of gold and mercury and in isotopes of the rare earth elements containing a few more than 82 neutrons.

The present paper is concerned with the detailed study of the alpha radioactive isotopes discovered in the survey, particularly in the heavy rare earth region. Also, several hitherto unreported alpha-active isotopes in the rare earth region are reported.

It may be worth mentioning at this point that with similar short, survey-type bombardments with 200 Mev protons on tungsten, tantalum, silver, palladium, samarium oxide, and tellurium (Te^{122}) oxide[12] it was not possible with the techniques employed here to detect induced alpha radioactivity other than that ascribable to heavy element contamination. The significance of these negative results is only qualitative; faster, more sensitive techniques may eventually uncover induced alpha activity in the regions where initially

negative results were obtained. In particular, the prospect of obtaining bombardments of good intensity with 100 Mev carbon ions in the Berkeley 60-inch cyclotron[13] has renewed interest in repeating by this new means the survey for alpha radioactivity in these regions. Through carbon ion bombardments of neodymium oxide it has been shown that the highly neutron-deficient isotopes exhibiting alpha activity can be produced without producing large amounts of less neutron-deficient isotopes, whose beta-gamma radiations make the search for small amounts of alpha activity more difficult. Larger amounts of the undesired slightly neutron-deficient isotopes, which add to the beta-gamma background, are inevitably produced by high energy bombardments with the conventional bombardment particles (H^1, H^2, He^4).

II. EXPERIMENTAL METHODS

A. Production of Alpha Active Isotopes

For the production of the alpha active isotopes to be investigated three particle accelerators at the University of California Radiation Laboratory have been used: the 184-inch cyclotron, the 60-inch cyclotron, and the 32 Mev proton linear accelerator.

Rare earth elements were bombarded as the powdered oxides. Materials of the highest available chemical purity were generally used as target materials. Freedom from heavy element impurities (uranium, thorium, bismuth, and lead) was especially important in experiments on short-lived activities where rare earth chemical separations were not feasible. In the 184-inch cyclotron the oxides were generally bombarded within envelopes of 0.00025 inch platinum clamped to a probe within the cyclotron tank. The probe

could be positioned within the tank to expose the target material to bombardment at a radius corresponding to the desired bombardment energy. Where metal foils were bombarded in the 184-inch cyclotron, they were clamped to target holders at the end of the probe. In the 60-inch cyclotron alpha particle, deuteron, or proton bombardments, the rare earth oxides were bombarded in the special target assembly developed for the transcurium isotope work at this laboratory and described in a recent article by Thompson et al.[14] This water cooled assembly exposes the target to bombardment by the deflected beam of the 60-inch cyclotron. The C^{12} ion bombardments of rare earth oxides were made within the vacuum tank, with the oxides wrapped in envelopes of 0.00025-inch tantalum foil, which were in turn clamped to a probe.

B. Detection of Alpha Activity

Alpha activity was detected following bombardments by counting samples in argon-filled ionization chamber counters adjusted with the aid of an oscilloscope such that they would count only the relatively large ionization pulses produced by alpha particles and not the "pile-ups" due to beta-gamma activity. Alpha particles of the energies encountered in this work (2.9 to 5.7 Mev) rapidly lose energy by ionization in traversal of matter. Therefore, it was desirable that samples for alpha counting be made as thin as possible. Samples were usually prepared by evaporation of active solutions on 5 mil thick platinum disks. The disks were heated to red heat in a flame to drive off all remaining moisture and to destroy organic material in order to minimize the self-absorption of alpha activity

in the sample. Despite the fact that target materials of good purity were used, there was often enough alpha activity present from heavy element contamination that it was found desirable to use as the detection device the special argon-filled ionization chamber and 48-channel differential pulse height analyzer.[15] By counting the alpha radioactivity in the pulse analyzer ion chamber a measurement of the pulse height was obtained. The alpha particle energy could then be calculated by comparison with the pulse heights produced by alpha emitter standards of known energy. Since the nuclear coulombic potential barrier for alpha particles is considerably lower for rare earth element nuclei than for heavy element nuclei, the alpha particles emitted by the rare earth element nuclei have energies less than that of any heavy element contamination alpha activities of comparable alpha decay half-lives by at least 1 Mev. Thus, from the observation of alpha particle energy and the alpha decay half-life of the activity alone, an estimate of the atomic number of the alpha active nuclide can be made with an uncertainty not greater than approximately ten.

For each alpha activity observed in the medium heavy elements it would be desirable to obtain the following information: (1) alpha particle energy, (2) half-life of activity, (3) atomic number of alpha active nuclide (element assignment), (4) mass number of nuclide, and (5) other modes of decay, their accompanying radiations, and branching ratios between the decay modes of the nuclide. The experimental methods employed for obtaining such information are discussed in Appendix I.

III. EXPERIMENTAL RESULTS

A number of the alpha emitting nuclides discovered among the neutron-deficient nuclides of the medium heavy elements have been studied individually. Table I lists a number of these alpha emitters with some of their properties. Following the table the experimental results are presented in greater detail. The ± sign with numerical data precedes the estimated limits of error, not the probable error. Experimental uncertainties in this work generally arise from so many sources as to render statistical evaluation of probable error impractical. Figure 1 shows the section of the isotope chart in which the rare earth alpha emitters are situated.

A. Europium Isotopes (Z = 63)

In the first preliminary report[2] concerning this investigation it was stated that bombardment of samarium (Z = 62) oxide with 200 Mev protons produced no significant amount of alpha activity. This and other early proton bombardments of samarium for short periods of time (less than one hour), which also gave negative results, indicated that alpha decay is not a prominent mode of decay among those neutron-deficient isotopes of europium and samarium, whose half-lives are long enough (>3 minutes) to have been observed.

Later it was found possible by longer bombardments of samarium targets with protons or deuterons to produce enough of one europium isotope, Eu^{147}, to observe its alpha decay branching. Comparable amounts of Eu^{145}, Eu^{148}, and Eu^{149} have not shown detectable alpha activity and are presumed to have partial alpha decay half-lives greater by at least a factor of five

Table I
Artificial Alpha Activities in Elements Below Lead

Element	At. No.	Mass No.	Alpha particle energy (Mev)	Measured half-life	Other modes	Branching ratio α/total	Partial alpha decay half-life	Prepared by
Eu	63	147	$2.8_8 \pm 0.1$	24 d ± 2	EC	$\sim 10^{-5}$	$\sim 6 \times 10^3$ y (within factor of 3)	$Sm^{147}(p,n)$ 8.5 Mev $Sm^{147}(d,2n)$ 19 Mev $Sm^{148}(d,3n)$ 19 Mev
Gd	64	148	$3.1_6 \pm 0.1$	> 35 y	--	--	$\sim 1.4 \times 10^2$ y (within factor of 3)	$Sm^{147}(\alpha,3n)$ 36 Mev $Eu^{151}(p,4n)$ 32 Mev
		149	3.0 ± 0.15	9 d ± 1	EC	$\sim 7 \times 10^{-6}$	$\sim 4 \times 10^3$ y (within factor of 3)	$Sm^{147}(\alpha,2n)$ 30 Mev
		150	2.7 ± 0.15	(long)	--	--		$Eu^{151}(d,3n)$ 19 Mev
Tb	65	149	$3.9_5 \pm 0.04$	4.1 h ± 0.2	EC, prob. no β^+	--	--	$Eu^{151}(\alpha,6n)$ 60 Mev $Gd(p,xn)$ 32 - 200 Mev
	65	150 or 151	$3.4_4 \pm 0.1$	19 h ± 1	--	--	--	$Gd(p,xn)$ 100 Mev $Eu^{151}(\alpha,xn)$ 60 Mev
Dy	66	$149 \leq A \leq 153$	4.21 ± 0.06	7 m ± 2	--	--	--	$Tb^{159}(p,xn)$ 100 Mev
	66	$149 \leq A \leq 153$	$4.0_6 \pm 0.04$	19 m ± 4	--	--	--	$Tb^{159}(p,xn)$ 100 Mev
	66	$149 \leq A \leq 153$	$3.6_1 \pm 0.08$	2.3 h ± 0.2	--	--	--	$Tb^{159}(p,xn)$ 100 Mev
Au	79	$183 \leq A \leq 187$	$5.0_7 \pm 0.1$	4.3 m ± 0.2	EC, β^+	α/K x-rays $\sim 10^{-4}$	~ 30d (within factor of 4)	$Au^{197}(d,pxn)$ 190 Mev $Pt(p,xn)$ 120 Mev
Hg	80	$A < 185$	$5.6_0 \pm 0.1$	0.7 M ± 0.2	--	--	--	$Au^{197}(d,xn)$ 190 Mev

than that of Eu^{147}. Following bombardment of separated isotope $_{62}Sm^{147}$ oxide with 8.5 Mev protons and of natural samarium oxide with 19 Mev deuterons, there has been observed a small amount of alpha activity which decays with about a three week half-life.

After these bombardments europium activities were separated from the samarium target material by means of the standard cation exchange column elutions described in Appendix 1.3. The eluting agent for the 83°C separation was pH 3.4 citrate solution. Low energy alpha activity was detected, following chemical separation, in the europium fraction.

Energy determination for the europium alpha activity was made by pulse analysis of a thin vaporized sample. The pulse analysis curve is shown in Figure 2, together with curves for the alpha energy standards Gd^{148} and Sm^{147} taken at about the same time. The alpha particle energy for the standard Sm^{147} (a uniform, vaporized sample of Oak Ridge separated Sm^{147} oxide) was taken as 2.18 Mev from the work of Jesse and Sadauskis.[16] The energy for the standard Gd^{148} is taken as 3.16 Mev, the determination of which is described in the following section, III B. By linear interpolation between the standards the alpha particle energy of this europium activity is determined to be 2.88 Mev with estimated limits of error ± 0.1 Mev. The alpha activity peak at 3.16 Mev in the europium sample is presumed to be due to Gd^{148}, formed by alpha particles contaminating the 60-inch cyclotron deuteron beam. The cation exchange separation does not separate gadolinium completely from europium under the conditions used in this work.

Chemical proof that the 2.88 Mev alpha activity is due to a europium nuclide and not to a gadolinium nuclide (which might conceivably have

been produced by alpha particles in the deuteron or proton beams of the 60-inch cyclotron) has been made by subjecting a mixture of a sample containing 2.88 Mev alpha activity and Gd^{148}(3.16 Mev) alpha tracer to a sodium amalgam reduction separation (Appendix I.3). Alpha pulse analysis of activity in the reduced fraction and of activity not reduced showed the predominance of 2.88 Mev alpha activity in the reduced fraction (europium) and of 3.16 Mev alpha activity in that not reduced (gadolinium).

From the decay curve (Figure 3) for the total count rate summed over the 2.88 Mev peak on the pulse analyzer, a value of 26 ± 4 days was deduced for the half-life of the activity. The large limits of error on the half-life determined from this curve arise from the statistical uncertainties attending these determinations of such low counting rates and the fact that decay could only be followed through a decay factor of three.

A general study of the neutron deficient europium isotopes by Hoff, Rasmussen, and Thompson[17] has been recently made by proton and deuteron bombardments of isotopically enriched samarium oxide[12] targets. The isotope Eu^{147} was found to decay predominantly by electron capture with a half-life of 24 ± 2 days as determined from Geiger counter decay curves. None of the other neutron deficient europium isotopes with mass numbers between 144 and 150 have half-lives near 24 days. Hence, the 2.88 Mev alpha activity in europium must arise from Eu^{147}.

Determination of the branching ratio between alpha decay and electron capture in Eu^{147} is difficult, since the detailed decay scheme for the electron capture process in Eu^{147} has not been studied yet. Hence, it is not known what to assume for the counting yield of a Geiger-Mueller

counter for the radiations accompanying electron capture decay of Eu^{147}. In order to make a rough estimate of the α/EC branching ratio in Eu^{147} the decay of the 24 day activity was followed by counting the same thin vaporized sample in the alpha pulse analyzer and in a windowless methane proportional counter (Nucleometer[18]) operated at 5 kev. The counting yield of the Nucleometer for electrpn capture decay is generally much higher than that of a conventional Geiger counter since the Nucleometer counts Auger and conversion electrons of low energies which would be stopped by the window of a Geiger counter. It was found in this laboratory for a few heavy element electron-capturing nuclides whose absolute disintegration rates can be calculated by observation of growth of their alpha active daughters that the mean counting yield of 30 percent for the electron capture process in the Nucleometer is not greatly in error for any of the examples studied. Assuming 30 percent counting yield for the electron capture decay of Eu^{147} in the nucleometer and a counting yield of 40 percent for the alpha decay in the alpha pulse analyzer, the branching ratio was calculated to be roughly $\alpha/EC \approx 1 \times 10^{-5}$. With this branching ratio the partial half-life for alpha decay is about 6×10^3 years. It is evident that the uncertainty of this partial alpha half-life estimate is largely due to the counting yield assumption for EC. It is probably reliable within a factor of three.

B. Gadolinium Isotopes (Z = 64)

1. 3.16 Mev Gd^{148}.--After a bombardment of dysprosium oxide with 200 Mev protons a small amount of a long lived, low energy alpha activity

was found. Subsequent bombardments showed it to be produced in much larger yield by 38 Mev alpha particle bombardment of natural samarium oxide or, in still greater yield, of samarium enriched[12] in Sm^{147}. Also this activity was produced in large yield by 50 Mev proton bombardment of europium oxide and in low yield with 32 Mev protons.

Bombarded material containing this activity was subjected to chemical separation by standard cation exchange column elutions and by sodium amalgam reduction as described in Appendix I.3. In all these separations the long lived alpha activity remained with the gadolinium fraction.

The alpha particle energy of this long lived gadolinium isotope was determined by pulse analysis and comparison with alpha-emitting standards of known energy by use of a pulse generator technique discussed in Appendix I.1. The energy was determined as 3.15_6 Mev from the plot in Figure 12. An earlier determination using fewer standards gave 3.18 Mev. The value $3.1_6 \pm 0.1$ Mev was selected, with conservatively large error limits selected because the alpha energy standards used were of much higher energy.

The 3.1_6 Mev alpha activity is too long lived for a half-life determination by the usual decay methods. A small sample (158 alpha disintegrations per minute) of this activity was counted at intervals during a period of one year in a standard 2π geometry ionization chamber alpha counter and did not decay detectably (<2 percent). These counting data established a lower limit of 35 years for the half-life.

The determination of mass number of the gadolinium isotope giving rise to this activity was made on the basis of the experimentally determined

excitation functions for its production by alpha particle bombardment of enriched samarium (Sm^{147}) oxide,[12] and by proton bombardment of natural europium. The alpha particle excitation function, shown in Figure 4, was determined by bombardment in the 60-inch cyclotron and the proton function by 31 Mev proton bombardment in the linear accelerator, using the modified stacked foil technique described in Appendix I.4. No special collimation to reduce the energy inhomogeneity of the alpha particle beam was used. In the transfer of bombarded oxides to platinum plates for counting, the oxides were dissolved in 12 N HCl, precipitated as the hydroxides by addition of ammonia gas, washed with water, and slurried onto platinum plates for counting.

The element assignment of the 3.16 Mev alpha activity to gadolinium reduced the mass assignment problem to that of deciding whether the excitation function of Figure 4 with reaction threshold 28 - 30 Mev is that of an (α,n), (α,2n), (α,3n) or (α,4n) reaction. The choice should be consistent with the observation in the proton excitation function of about a 30 Mev threshold for the $Eu^{151}(p,xn)Gd^{152-x}$ reaction. Since statistical theory predicts, as bombarding energy is increased, a very gradual onset of reactions in which several neutrons are evaporated, the actual energetic thresholds are probably below the estimates from these insensitive experiments. Theoretical estimates of (α,xn) and (p,xn) energetic thresholds were made by calculation from masses in the atomic mass table of Metropolis and Reitwiesner[19] (based on a semiempirical mass formula of Fermi). Table II lists these values.

Table II

Calculated Reaction Thresholds from Fermi Semiempirical Mass Equation

Reaction	Threshold (Mev)
$Sm^{147}(\alpha,2n)Gd^{149}$	16.9
$(\alpha,3n)Gd^{148}$	24.5
$(\alpha,4n)Gd^{147}$	34.0
expt.	~28 - 30
$Eu^{151}(p,3n)Gd^{149}$	17.9
$(p,4n)Gd^{148}$	25.5
$(p,5n)Gd^{147}$	34.9
expt.	~30

The comparison of the experimental and theoretical values in Table II indicate a probable mass assignment of the 3.16 Mev alpha activity to Gd^{148}, although the possibility of an assignment to Gd^{147} is not too improbable, particularly when it is realized that the calculated thresholds would be lowered by a decrease in neutron binding energy that might be expected for several neutrons beyond the closed shell at 82 (cf. Harvey[20]). The mass assignment to 148 rather than to 147 seems more consistent with the long half-life (>35 years) of this activity, for the even-odd nuclide Gd^{147} should have about 2 Mev more energy available for electron capture decay than even-even nuclide Gd^{148}.

It is possible to make a very rough estimate of the partial half-life for alpha decay of Gd^{148} from the yield of 3.16 Mev alpha activity produced

in the excitation function bombardment of Sm^{147} with alpha particles if an approximate value is assumed for the $(\alpha,3n)$ reaction cross section. From the observed yield for 36 Mev alpha particles with a beam of measured intensity it was determined that $T_{1/2\alpha}\sigma(\alpha,3n) \approx 1.4 \times 10^2$ years barn^{-1}. The (α,xn) cross sections in the rare earth region are not known. If it is assumed that this cross section is about one barn, as was found for the $(\alpha,3n)$ reaction on Bi^{209} at the same energy above the $(\alpha,3n)$ threshold,[21] the estimate of the alpha half-life of Gd^{148} would be 1.4×10^2 years. This estimate may be reliable to a factor of three.

The presence of long lived electron-capturing gadolinium isotopes in the samples containing Gd^{148} make it impossible at present by simple beta-gamma counting to set a significant lower limit on the branching ratio between alpha decay and electron capture, if electron capture is a mode of decay. The possibility that Gd^{148} may be stable with respect to electron capture decay can neither be ruled out nor confirmed by present data.

2. Alpha Activity of Other Gadolinium Isotopes.—Following a bombardment of 200 mg of samarium oxide with 31 Mev alpha particles a chemical separation by standard cation exchange column elution at room temperature (Appendix I.3) was made. A sample plate of the gadolinium and europium chemical fraction was prepared by vaporization. A small amount of 3.16 Mev Gd^{148} was observed in the sample; in addition there was some activity with alpha energy of about 3.0 ± 0.15 Mev which decayed with a half-life of about a week. This 3.0 Mev activity was not observed following intensive deuteron (19 Mev) bombardments of Sm_2O_3 and, hence, cannot be europium. The 3.0 Mev activity was assigned to Gd^{149}, which

has been studied by Hoff et al.[17] and found to decay mainly by orbital electron capture with a half-life of 9 ± 1 days. The α/EC branching ratio is about 7×10^{-6} from comparison counting in the alpha pulse analyzer and the methane proportional counter (Nucleometer) on the same counting yield assumptions as were made for the Eu^{147} α/EC estimation (i.e., α, 0.4; EC, 0.3). This branching ratio figure is probably good to within a factor of three. This branching ratio would correspond to a partial alpha half-life of about 4×10^3 years.

It has been suggested by Kohman[22] that Gd^{150} might be stable with respect to electron capture although it is missing in nature. Thus, like Sm^{146}, Gd^{150} might be so missing because its alpha decay half-life is much less than the age of the elements (i.e. $T_{1/2} \lesssim 10^8$ years). In the hope that the alpha half-life of Gd^{150} might be short enough to detect by cyclotron production a 45 microampere hour cm^{-2} irradiation of 40 mg of Eu_2O_3 with 19 Mev deuterons was made. Gd^{150} should have been produced in fair yield by the reaction $Eu^{151}(d,3n)Gd^{150}$.

Following bombardment the Eu_2O_3 target material was dissolved in 6N HCl, the rare earth hydroxides were precipitated with ammonia, and after redissolving with a minimum of 6N HCl a chemical separation of europium (and partially samarium) from gadolinium was made by the sodium amalgam procedure (Appendix 1.3).

A thin uniform sample of part of gadolinium fraction on a platinum plate was prepared by the volatilization technique (Appendix 1.1). This sample exhibited (Fig. 5) a minute amount (0.45 alpha disintegrations/minute)

of alpha activity of 2.7 ± 0.15 Mev energy. The presence of short range alpha activity in the gadolinium fraction was further confirmed by Dunlavey,[23] who introduced some of the gadolinium fraction into the emulsion of an Ilford C2 photographic plate and allowed it to stand for several days before developing. Microscopic examination of the developed plate revealed a number of alpha particle tracks with a mean range of about 9.7 microns. This range corresponds to an alpha particle energy of 2.7_4 Mev on the range-energy curves of Rotblat.[24]

The assignment of this alpha activity to Gd^{150} can at present be no more than a tentative suggestion based principally on semiempirical considerations of the probable expected alpha decay energy for Gd^{150}. Insufficient time has elapsed to rule out the possibility that this activity could arise from 155 day[25] Gd^{151}.

Sun et al.[26] have reported observing an alpha activity of 7.0 hour half-life from a bombardment of Sm_2O_3 with 30 Mev alpha particles. This activity was assigned by them to gadolinium, since it was not produced by proton (8 Mev) and deuteron (15 Mev) bombardments on Sm_2O_3. They do not mention any alpha particle energy measurements nor any chemical separations. In apparent contradiction to this no 7.0 hour rare earth alpha activity has been observed here following bombardments of Sm_2O_3 with 30 Mev alpha particles and Eu_2O_3 with 30 Mev protons. In nearly all bombardments here it is observed that some alpha activity due to traces of heavy element (uranium, thorium, bismuth, or lead) impurities in the target materials is produced. It has generally been necessary to make alpha energy measurements or chemical separations or both to

make sure that observed alpha activity was due to rare earth nuclides and not to heavy element contaminants. For example, the 7.5 hour alpha emitter At^{211} would be produced in good yield by 30 Mev alpha particles on an extremely small amount of bismuth impurity by the reaction $Bi^{209}(\alpha,2n)At^{211}$. This possibility should not be ignored. An alpha particle energy measurement on any newly discovered alpha emitter in the rare earth region needs to be made as proof in view of the probability of heavy element impurities in small amounts.

C. Terbium Isotopes (Z = 65)

In a preliminary report[2] the discovery of rare earth alpha emitters with alpha particle energies of 4.2 and 4.0 Mev and half-lives of ~7 minutes and ~4 hours, respectively, was reported and their tentative assignment to terbium made. Subsequent work provided chemical proof for the assignment of the latter activity to terbium (3.9_5 Mev, 4.1 hour). The seven minute activity, whose half-life is too short for chemical identification of the element, was reassigned to dysprosium (Z = 66) on the basis of later bombardment data. Its appearance in low yield after proton bombardments of gadolinium oxide in the first experiments was probably due to a small amount of terbium present in the target material. A second alpha emitter in terbium (3.44 Mev, 19 hour) was also observed and studied.

1. <u>3.95 Mev Tb^{149}</u>.-- This alpha emitter of 4.1 hour half-life was the first of the artificial rare earth alpha emitters to be observed,[2] following a bombardment of gadolinium oxide with 200 Mev protons.

Subsequently, the 4.1 hour alpha activity was also produced by bombardment of terbium oxide with 120 Mev protons, of dysprosium oxide and ytterbium oxide with 200 Mev protons, and of europium oxide with alpha particles of 90 Mev, 120 Mev, and (in ~3 percent of the 90 Mev yield) 60 Mev alpha particles. It was produced in very low yield in a bombardment of gadolinium oxide with 31 Mev protons.

Following a number of these bombardments chemical separations of the rare earth products were made by the standard cation exchange column elutions described in Appendix I.3. The 4.1 hour alpha activity was always observed exclusively in the terbium chemical fractions.

Fig. 6 shows alpha pulse analysis curves of the 4.1 hour terbium alpha activity and of a U^{234} - U^{235} standard, both being thin, uniform samples prepared by volatilization. From the positions of the leading edges of the U^{234} and the U^{235} peaks the energy scale can be determined. The leading edge of the Tb^{149} peak corresponds to an energy of 3.93 Mev. Other energy determinations were in close agreement with this. A weighted average of three determinations gives a best value of alpha particle energy of 3.95 ± 0.04 Mev.

In Fig. 7 is plotted the decay curve of the total alpha activity in the unseparated material produced in a 15 minute bombardment of gadolinium oxide with 75 Mev protons. The 4.1 hour activity is seen to be by far the predominant activity. From this decay curve, making a minor correction for the presence of the 19 hour alpha activity, the half-life of Tb^{149} was set at 4.1 ± 0.1 hours.

The mass number 149 was assigned to this activity on the basis of a mass spectrographic determination previously reported.[27]

No positron activity with 4.1 hour half-life could be observed in samples containing Tb^{149}, but it has not been so far possible from direct counting data to make a significant estimate on the branching ratio between alpha decay and electron capture, as all samples containing Tb^{149} also contained presumably some 5 hour Tb^{156}, which decays by electron capture.[28]

2. 3.4 Mev Terbium.-- High energy (100-200 Mev) proton bombardments of gadolinium oxide, terbium oxide, and dysprosium oxide, and alpha particle bombardments of europium oxide were observed to produce some alpha activity of 19 hour half-life.

Following proton bombardments of gadolinium oxide and of dysprosium oxide, the terbium activities were separated from the gadolinium target material by means of the standard cation exchange column elutions described in Appendix I.3. For the 87°C separations, pH 3.4 citrate solution was used as the eluting agent. This alpha activity was found exclusively in the terbium chemical fraction. An alpha pulse analysis curve of column-separated terbium alpha activities from a bombardment of dysprosium oxide with 200 Mev protons is shown in Figure 8. From the pulse analysis of a standard sample of U^{234} and U^{235} the energy spread was determined as 26.1 kev/register. The separation between the leading edges of peaks for the 19 hour terbium and 4.1 hour terbium alpha activities is 20.3 registers, corresponding to an energy difference of about 0.53 Mev. This difference was checked in several other measurements. Since the 4.1 hour activity has an alpha particle energy of 3.95 ± 0.04 Mev, the energy

of the 19 hour terbium is calculated to be 3.4_2 Mev. From an average of several measurements the energy was set at $3.4_4 \pm 0.1$ Mev.

In Figure 9 is plotted the decay curve of the 3.4 Mev peak, where the counting rates were determined by taking the total counts under the peak on the pulse analysis curves and subtracting a background. From this curve the half-life of the activity was determined as 19 ± 1 hours.

Only a tentative mass assignment of the 19 hour activity can be made at present. The observation was made from a series of three alpha particle bombardments of europium oxide at 60, 90, and 120 Mev that the yield of 19 hour activity relative to the 4.1 hour is greatest for the 60 Mev bombardment, in which the yield of the 4.1 hour activity is much lower than at 90 or 120 Mev. The 4.1 hour Tb^{149} can be produced by $Eu^{151}(\alpha,6n)Tb^{149}$ or $Eu^{153}(\alpha,8n)Tb^{149}$ reaction. In the 60 Mev bombardment the $Eu^{151}(\alpha,6n)Tb^{149}$ reaction was probably proceeding just-above its energetic threshold and was thus in low yield, while the $Eu^{151,153}(\alpha,5n)Tb^{150,152}$ and $Eu^{151,153}(\alpha,4n)Tb^{151,153}$ reactions would have shown considerably higher yields at 60 Mev than at 90 Mev or 120 Mev. From such considerations it would seem that the mass number of the 19 hour activity is probably greater than 149, the mass number of the 4.1 hour activity. No alpha activity ($\alpha/EC < 5 \times 10^{-6}$) was found by Wilkinson[28] for terbium isotopes which could be produced by bombardment of Eu_2O_3 by 38 Mev alpha particles, that is, with mass numbers 153 and greater. Hence, the 19 hour alpha emitter probably has the mass number 150, 151, or possibly 152. It should be possible by further excitation function work with alpha particles on europium to make an exact mass assignment.

Theoretical half-life energy relations for alpha decay lead to the conclusion that the predominant mode of decay of the 19 hour terbium alpha emitter must be orbital electron capture or positron emission. The presence of 17.2 hour Tb^{154}, however, which decays by electron capture and positron emission,[28] in all samples of the 19 hour terbium has hitherto made impossible any direct determination of the branching ratios between the expected modes of decay of the 19 hour terbium alpha emitter.

D. Dysprosium Isotopes (Z = 66)

The three dysprosium alpha emitters listed in Table I were observed following high energy proton bombardments of several rare earth elements with atomic number greater than 64. They were also produced by 100 Mev C^{12} ion bombardments of neodymium (Z = 60) oxide by $Nd(C^{12},xn)Dy$ reactions. The alpha energies and the half-lives of these isotopes were determined following bombardments of terbium (Z = 65) oxide with protons of about 100 Mev energy.

With regard to the possible mass assignments of the three dysprosium activities of 4.2_1, 4.0_6, and 3.6_1 Mev energies, it is to be noted that all three are produced in good yield by 100 Mev protons on terbium. The 4.0_6 and 3.6_1 Mev activities are apparently produced in extremely low yield by 70 Mev protons on terbium. The cross sections for production of all three dysprosium alpha emitters undergo large increases somewhere between 70 and 100 Mev. An approximate theoretical calculation, of the type that has been outlined by Fermi,[29] of the most probable number of neutrons to be evaporated from the compound nucleus Dy^{160}, ($Tb^{159} + H^1$), excited to a

maximum of energy by 70 Mev or 100 Mev protons is 6 - 7 or 9 - 10, respectively. The dysprosium alpha emitters then would most likely be products of $Tb^{159}(p,xn)Dy^{160-x}$ reactions with $7 \leq x \leq 11$, allowing the limits $153 \leq A \leq 149$ to be set on the mass numbers. It is hoped that further studies of bombardment yields of these alpha emitters in the proton energy range between 70 and 120 Mev, will enable one to set better limits on the individual mass numbers.

No information concerning other modes of decay, such as electron capture or positron emission, has been obtained yet for any of the three dysprosium alpha emitters.

1. 4.2 Mev Dysprosium.--Bombardments of terbium oxide with 100 Mev protons and bombardments of neodymium oxide with ~100 Mev C^{12} ions were observed to produce some alpha activity with a seven minute half-life.

A probable element assignment is made by noting the bombardments in which this activity is produced and those in which it is not. Its production by protons on terbium and by carbon ions on neodymium ($Z = 60$) restricts the atomic number of the seven minute activity to 66 or less. Also, its atomic number must be greater than 65, for it was not produced by bombardments of europium oxide with alpha particles at energies of 60, 90, and 120 Mev. Hence, the seven minute activity is attributed to a dysprosium nuclide ($Z = 60$).

Energy determination for the seven minute dysprosium activity has been made from the first and second of the alpha pulse analysis curves of activities from a bombardment of terbium oxide with 100 Mev protons for ten minutes, shown in Figure 10. The separation between the 19 minute and 7 minute peaks is seen to be 4.2 ± 0.6 registers. Calibration of the

pulse analyzer at these gain settings, using U^{235} and Gd^{148} standards, determines the energy scale as 35.4 kev per register. The energy difference between the peaks is, therefore, $4.2 \times 0.035_4 = 0.15 \pm 0.02$ Mev. The alpha particle energy of the 19 minute dysprosium activity was determined as 4.06 ± 0.04 Mev. The alpha particle energy of the seven minute activity is set then at $4.2_1 \pm 0.06$ Mev.

Figure 11 shows a plot of the decay of the two highest energy dysprosium alpha peaks. The counting rates were calculated from the summation of the counts under each pulse analysis peak. The half-life of 7 ± 2 minutes is determined from the slope of the decay curve. Other half-life determinations agree within these limits.

2. 4.0_6 Mev Dysprosium.-- Bombardments of terbium oxide with 100 Mev protons and of dysprosium oxide with 200 Mev protons were observed to produce alpha activity with a 19 minute half-life.

The assignment of this activity to dysprosium is made on the basis of its appearance in the 100 Mev proton bombardment of terbium oxide and not in alpha particle bombardments of europium oxide at energies of 60 Mev, 90 Mev, and 120 Mev. The alpha particle energy of the 19 minute dysprosium activity is determined from the plot in Figure 12 as $4.0_6 \pm 0.04$ Mev by pulse analysis, using a pulse generator comparison technique described in Appendix I.1.

Figure 11, referred to in the previous section, shows a plot of the decay of the 4.06 Mev alpha peak. From this plot the half-life of the activity is set at 19 minutes. Other half-life determinations check fairly well, suggesting the error limits $\pm$ 4 minutes.

3. 3.6 Mev Dysprosium.--Bombardments of terbium oxide with 100 Mev and 120 Mev protons and bombardments of neodymium oxide with ~100 Mev C^{12} ions have produced some alpha activity of 2.3 hour half-life.

The element assignment to dysprosium is made on the basis of a chemical separation of the products of a 120 Mev proton bombardment of terbium oxide. The separation was made by standard cation exchange column elution at 78° C using pH 3.3 citrate solution as eluting agent, as described in Appendix I.3.

The alpha particle energy has been determined best by the pulse generator technique described in Appendix I.1 and is shown on the plot in Figure 12. The alpha particle energy is determined to be $3.6_1 \pm 0.08$ Mev.

Figure 13 is a semilog plot of the decay through two half-life periods of the alpha counts under the 3.6_1 Mev peak on the pulse analyzer. The half-life is determined from the slope to be 2.3 ± 0.2 hours.

E. Isotopes of Other Rare Earth Elements

There is good evidence of short half-life alpha activities in rare earth elements with atomic number greater than 66, that of dysprosium, although no systematic study of them has yet been made.

A short bombardment of dysprosium oxide with 200 Mev protons produced, in addition to several activities in terbium and dysprosium which have been previously discussed (Sections IIIC and D), some alpha activity with about a four minute half-life and 4.2 ± 0.15 Mev energy. This activity probably arises from a holmium ($Z = 67$) isotope.

A short bombardment of samarium ($Z = 62$) oxide with 100 Mev C^{12} ions was observed to produce some alpha activity with about a 3 1/2 minute

half-life. No energy measurement of the alpha particles was made, so the possibility that this activity came from heavy element impurities in the target material cannot be ruled out, but it seems probable that this activity was due to a holmium (Z = 67) or erbium (Z = 68) nuclide.

It appears that no appreciable alpha branching is exhibited by neutron-deficient nuclides of atomic number 67 to 69 with half-lives of intermediate length (<1 hour). A two hour bombardment of erbium (Z = 68) oxide with 200 Mev protons was made, and chemical separation by cation exchange column was completed two hours after the end of the bombardment. No alpha activity was found in the fractions corresponding to elements with atomic number greater than 66 (dysprosium). (The 2.3 hour dysprosium alpha activity and the 4.1 hour terbium activity were prominent in their respective chemical fractions.) In a search for long-lived alpha activity a target of ytterbium oxide (Z = 70) was given an intensive irradiation by protons of 150 and 250 Mev for a total of about forty hours. After several days a chemical separation by standard cation exchange column elutions was made. No alpha activity in any of the rare earth elements was found except for the 3.16 Mev alpha activity in gadolinium due to Gd^{148}.

No alpha activities in nuclides with atomic numbers less than 63 (europium) have yet been observed in this investigation, excepting, of course, the natural alpha emitting isotope Sm^{147}. Only a few short survey bombardments have been made in this region.

The best chance of observing alpha activity in an artificially produced samarium isotope is probably in the 84 neutron isotope Sm^{146}, which is presumed beta stable[22] but is missing in nature.

It seems likely that some of the nuclides containing 84 or a few more neutrons of the elements of atomic number greater than 66 will exhibit appreciable decay by alpha particle emission. It is to be expected, however, that the increased energies available to the electron capture or positron decay process will result in very short half-lives for these isotopes. Those nuclides with half-lives greater than a few minutes in these higher elements are probably so many neutrons beyond the 84 neutron configuration favorable to alpha decay that they will show unobservably low alpha decay branching.

F. Gold Isotope* (A = 79)

Following bombardments of gold foils with 200 Mev protons or 190 Mev deuterons and of platinum foils with protons of as low energy as 100 Mev, an alpha activity of 4.3 minute half-life was observed.[2]

This activity was assigned to gold on the basis of a chemical separation procedure utilizing extraction of the chloride into ethyl acetate as described in Appendix I.3. The alpha activity in the gold fraction was counted within thirty seconds following chemical separation and no evidence of deviation from the simple 4.3 minute decay was found. From this evidence, it can be stated that the alpha particles are emitted either by the 4.3 minute gold nuclide or by its platinum daughter nuclide (from positron or

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* The alpha activity in gold was discovered by S. G. Thompson and A. Ghiorso, and some of the work reported in this section was done by them. The material is included here merely for completeness.

electron capture decay) with the half-life of the daughter less than 15 seconds, the latter possibility seeming unlikely but possible.

By alpha pulse analysis the energy of the 4.3 minute alpha particles was determined as 5.07 ± 0.1 Mev. Comparison standards Pu^{239} (5.14 Mev), Am^{241} (5.48 Mev), and Cm^{242} (6.08 Mev) were used in the determination.

The half-life of 4.3 ± 0.2 minutes has been determined from alpha decay curves extending through a decay factor of 100.

The limits set on the mass number of the gold alpha emitter are rather indefinite, based on evidence concerning bombardment energies necessary for its production. Tentative limits of $183 \leq A \leq 187$ have been set.

Chemically separated samples containing the gold alpha activity were counted with standard Geiger counters, with and without beryllium and lead absorbers. A component with approximately four minute half-life can be resolved from the decay curves. From comparison of counting rates with different combinations of absorbers it was possible to determine an approximate figure for the abundance of electromagnetic radiation with four minute half-life of about the energy of platinum K x-rays. The ratio of alpha disintegrations to four minute K x/rays was about 10^{-4}. If both radiations arise from the same isotope and if roughly one K x-ray per electron capture disintegration is emitted, then the α/EC branching ratio would be 10^{34} as reported in the 1949 paper.[2] This branching ratio should be regarded as very tentative, pending further work to check the assumptions involved.

G. Mercury Isotope* (Z = 80)

Following bombardments of gold foils with 190 Mev deuterons some alpha activity of 0.7 minute half-life was observed.

This activity was assigned to mercury on the basis of a chemical separation by volatilization (see Appendix I.3) from the gold foil.

The alpha particle energy of the 0.7 minute activity was determined to be 5.60 ± 0.1 Mev by alpha pulse analysis. Its half-life was determined to be 0.7 ± 0.2 minute from decay curves of the 5.6 Mev alpha peak from the pulse analyses.

No work to determine the threshold bombardment energy for production of this activity has been done beyond the original 190 Mev deuteron bombardments; hence, no significant limits on the mass number can be set at this time.

IV. DISCUSSION

A. Alpha Decay Rate

The alpha emitters in the rare earth region are of special interest in that they provide data on the rate of alpha decay in a region considerably removed from the heavy element alpha emitters.

Of the rare earth alpha emitters with known decay energies only the naturally occurring Sm^{147} has its partial half-life for alpha decay well determined. The partial alpha decay half-lives of Eu^{147}, Gd^{149}, and Gd^{148} are known with a factor of three uncertainty. For most of the other alpha emitters studied no estimate of α/EC branching ratio can now be made; for

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* The work reported in this section concerning the alpha activity in mercury was performed by S. G. Thompson and A. Ghiorso and is included here merely for completeness.

these, of course, it can be stated that the partial alpha half-lives are greater than the experimentally-observed half-lives. Conclusions from so few data must be very tentative. Yet it was felt worthwhile to present the results of extrapolation to the rare earths of several decay formulas used in the heavy element region.

For comparison alpha decay half-lives have been calculated by extrapolation of formulas in which parameters have been adjusted to give good agreement with the alpha decay rates of the even-even type alpha emitters among the heavy elements (excluding the polonium isotopes[30]). Four different formulas (the first three using WKB approximation expressions for the barrier penetrability factor and the fourth a similar expression calculated by the "method of steepest descents") have been employed, the chief point of difference between the formulas being the different expressions used for the effective nuclear radius for alpha particles.

The first three formulas used can all be expressed generally as a product

$$\gamma = \gamma_0 (E)^{1/2} P, \tag{4.1}$$

where γ is the alpha decay constant in second^{-1}, E is the alpha decay energy in Mev, P is the exponential barrier penetration factor, and γ_0 is the "decay constant without barrier at 1 Mev" in second^{-1}. The fourth formula derived by Preston[31] and used recently by Kaplan[32] has the same exponential factor P but has a coefficient showing a more complicated dependence on decay energy than the simple square root.

The first formula was derived, as Cohen[33] has done, using a nuclear radius expression determined from the comparison of experimental

alpha particle bombardment excitation functions[21,34,35,36] with the theoretical calculations of Weisskopf.[37] Then the parameter γ_0 was calculated separately for many even-even heavy element alpha emitters and the geometric mean 5.03×10^{18} seconds^{-1} taken (corresponding to a level width without barrier at 6 Mev of 8.1 kev in excellent agreement with Cohen's[33] correction to Bethe's many body alpha decay treatment[38]). The nuclear radius expression based on the work of Jungermann[36] on Th^{232} (alpha, fission) cross sections

$$r = (1.43\ A^{1/3} + 1.48) \times 10^{-13}\text{cm} \qquad (4.2)$$

(A = mass number of daughter nucleus) was used. The additive constant 1.48×10^{-13}cm for the alpha particle radius is the value used by Weisskopf.[37]

The second formula is the form of the one-body alpha decay expression used by Perlman and Ypsilantis[30] with $\gamma_0(E)^{1/2}$ given by v_0/r where v_0 is the velocity of the alpha particle (center of mass system) at infinite distance from the nucleus. This corresponds to $\gamma_0 = 7.7 \times 10^{20}$ seconds^{-1} for the rare earth studies. They found good agreement in their calculations using the sample radius expression

$$r = 1.48\ A^{1/3} \times 10^{-13}\ \text{cm}, \qquad (4.3)$$

and this radius formula is also used for the rare earth calculations.

The third formula is from the original many-body treatment of Bethe.[38] A γ_0 value of 6.24×10^{14} seconds^{-1} was used. The simple radius formula

$$r = 2.05\ A^{1/3} \times 10^{-13}\ \text{cm} \qquad (4.4)$$

originally given by Behe was used after checking it with decay data of Pu^{236} and finding it to give agreement.

The fourth formula is that derived by Preston[31] and also applied recently by Kaplan.[32] The values tabulated in Table III were calculated by taking, as Preston and Kaplan have done, the smallest solution for (E - U), the kinetic energy of the alpha particle within the nucleus and using nuclear radius values given by Present's[39] formula.

$$r = r_o^* A^{1/3}\left[1 + 0.8\ (A - 2Z)^2 A^{-2} - 0.3A^{-1/3} + 0.010\ Z^2\ A^{-4/3}\right] \qquad (4.5)$$

with $r_o^* = 1.50 \times 10^{-13}$ cm, the value found by Kaplan[32] by applying the Preston alpha decay formula to the even-even type alpha emitters in the heavy region.

The penetration factor P in all cases was calculated by the exponential expression (600) of Bethe.[38]

Table III lists for comparison the calculated alpha half-life values and the experimental values for the light alpha emitters. Column 4 contains an additive correction factor to the alpha decay energy necessitated by the consideration of the potential screening due to the electron cloud, as pointed out by Ambrosino and Piatier.[40] This is the first of two corrections of about the same magnitude proposed by them and has been estimated from the Fermi-Thomas atomic model and experimental binding energy data (cf. Appendix II) as

$$E_{sc} = 72.8\ (Z - 1)^{4/3}\ \text{kev.} \qquad (4.6)$$

It is felt that the inclusion of both corrections would involve a duplication, as is pointed out in Appendix II.

Three of the rare earth nuclides included in the comparison are not of the even-even type, so that, in general, the agreement with a decay rate formula for even-even alpha emitters would not be expected. In

Table III

Comparison of Experimental with Theoretical
Alpha Decay Half-Lives for Light Alpha Emitters

Nuclide	Alpha particle energy (Mev)	E_{sc} (Mev) (Electron Screening Cor.)	E_{α} nuclear energy (Mev)	Partial alpha half-life (Exp)	Est Error limit factor	Calc. half-life exp. radius	Ratio Calc.: exp.	Calc. half-life one body Perlman-Ypsilantis	Ratio Calc.: exp.	Calc. half-life many body Bethe	Ratio calc.: exp.	Calc. half-life one body Preston-Kaplan	Ratio calc.: exp.
Sm^{147}	2.18	0.02	2.26	1.4×10^{11}y	1.5	1.8×10^{12}y	7.9	8.2×10^{11}y	5.9	$2.8_2 \times 10^{13}$y	200	1.6×10^{13}y	114
Eu^{147}	2.88	0.02	2.98	$\sim 6 \times 10^{3}$y	3	$1.8_3 \times 10^{4}$y	3.0	$1.4_8 \times 10^{4}$y	2.5	4.3×10^{5}y	72	2.5×10^{5}y	42
Gd^{148}	3.16	0.02	3.27	$\sim 1.4 \times 10^{2}$y	3	$2.0_9 \times 10^{2}$y	1.5	$1.0_8 \times 10^{2}$y	0.77	4.7×10^{3}y	34	2.0×10^{3}y	14
Gd^{149}	3.0	0.02	3.1	$\sim 4 \times 10^{3}$y	3	$1.0_3 \times 10^{4}$y	2.6	3.5×10^{3}y	0.87				
$Gd^{(150)}$	2.7	0.02	2.8			8.8×10^{6}y		$4.0_8 \times 10^{6}$y					
Tb^{149}	3.95	0.02	4.07	>4.1h		9.2h		5.5h		7.0d		3.9d	
$Tb^{(151)}$	3.44	0.02	3.55	>19h		3.9y		1.95y					
$Dy^{(151)}$	4.21	0.02	4.34	> 8m		42m		57m					
$Dy^{(151)}$	4.06	0.02	4.19	>19m		5.2h		7.6h					
$Dy^{(151)}$	3.61	0.02	3.72	>2.3h		0.83y		1.15y					
$Au^{(188)}$	5.07	0.03	5.21	>4.3m (~30d)	?	17h		8.0h					
$Hg^{(189)}$	5.60	0.03	5.76	>0.7m		33m		3.6m					

( ) indicates value of mass number assumed for calculation where mass number is not known.

accordance with observation in the heavy region, however, the odd nucleon types would be expected to have half-lives longer than predicted by the even-even formula rather than shorter, as are actually observed.

Little can be concluded from the alpha emitters whose α/EC branching ratios are not known. All the extrapolations give alpha half-lives greater than the lower limits set by the experimentally observed half-lives. The only comparisons of any significance that can be made are those for Sm^{147}, Eu^{147}, Gd^{149}, and Gd^{148}. The first two treatments predict half-lives in fair agreement with experiment, while the third and fourth predict half-lives too long. Note, though, that the extrapolations by the third and fourth treatments appear in relatively bad light with respect to the first in that the radius formulas in the third and fourth do not have the additive constant for the alpha particle radius, and the fourth treatment suffers further in using Present's formula which gives an even smaller radius than the simple $aA^{1/3}$ formulas. The use of radius formulas with additive constants could provide an extra ten percent in radius needed to bring the predictions by the third and fourth treatments into as good agreement as those of the first or second. One can only conclude in this regard that by use of a decay formula with a frequency factor as high as those in the one body models, a radius formula without additive constant is adequate, but with formulas employing lower frequency factors, agreement is best secured by use of a radius formula with an additive constant.

None of the treatments give the correct energy dependence of the half-life. In all cases the Eu^{147} and Sm^{147} predictions are successively worse (i.e. predicted half-lives greater than experimental) than the Gd^{148}

and Gd^{149} predictions. These discrepancies may be due to experimental error in the value of 2.18 Mev for the alpha particle energy of Sm^{147}. (Sm^{147} was used as the standard in determination of Eu^{147} energy.) There is wide variation in energy values reported in the literature (cf. NBS Nuclear Tables[41]). So great is the sensitivity of the barrier penetration factor to a change in energy at the low energy of Sm^{147}, that an increase in energy of only 52 kev would increase the exponential penetration factor by a factor of six, enough to bring good agreement with experiment. For reference the calculated dependence of the penetration factor on energy is tabulated in Table IV and graphed for Sm^{147} in Figure 14.

Table IV

Rate of Change of Penetration Factor with Energy

Nuclide	Radius used (10^{-13} cm)	Alpha energy (Mev)	$\frac{-d \log_{10} P}{dE(\text{Mev})}$
Gd^{148}	8.99	3.27	8.6
Eu^{147}	8.97	2.98	9.7
Sm^{147}	10.78	2.26	14.7
Sm^{147}	8.97	2.26	15.0
Sm^{147}	7.76	2.26	14.1

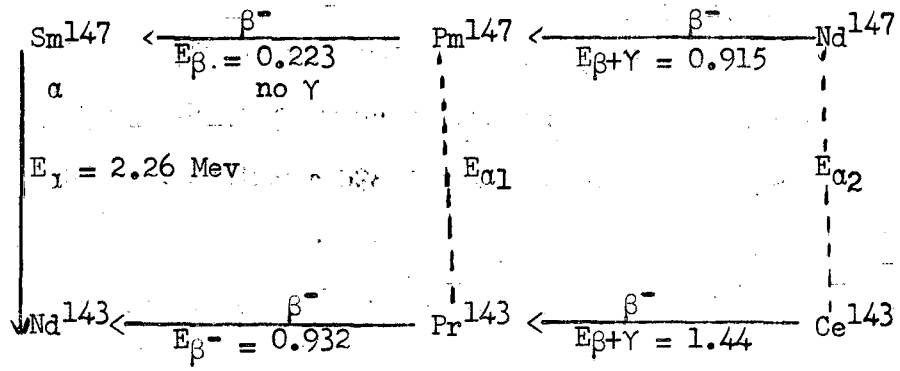
There is also the possibility, as Wheeler[42] suggested, that the excessively large decay rate of Sm^{147} is due to an effective nuclear radius increase due to a deviation of the Sm^{147} nucleus from spherical

symmetry, as would be shown by a large nuclear quadrupole moment, concerning which no measurements have yet been published.

B. Rare Earth Alpha Decay Energy Systematics and the Neutron Closed Shell of 82

With knowledge of the alpha decay energies of many isotopes of the heavy elements and the beta decay energies of a few it has been possible to calculate the relative masses[43] of nuclides of given radioactive families with great accuracy by the method of closed decay cycles. The determination of a few neutron binding energies made it possible to relate the mass values between the various decay families and thus to calculate the relative masses of a very large number of heavy nuclides.

Before studying the systematics of alpha decay energies in the rare earth region it would be well to attempt to calculate any unobserved alpha energies possible by the method of closed decay cycle energy balances.[9] The application of such calculations to the rare earth alpha emitters is unfortunately quite limited, since in the region where alpha decay occurs the principal decay process is orbital electron capture, for which it is not possible to make direct experimental determinations of the decay energy. It is possible, though, to calculate the alpha decay energies of Pm^{147} and Nd^{147} as follows: (The mass assignment of the samarium alpha activity to 147 has been made by Weaver[44] and by Rasmussen *et al.*,[27] and the alpha particle energy 2.18 Mev as determined in an argon-filled ionization chamber by Jesse and Sadauskis[16] is used with recoil energy and screening correction added to give the nuclear decay energy).



Conservation of energy requires that

$$E_{\alpha_1} = 2.26 + 0.223 - 0.932 = 1.56 \text{ Mev}$$

$$E_{\alpha_2} = E_{\alpha_1} + 0.915 - 1.44 = 1.04 \text{ Mev}$$

(Beta decay energies of Pm^{147} and Pr^{143} are from the NBS table.[41]) Nd^{147} decay energies are from the work of Emmerich and Kurbatov,[45] and Ce^{143} energies are from the work of Mandeville and Shapiro.[46] These alpha decay energies correspond to alpha half-lives much too long for experimental detection.

The list of rare earth nuclides with known alpha decay energies can be augmented for a study of decay energy trends by the addition of a few nuclides for which upper or lower limits on alpha decay energy can be set by consideration of the relation between alpha decay energy and rate. Approximate upper energy limits can be set for the naturally occurring rare earth nuclides not observed to undergo alpha decay. Included in Table V are Nd^{144}, Sm^{144}, and Sm^{148} for which upper energy limits have been set on the assumption that the half-lives for alpha decay must be greater than 10^{14} years to have escaped experimental detection of decay.[47] The lower energy limit in Table V for the presumably beta stable Sm^{146} not present in nature was set on the assumption that its alpha decay half-life must be less than 10^{8} years to have decayed away since the origin of the elements.

Table V also lists the alpha decay energies calculated from Fermi's semiempirical mass equation using the tables of Metropolis and Reitwiesner[19] and assuming a mass for He^4 of 4.00390 atomic mass units and the mass energy conversion factor of 931.4 Mev = 1 a.m.u.

In Figure 15 the energy data of Table V are plotted against neutron number.

The gadolinium isotopes constitute the only isotopic sequence for which the mass numbers are fairly certain. Mass assignments on the plot for the dysprosium isotopes and 3.44 Mev terbium are guesses based on the expectation that the various curves of Figure 15 should be nearly parallel as are those of Figure 16, a plot of alpha energies against mass number in the heavy region.

It is evident from Fig. 15 that a maximum in alpha decay energy occurs for the samarium isotopes at 83 or 84 neutrons. In Section III A evidence was presented to show that Eu^{145}, Eu^{148}, and Eu^{149} must have alpha half-lives much longer than that of Eu^{147}, since no alpha activity ascribable to them has been observed. In accordance with the relation between alpha decay energy and decay rate it can, thus, be assumed that Eu^{147} with 84 neutrons, for which alpha decay has actually been observed, has a greater alpha decay energy than any of its neighboring europium isotopes (with the possible exception of Eu^{146}, for which a significant upper limit on the alpha half-life has not yet been determined). For the gadolinium and terbium isotopes, also, the maximum in alpha decay energy probably is found for the 84 neutron nuclides. Furthermore, no alpha activity ascribable to a nuclide with less than 84 neutrons has yet been observed.

Table V

Alpha Decay Energies of Rare Earth Nuclides

Nuclide	N	E_α (Exp.) Mev Inc. Screening Correction	"Normal" E_α (Calc. from semi-emp. mass eqn.) Mev	Difference Mev
Sm^{147}	85	2.26	0.08	2.18
Eu^{147}	84	2.98	0.73	2.25
Gd^{148}	84	3.27	1.19	2.08
149	85	3.1	1.01	2.1
150	86	2.8	0.87	1.9
Tb^{149}	84	4.08	1.60	2.48
(151)	(86)	3.56	1.28	2.28
Dy(150)	(84)	4.35	2.05	2.30
(151)	(85)	4.20	1.87	2.33
(152)	(86)	3.73	1.73	2.00
Pm^{147}	86	1.56(calc)	-0.59	2.15
Nd^{147}	87	1.04(calc)	-1.28	2.32
Nd^{144}	84	<2.0	-0.68	<2.68
Sm^{144}	82	<2.1	+0.64	<1.5
Sm^{148}	86	<2.1	-0.07	<2.2
Sm^{146}	84	>2.4	+0.28	>2.1

( ) indicates mass number not known but merely assumed for calculations.

The normal trend for alpha decay energy to increase with decreasing neutron number is seen from the family of parallel lines in Fig. 15 defined by the alpha decay energies calculated from Fermi's semiempirical mass equation in which no account is taken of fluctuations in nuclear binding energies caused by quantum effects, such as those ascribed to nuclear closed shells. The true alpha decay energies are not only greater than mass equation values by about 2 Mev, but exhibit a discontinuity in the normal trend, dropping to lower alpha decay values for neutron numbers less than 84.

Although the neutron binding energies of these alpha active rare earth nuclides cannot be calculated explicitly as for the alpha emitters in the neighborhood of 126 neutrons,[20] the analogy between the alpha decay energy curves in the two regions, with their maxima at 84 and 128 neutrons, respectively, would strongly suggest that the maxima at 84 neutrons in the rare earth alpha decay energies are a consequence of the decreased neutron binding energies just beyond the closed shell[48] of 82 neutrons in analogy to the maxima at 128 neutrons resulting from the abnormally low neutron binding energies just beyond the closed shell of 126 neutrons.

The effect on alpha decay energies of the 82 neutron closed shell was early suspected after discovery of rare earth alpha activity. Before any element or mass assignments had been made this relation of the closed shell was discussed by Thompson _et al._[2] and by Perlman _et al._[49] and predictions of the probable mass numbers of alpha active nuclides made. Subsequent experimental work has borne out these predictions to a remarkable degree.

The neutron binding energy comparisons of Harvey[20] show clearly an abnormal drop in the neutron binding energies just beyond 126 neutrons, and the few cases compared near 82 neutrons seem to indicate a like discontinuity. The few experimentally determined neutron binding energies in Harvey's compilation near 82 neutrons can be used as a basis for calculation of the neutron binding energies of isobaric nuclei using beta decay energies for closed cycle energy balance calculations similar to the cycle calculations of alpha decay energies earlier in this section. Beta decay energies and decay schemes from the NBS nuclear tables[41] have been used, except for the positron decay energy of Pr^{140}, which was recently redetermined[50] spectrographically as 2.23 Mev. Table VI summarizes these calculations. The differences, Δn, between actual and mass equation calculated neutron binding energies is plotted against neutron number in Fig. 17, as Harvey[20] has done. A drop of 1.5 Mev in neutron binding energies in crossing the 82 neutron configuration is seen. This discontinuity in the neutron binding energy evidently extends into the region of higher atomic number, where it affects the alpha decay energies.

The decreased neutron binding energies just beyond 82 neutrons facilitate the observation of alpha emission in two respects. First, alpha decay energies are increased, and, second, the energies available to the electron capture or positron emission decay processes are decreased.

From small extrapolations of the curves in Fig. 15 one might estimate the decay energies of a few nuclides, the alpha activities of which have not yet been detected, such as, Sm^{146}, ~2.5 Mev, Sm^{148}, ~2.0 Mev, Pm^{145}, ~2.0 Mev. With such energies alpha activity of Sm^{146} should be barely detectable and that of Sm^{148} and Pm^{145} undetectable by present techniques.

Table VI

Neutron Binding Energies Near 82 Neutrons

(Binding energy of last neutron in nuclide.)

Nuclide	Z	N	Bn Exp. (Mev)	Bn Mass Eqn. (Mev)	Δn ($B_{exp} - B_{mass\ eqn}$) (Mev)
Nd^{141}	60	81	7.9	7.25	+0.7
Pr^{141}	59	82	9.5	8.40	+1.1
Ba^{139}	56	83	5.2	5.63	-0.4
Ce^{141}	58	83	5.7	6.26	-0.6
La^{141}	57	84	6.8	7.43	-0.6
Ba^{141}	56	85	~5.0	5.30	-0.3
Nd^{150}	60	90	7.4	7.16	+0.2
Nd^{147}	60	83-87 Ave. 4 neutrons to Nd^{147}			-0.62

Underlining denotes experimental value from compilation of Harvey.[20]

Since the magnitude of the discontinuity in neutron binding energies at 82 neutrons is about 1.5 Mev, the 83 and 82 neutron isotopes might be expected to have on the order of 1.5 and 3 Mev, respectively, less energy for alpha decay than the 84 neutron nuclide of the same Z. This energy difference easily accounts for the undetectably low alpha decay rate of Sm^{144} and indicates that the alpha decay of nuclides with less than 84 neutrons may only become observable in elements with atomic number greater than 65, that of terbium.

It is interesting to note that as the atomic number is increased at constant neutron number 84 the increase in alpha decay energy in going from atomic number 64 to 65 is much greater than that in going from 63 to 64. In Table V, too, it is seen that the energy difference column shows the Tb^{149} alpha energy to show the highest difference, some 0.3 Mev higher than the average of the lower elements. The observation might indicate that the 65th proton is less tightly bound than the general average in the region, that 64 might be a minor "magic number" proton configuration. A small splitting between the $2d_{5/2}$ and $2d_{3/2}$ levels on the Mayer[48] nuclear shell model could account for a slight extra stabilization of the 64 proton configuration.

Similarly, the observance of alpha activity in a gold and in a mercury nuclide might be an indication of a minor neutron magic number around 102 - 106 neutrons (from the mass number limits $183 \leq A \leq 187$ on the gold alpha emitter). The numbers 100 at the splitting between the $2f_{7/2}$ and $2f_{5/2}$ and 106 at the end of the $2f_{5/2}$ level can be derived from the Mayer shell model. Clearly, until there is much more information on alpha decay energies of light gold and mercury nuclides, the minor magic number possibility must remain only speculation.

V. DISCUSSION ON ALPHA DECAY RATE THEORY

Summary of Symbols Used

γ Alpha decay rate constant

e^{-2c} Exponential barrier penetration factor by WKB approximation

$$C = (1/\hbar)\int_R^{r_{tp}} \left[2M(V - E)\right]^{1/2} dr$$

r Center to center distance between alpha particle and recoil nucleus.

R Nuclear penetration radius, the effective nuclear radius for alpha decay

r_{tp} The value of r at the classical turning point. $E = V = \frac{2Ze^2}{r_{tp}}$

M Reduced mass of alpha particle.

V Potential energy of alpha particle, a function of r, $2Ze^2/r$ for $r > R$.

E Total nuclear disintegration energy. Alpha particle energy + recoil energy + change in total electron binding energies in alpha decay.

B Barrier height energy $B = 2Ze^2/R$, where e is the unit charge.

A Mass Number

$\omega = C = g\gamma(x)$

g In Bethe's[56] notation $P = e^{-2C} = e^{-2g\gamma(x)}$, where $g = (4MZe^2R)^{1/2}/\hbar$

$\gamma(x) = (\arccos x^{1/2})/x^{1/2} - (1 - x)^{1/2}$

$x = E/B$

$y = (1 - x)^{1/2}$

$\alpha = \arccos x^{1/2}$

v = Velocity of alpha particle

D = average nuclear level spacing between levels of zero total angular momentum and even parity.

D* an energy value defined by equation (5.32) and presumably of the same order of magnitude as D.

W Total nuclear energy eigenvalue.

S(W) The logarithmic derivative of the alpha particle wave function at the nuclear radius.

W_S The real part of W, the nuclear energy eigenvalue.

Γ The level width of the original nuclear state $\Gamma = \hbar\gamma$

k The wave number of the alpha particle at infinite distance

K The wave number of the alpha particle within the nucleus

a The nuclear penetration radius for alpha particles.

A. Alpha Decay Rate Correlations, General

In most treatments of alpha decay rates a rate expression of the form

$$\gamma = f\ P \tag{5.1}$$

has been applied, where γ is the alpha decay rate constant, f is a frequency factor or hypothetical decay constant in the absence of a potential barrier, and P is the penetration factor or probability of an alpha particle of the given energy tunneling through the negative energy region of the coulombic potential barrier. The form of the penetration factor is given by Bethe[51] as $P = e^{-2c}$ from the WKB[52] approximation calculation, where

$$C = \frac{1}{\hbar} \int_R^{r_{tp}} \sqrt{2M(V-E)}\ dr \tag{5.2}$$

The integration is carried out over the region where $V > E$. The potential V is generally assumed to be

(for S-wave emission)

$$V = \begin{cases} \dfrac{2\ Ze^2}{r} & \text{for } r > R \\ U, \text{ some constant value less than } E, & r < R. \end{cases} \tag{5.3}$$

and the integration is straightforward.

In the alpha decay expression, then, there are two unknown nuclear quantities. The first is f, the decay constant without barrier, and the second is R, the effective nuclear radius. For the moment the effect on penetrability of a third unknown, the internal potential U, which determines the kinetic energy of the alpha particle within the nucleus, will not be considered, as it does not enter into the simple WKB approximation Eq.(5.2) for the penetrability.

If a value for either f or R is assumed or determined independently, the other quantity can be calculated using the decay rate and energy data of the known alpha emitters. In the past a number of different estimates

of f have been made and corresponding calculations of R performed by various authors. These estimates of f range over a factor of 10^6, falling roughly into two groups--estimates from one body models[53] (10^{21} to 10^{19} sec^{-1}), and estimates from many body models[54] (10^{19} to 10^{15} sec^{-1}).

If the odd nucleon type alpha emitters are excluded, since many show abnormally low decay rates, the calculations from even-even alpha emitter data with almost any assumption of f between 10^{21} and 10^{15} sec^{-1} lead with but few exceptions to remarkably self-consistent sets of R values, generally of the right order of magnitude as inferred from other nuclear data giving a measure of nuclear size.* Although an alpha decay rate expression based upon an assumed f and calculated R is of much value in relating new alpha decay data (i.e., predicting half-lives from predicted energies, etc.) it would be desirable to have an expression in which f and R had values most nearly approaching those in the true physical situation. The best that can be done in this direction at present might be to reverse the older procedure of assuming f and calculating R, for recently excitation function data have been obtained which allow a determination of the effective nuclear radius for alpha particles by comparison with theoretical calculations by Weisskopf[37] of total cross sections for bombarding alpha particles. In particular the work of Jungermann[36] on the $Th^{232}(\alpha,fission)$ cross section

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* As Present[55] pointed out, though, nuclear radius values derived from different types of nuclear evidence are not strictly comparable. For instance, nuclear radii determined from beta decay energies of mirror nuclei are not strictly comparable with "penetration" radii calculated from charged particle reactions, and neither of these, with radii determined from total cross sections for fast neutrons.

gives by comparison with Weisskopf's calculations an effective nuclear radius of Th^{232} for alpha particles of 10.27×10^{-13} cm. This value is intermediate between the larger radius derived from the Bethe[38] many body treatment of alpha decay and the values derived in various one body treatments. While the experimental nuclear radius for alpha particles is determined mainly by the penetration of more energetic alpha particles than those in alpha decay, this value should be very close to the radius value effective in alpha decay.

Taking the total effective nuclear radius of Th^{232} for alpha particles to be 10.27×10^{-13} cm and the additive constant ("alpha particle radius" plus range of nuclear forces) to be 1.48×10^{-13} cm (as assumed by Weisskopf in his calculations)[37], the radius expression

$$R = (1.43\ A^{1/3} + 1.48)10^{-13}\text{cm} \tag{5.4}$$

is derived. The correlation is little affected by the value chosen for the additive constant, since $A^{1/3}$ varies only slightly over the region of the heavy element alpha emitters.

B. Correlation of Experimental Data with a Many Body Model Based on Jungermann's Value of the Alpha Penetration Radius of Th^{232}.

Using expression (5.4) for the effective nuclear radius, calculations of f for the known even-even alpha emitters have been made and are set forth in Table VII. The expression for alpha decay rate used was the simple

$$\gamma = fe^{-2c} = fe^{-2g\gamma(x)} \tag{5.5}$$

with g and $\gamma(x)$ defined as in Bethe.[56]

$$g = (4mZe^2R)^{1/2}/\hbar$$

$$\gamma(x) = x^{-1/2} \text{ arc cos } x^{1/2} - (1 - x)^{1/2} = \frac{\text{arc sin } y}{(1 - y^2)^{1/2}} - y \tag{5.6}$$

$$x = E/B$$

and $y = (1 - x)^{1/2}$

Table VII
Alpha Decay Rate Calculations for Ground State Transitions
of Even-Even Nuclides

Nuclide	Alpha Decay Energy Mev	Electron Screening Correction (Mev)	E Total (Mev)	$-\log_{10}P$ ($P = e^{-2G}$)	$-\log \gamma$ (Decay Const.) (γ in sec^{-1})	$\log_{10}f$ $= \log P - \log \gamma$ (f in sec^{-1})	$-\frac{1}{2}\log\left(\frac{E}{B-E}\right)$	$\log \gamma_0 = \log f - \frac{1}{2}\log\left(\frac{E}{B-E}\right)$ (γ_0 in sec^{-1})
Cm^{242}	6.21	0.032	6.24	25.78	7.41	18.37	0.25	18.62
Cm^{240}	6.37		6.40	25.17	6.57	18.60	0.24	18.84
Pu^{238}	5.59	0.031	5.62	28.26	9.62	18.64	0.28	18.92
Pu^{236}	5.85		5.88	26.83	8.09	18.74	0.26	19.00
U^{238}	4.25	0.030	4.28	36.20	17.31	18.89	0.34	19.23
U^{236}	4.57		4.606	33.64	15.03	18.61	0.32	18.93
U^{234}	4.84		4.87	31.81	13.03	18.78	0.31	19.09
U^{232}	5.40		5.43	28.40	9.50	18.90	0.28	19.18
U^{230}	5.96		5.99	25.42	6.41	19.01	0.26	19.27
Th^{232}	4.05	0.029	4.08	36.76	17.80	18.96	0.35	19.31
Th^{230}	4.76		4.79	31.37	12.66	18.71	0.31	19.02
Th^{228}	5.52		5.55	26.78	8.08	18.70	0.27	18.97
Th^{226}	6.41		6.44	22.48	3.43	19.05	0.23	19.28
Ra^{226}	4.877	0.028	4.905	29.60	10.89	18.71	0.30	19.01
Ra^{224}	5.784		5.812	24.50	5.66	18.84	0.25	19.09
Ra^{222}	6.63		6.66	20.78	1.74	19.04	0.21	19.25
Em^{222}	5.587	0.027	5.614	24.60	5.68	18.92	0.26	19.18
Em^{220}	6.398		6.425	20.92	1.90	19.02	0.22	19.24
Em^{218}	7.25		7.28	17.83	-1.44	19.27	0.18	19.45
Em^{212}	6.29		6.32	21.50	3.30	18.20	0.23	18.43

Table VII continued)

Nuclide	Alpha Decay Energy Mev	Electron Screening Correction (Mev)	E Total (Mev)	$-\log_{10}P$ ($P = e^{-2G}$)	$-\log \gamma$ (Decay Const.) (γ in sec^{-1})	$\log_{10}f$ $1 = \log P - \log \gamma$ (f in sec^{-1})	$-\frac{1}{2}\log\left(\frac{E}{B-E}\right)$	$\log \gamma_0 = \log f - \frac{1}{2}\log\left(\frac{E}{B-E}\right)$ (γ_0 in sec^{-1})
Po^{218}	6.110	0.026	6.136	21.37	2.42	18.95	0.23	19.18
Po^{216}	6.902		6.928	18.28	-0.64	18.92	0.19	19.11
Po^{214}	7.826		7.852	15.37	-3.68	19.05	0.15	19.20
Po^{212}	8.945		8.971	12.53	-6.36	18.89	0.11	19.00
Po^{210}	5.403		5.429	24.82	7.24	17.58	0.26	17.84
Po^{208}	5.24		5.27	25.75	8.14	17.61	0.27	17.88

The change of variable from x to y was made here to facilitate accurate calculation of a table of the function γ. This table is in Appendix III.

C. Discussion of Results of the Correlation in Section V.B.

The calculated values of f are remarkably constant except for nuclides with 126 or less neutrons. The average of log f for 23 nuclides with $N > 126$ is 18.84 ± 0.15. No significant trends of f vs. E or f vs. Z are evident other than a slight decrease of f with increasing Z. If f really should be constant for different Z, this trend would indicate that a slightly smaller choice of nuclear radius might be better.

Note that U^{238} and Th^{232} which were seen to decay too rapidly for their energies in the comparison of Perlman and Ypsilantis[30] (designated PY hereafter) are essentially in agreement with other nuclides here. The agreement is probably due mainly to inclusion of the screening correction.*

Note that hindrance (less than average decay rate) here is not associated with nuclides whose alpha decay <u>daughters</u> have proton or neutron numbers less than or equal to a closed shell " magic" number (82 for protons or 126 for neutrons). Instead, the hindrance is present when the nuclide undergoing alpha decay has itself the closed shell configuration or less. Thus, the polonium nuclides with $N > 126$ show no hindrance in alpha decay, although the daughter lead isotopes have the magic number 82 protons. Such evidence might suggest that the noticeable hindrance (factor of ten) in the nuclides with $N \leq 126$ is associated with a discontinuity in f at closed shell configurations rather than with a discontinuity in the nuclear radius.

The most remarkable aspect about the numerical results of this and other recent correlations[30,31,32] of even-even alpha emitter decay rate

*cf. Appendix II.

data is that the simple theory works so well to give a constant frequency factor or a smoothly varying nuclear radius, whichever is calculated as the undetermined quantity. For decay rates ranging over a factor of 10^{24} (Po^{212} to Th^{232}), the calculation of the penetration factor in an idealized model alone explains the variation. The agreement seems to some extent fortuitous when it is recalled what assumptions are involved in the calculation of P.

One inevitably assumes the nucleus to be a rigid charged sphere with infinitely short range attractive nuclear forces between alpha particle and nuclear matter. That is, one assumes the alpha particle at distances greater than the penetration radius to move in the pure electrostatic coulombic potential of a point charge of the magnitude of the nuclear charge and at distances less than the penetration radius to move in a constant internal potential. Now, the nuclear forces are certainly not of zero range, so the true potential would be a smooth continuous function of r near the radius, not the discontinuous idealized potential. Furthermore, the nucleus is not a rigid sphere but is constantly vibrating.

From the liquid drop model equations (8) and (25) of Bohr and Wheeler[57] one calculates that the magnitude of the zero point vibration in Th^{232} of the first possible mode ($n = 2$) of capillary oscillation corresponds to a maximum radius excursion of about 9% from the average value. Thus, one should treat a time-varying radius in alpha decay. The penetration radius derived for a static model would represent a radius nearer the maximum than the average radius in the zero point vibration. The treatment for the dynamic model would be complicated further by time-varying electric quadrupole and higher moments.

Refinement of the present theory by inclusion of the above corrections hardly seems justified at the present time, but it is important to bear in mind that the simple alpha decay models used for the derivation of the present-day decay expressions do neglect considerations which are certainly not negligible. These omissions evidently affect the decay rates of most known even-even alpha emitters to about the same extent, since the simple theory gives such excellent self-consistent correlations. Nevertheless, the fact that important considerations have been neglected constitutes a good argument against a too literal acceptance of the absolute values of frequency factors or penetration radii derived from alpha decay data.

With these reservations in mind, let us see what different nuclear frequencies might be derived of the same order of magnitude as the frequency factor f determined from averaging over the calculated values in Table VII. The average of log f was found to be 18.84 ± 0.15. That is, the geometric mean of f is $7 \cdot 10^{18}$ sec.$^{-1}$.

One body models of alpha decay (virtual level theory) yield frequency factors of magnitude $f = v_i/2R$[58] or $f = v_i/R$[59], where v_i is the velocity of the alpha particle inside the nucleus and R is the nuclear radius. The potential for an alpha particle within the nucleus is often assumed to be zero, giving for example, for Po^{214} with $R = 8.67 \cdot 10^{-13}$ cm. and $E_\alpha = 7.83$ Mev, corresponding to $v = 1.94 \cdot 10^9$ cm. sec^{-1}, a value of

$$f = \frac{1.94 \cdot 10^9}{8.67 \cdot 10^{-13}} = 2.2 \cdot 10^{21} \text{ sec}^{-1} \qquad (5.7)$$

This frequency factor is not constant for all the alpha emitters compared but varies directly as the square root of the decay energy and inversely as the nuclear radius.

Preston[31] and Kaplan,[32] by considering the alpha particle within the nucleus to be in its lowest square well level, 1S, derive a frequency factor nearly independent of decay energy, but inversely proportional to R^2. The energy of the alpha particle within the nucleus is taken[32] approximately to be

$E_i = h^2/8MR^2$, or about 0.51 Mev for heavy element alpha emitters.

Then for a nuclear radius of $R = 9.10^{-13}$ cm the frequency factor

$$f = v_i/2R \approx 2.7 \cdot 10^{20} \text{ sec}^{-1} \tag{5.8}$$

At the other extreme Bethe's many body model[51] assumes a frequency factor of

$$f \approx 10^{15} \text{ sec}^{-1} \tag{5.9}$$

by analogy with neutron emission by excited nuclei.

Cohen[33] suggests correcting Bethe's estimate to take into account the fact that level widths (emission rate constant times $\hbar$) are generally proportional to the average level spacing. Since level spacing in the region of the neutron resonances is on the order of 10 ev and the level spacing near the ground states of heavy nuclei is on the order of 100 kev, this correction multiplies Bethe's frequency factor by 10^4 giving

$$f \approx 10^{19} \text{ sec}^{-1} \tag{5.10}$$

Fluegge,[60] for reasons not given, estimates

$$f \approx 10^{17} \text{ sec}^{-1} \tag{5.11}$$

It is rather interesting that nuclear frequency factors of the same order of magnitude can be derived in a totally different manner. Bohr and Wheeler[61] estimate the lowest mode ($n = 2$) of capillary oscillation in the liquid drop model of the U^{238} nucleus to have a frequency factor equal to about $2 \cdot 10^{20}$ sec^{-1}. This frequency factor might have physical significance

to the alpha decay process, if one takes into account the fact that alpha emission is probably favored by the minimum barrier during the times of maximum excursions of the nuclear surface from the center during capillary oscillations.

Also, if one considers the finite compressibility of nuclear matter, one can envision a frequency factor determined by the zero point vibration of compression wave modes within the nuclear matter. An estimate of the frequency factor might be obtained by dividing the velocity of sound within nuclear matter, v_s, by the nuclear diameter.

$$f = v_s/2R \tag{5.12}$$

Rosenfeld[62] estimates

$$C^2 \approx 8.1 \cdot 10^{-3} \tag{5.13}$$

where $C = v_s/c$ and c is the velocity of light

Therefore $v_s \approx 3.10^{10}\ (.0081)^{1/2} = 2.7 \cdot 10^9$ cm sec^{-1}

hence, taking $R = 9.10^{-13}$ cm.,

$$f = 1.5 \cdot 10^{21}\ sec^{-1} \tag{5.14}$$

There is thus a large choice of frequency factors, ranging between 10^{21} sec^{-1} and 10^{15} sec^{-1}.

For a simple harmonic oscillator the relation

$$h\nu = D \tag{5.15}$$

holds, where ν is the frequency of the oscillator and D is the level spacing. By analogy a nuclear frequency factor for alpha emission might be

$$f = 2\nu = 2D/h = D/\pi\hbar \tag{5.16}$$

Taking Cohen's[33] estimate, D ≈ 100 kev, deduced from alpha fine structure separations, the frequency factor becomes

$$f = 4.8 \cdot 10^{19}\ sec^{-1} \tag{5.17}$$

Now certainly the nucleus is not a simple harmonic oscillator, but as Feshbach, Peaslee, and Weisskopf[63](designated FPW hereafter) point out, the nucleus at a given excitation may be considered an oscillator with much less confinement of the wave function than in the single particle model. Thus, the expression of Eq. (5.16) may be of real significance in the interpretation of the alpha decay process with a many body model. The remarkable connection between average level spacing and neutron emission probabilities, as discussed by FPW and Wigner,[64] leads one to feel that a similar connection probably holds in the alpha emission process.

D. Quantitative Formulation of a New Many Body Alpha Decay Formula

Since the FPW treatment of neutron emission probabilities yields expressions in such remarkable general agreement with experimental data, it would appear worthwhile to attempt an analogous treatment of the alpha decay process to obtain a decay rate expression with real physical significance.

The FPW treatment derives an approximate expression for the logarithmic derivative of the compound nucleus wave function with respect to coördinates of the emitted particle, the derivative being evaluated at the nuclear radius. This expression is equated to the logarithmic derivative of the external outgoing wave function of the emitted particle, also evaluated at the nuclear radius. The complex energy eigenvalue substitution gives two equations, one for the real and one for the imaginary parts. From the latter the partial level width for emission of the particle is derived.

In the derivation to follow there is no attempt to provide a rigorous justification of the procedure. The detailed justification can be found in FPW; here their arguments are repeated only where modifications have been introduced for the alpha decay treatment.

For the external wave function of the alpha particle in a pure coulombic field we make use of the "steepest descents" approximation of Preston[31] for the logarithmic derivative of the outgoing coulomb wave function.

1. The case of S-wave emission

a. The Internal Solution

We define S(W), the logarithmic derivative of the total nuclear wave function by the expression

$$S(W) = \left[\frac{1}{r\Psi}\frac{\partial(r\Psi)}{\partial r}\right]_{r=a} \qquad (5.18)$$

where W is the total energy of the system,

$\Psi(r_1 \ldots r_A)$ is the total wave function of the system,

$r_1 \ldots r_A$ are the coordinates of the nucleons,

r is the distance of the alpha cluster of two protons and two neutrons from the center of the residual nucleus and "a" is the nuclear radius. By considerations analogous to those in FPW[65] it can be shown that

$$S(W) = \left[\frac{1}{r\varphi}\frac{\partial(r\varphi)}{\partial r}\right]_{r=a} \qquad (5.19)$$

where $\varphi_{(r)}$ is the radial part of the alpha particle wave function. That expression (5.19) is valid follows from the fact that when the alpha cluster is formed and is at the nuclear radius with the proper energy for emission the total nuclear wave function is instantaneously just the product.

$$\left[\Psi_D(r_5 \ldots r_A)\ \varphi(r)\ \chi(r_1 - r, \ldots r_4 - r)\right]_{r=a} \qquad (5.20)$$

where Ψ_D is the total wave function of the daughter nuclide and χ the internal wave function of the alpha particle. Substitution of (5.20) into (5.18) gives (5.19). The probability of Ψ assuming the configuration (5.20) may be very small, but the probability divides out in evaluating S(W). Hence, this treatment automatically eliminates the need to estimate explicitly the "probability of formation" of the alpha particle.

Ψ is not a real stationary state, since the state decays in time by alpha emission. This transient behavior can be expressed by giving the energy eigenvalue an imaginary component.

$$W = W_S - i \cdot \frac{\Gamma}{2} \tag{5.21}$$

where W_S and Γ are real, Γ being the level width of the state and equal t o $\hbar\gamma$. Then, in the time dependence factor of $|\Psi|^2$

$$|\Psi|^2 = \left| e^{-i(W_s - \frac{i\Gamma}{2})\frac{t}{\hbar}} \right|^2 = e^{-\frac{\Gamma t}{\hbar}} = e^{-\gamma t} \tag{5.22}$$

We follow the same reasoning as FPW and write

$$S(W) = \frac{f(W)}{a} = -K \tan Z(W) \tag{5.23}$$

where K is the wave number of the alpha particle within the nucleus and Z(W) is some monotonically increasing function of W. Here there is a difference from the neutron resonance formulation of FPW. Namely, alpha emission resonance levels occur, for a nucleus with infinite outside potential, at real energies W_r, for which $Z(W_r) = (n + 1/2)\pi$, with n an integer, not at $Z = n\pi$ as for neutron resonances, for $S_i(W)$ must be infinite at the hypothetical alpha resonances and zero at the neutron resonances considered by FPW. Thus, we define y(W)

$$y(W) = Z(W) - \pi/2 \tag{5.24}$$

$$S_i(W) = -K \tan (\pi/2 + y(W)) \tag{5.25}$$

$$= +K \cot y(W)$$

and $y(Wr) = n\pi$

W, the energy eigenvalue of the nucleus, may be written

$$W = Wr + \Delta E - i\Gamma/2, \tag{5.26}$$

with Wr the resonance energy for the nucleus with infinite outside potential and $\Delta E - i\Gamma/2$ the energy shift caused by considering the actual

coulombic barrier effect. Both the real and imaginary components of this shift will be expected to be small compared to the energy spacing between resonance levels, so that we may estimate $S_i(W)$ as FPW do by making Taylor-Laurent expansion at the resonance level, W_r, of interest and taking only the first two terms. For the alpha case here $S_i(Wr)$ is a singular point and the expansion cannot be made. Therefore, it is necessary to match the reciprocals, $1/Si(W) = 1/So(W)$ at the nuclear radius. Wr is a regular point of the analytic function $1/Si(W)$ and can be expanded as

$$1/Si(W) = 1/Si(Wr + \Delta E - i\ /2) \approx 1/Si(W_r) + (\Delta E - i\Gamma/2)\ \frac{d}{dW}(1/Si(W))_{W = W_r} \qquad (5.27)$$

$$1/Si(W) = 1/K \cot y(W) = (1/K) \tan y(W) \qquad (5.28)$$

$$(d/dW)(1/Si(W)) = (1/K)\sec^2 y(W) \cdot dy/dW \qquad (5.29)$$

$$\text{Therefore } 1/Si(W) = \frac{1}{K} \tan y(W_r) + \frac{(\Delta E - \frac{i\Gamma}{2})}{K} \sec^2 y(W_r)(\frac{dy}{dW})_{W = W_r} \qquad (5.30)$$

but $y(W_r) = n\pi$

so $\tan y(W_r) = 0$

$$\sec^2 y(W_r) = 1 \qquad (5.31)$$

We define D* analogously to FPW by the expression

$$dy/dw = dz/dW = \pi/D^* \qquad (5.32)$$

It is to be expected that D* will be of the same order of magnitude as D, the level spacing, an expectation which is well borne out by experimental data on neutron resonances.

Then we have the final expression

$$[1/Si(W)]_{r=a} = (1/K)(\Delta E - i\Gamma/2)\left(\frac{\pi}{D^*}\right) \qquad (5.33)$$

b. The external solution.

For the external solution to be joined we need the coulomb wave function representing the outgoing wave.

The external radial wave function $X = r\varphi$ must satisfy the Schrödinger equation below:

$$\frac{d^2X}{dr^2} + \left[\frac{2mE}{\hbar^2} - \frac{2Ze^2 . 2m}{\hbar^2 r}\right] X = 0 \tag{5.34}$$

$$d^2X/dr^2 + (k^2 - \frac{2mB}{\hbar^2} . a/r) X = 0 \tag{5.35}$$

where k is the wave number at infinity,

B is the barrier height and a is the nuclear radius.

Making the substitutions $x = kr$

$$\beta = \frac{2mBa}{\hbar^2 k}, \quad \text{we have} \tag{5.36}$$

$$\frac{d^2X}{dx^2} + (1 - \beta/x)X = 0 \tag{5.37}$$

Preston[31] has obtained an approximate solution for the outgoing wave function by using the method of steepest descents to approximate the contour integral expression[66] for one of the solutions to the confluent hypergeometric equation

$$\frac{d^2y}{dx^2} + (1 - \beta/x) y = 0 \tag{5.38}$$

$$y^{(1)}(x) = e^{-\frac{\beta\pi}{2}} e^{ix} \int_0^\infty \left(1 - \frac{2ix}{t}\right)^{-\frac{1}{2}i\beta} e^{-t}\, dt \tag{5.39}$$

We use for the logarithmic derivative the expression[67] derived by Preston

$$S_0(W) = \left[\frac{1}{X(\omega)} \frac{dX(\omega)}{dr}\right]_{r=a} = -k \tan\alpha \left(\frac{1 - \frac{1}{2}i e^{-2\omega}}{1 + \frac{1}{2}i e^{-2\omega}}\right) \tag{5.40}$$

where k is the wave number at infinity

$k = \frac{1}{\hbar}(2ME)^{1/2}$

$\alpha = \arccos(E/B)^{1/2}$

$$\omega = \frac{\text{arc cos}(E/B)^{1/2}}{(E/B)^{1/2}} - (1 - E/B)^{1/2}*$$

$$\frac{1}{S_0(W)} = -\frac{1}{k}\cot\alpha\left(\frac{1+\frac{1}{2}ie^{-2\omega}}{1-\frac{1}{2}ie^{-2\omega}}\right) = -\frac{1}{k}\cot\alpha\left(\frac{1-\frac{1}{4}e^{-4\omega}}{1+\frac{1}{4}e^{-4\omega}} + i\frac{e^{-2\omega}}{1+\frac{1}{4}e^{-4\omega}}\right) \quad (5.41)$$

Neglecting $e^{-4\omega}$ with respect to 1, since it is of order 10^{-40} we obtain

$$\left[\frac{1}{(So(W)}\right]_{r=a} \approx -\frac{1}{k}\cot\alpha\,(1 + ie^{-2\omega}) \quad (5.42)$$

C. Final solution for ΔE and Γ.

Equating now the reciprocals of the logarithmic derivatives at the nuclear radius we have

$$\frac{1}{K}\left(\Delta E - i\frac{\Gamma}{2}\right)\frac{\pi}{D^*} = -\frac{1}{k}\cot\alpha\,(1 + ie^{-2\omega}) \quad (5.43)$$

Equating the real and imaginary parts we have

$$\Delta E = -(K/k)(D^*/\pi)\cot\alpha \quad (5.44)$$

and

$$\Gamma = \frac{K}{k}\,\frac{2D^*}{\pi}\,(\cot\alpha)e^{-2\omega} \quad (5.45)$$

$$\text{or } \gamma = \frac{K}{k}\,\frac{2D^*}{\pi\hbar}\,(\cot\alpha)\,e^{-2\omega} \quad (5.46)$$

If we let U be the potential energy of the alpha particle just inside the nuclear radius we can write $K = \left[2\,M(E-U)\right]^{1/2}/\hbar$

$k = (2ME)^{1/2}/\hbar$

$\cot\alpha = \text{c ot arc cos}(E/B)^{1/2} = \left[E/(B-E)\right]^{1/2}$

so $\gamma = \left[\frac{E-U}{E}\right]^{1/2} \frac{2D^*}{\pi\hbar}\left(\frac{E}{B-E}\right)^{1/2} e^{-2\omega}$

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* Note that ω is the same as Bethe's[68] $C = g\gamma(x)$ from the WKB approx., as used in Table VII calculations.

$$\gamma = \frac{2D^*}{\pi\hbar}\left(\frac{E-U}{B-E}\right)^{1/2} \qquad e^{-2\omega} \qquad (5.47)^*$$

Equation (5.44) for ΔE is of no particular interest. It merely indicates that the resonance energy is lower for the finite repulsive potential than for the infinite potential outside the nucleus. This is as one should expect, the wave function that is the less confined corresponding to the lower energy eigenvalue.

The formula for γ can be used if values for U and D* are estimated. There is no easy way to determine U. One might think of U as roughly the difference between the potential experienced by nucleons in the nucleus and the potential experienced by nucleons in an alpha particle. The former potential has been estimated from the nuclear model of a degenerate Fermi gas as approximately the sum of the Fermi energy and the nucleon binding energy or ~28 Mev. The potential of nucleons in an alpha particle might be of the same order of magnitude or a little less. Thus we would estimate U as zero potential (referred to zero potential at infinity), as Biswas and Patro and PY[59] have, in effect, done.

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* Equation (5.47) can be easily extended to include emission by even-even nuclei of alpha particles with angular momentum ($\ell > 0$). The derivation, not given here, is straightforward and makes use of Preston's[31] equation B.3 for the logarithmic derivative of the coulomb wave functions with $\ell > 0$. We have:

$$\gamma_\ell = \frac{2D^*}{\pi\hbar}\left[\frac{E-U}{B-E}\right]^{1/2} Q_\ell \, e^{-2\omega} \qquad (5.47a)$$

where Q_ℓ is a function of tan α and R. $Q_0 = 1$. Q_ℓ is always less than one and decreases with increasing ℓ. Preston gives explicit expressions for Q_1, Q_2, Q_3, and Q_4.

D* should be comparable to D, the average level spacing for the alpha emitting nucleus of levels capable of emitting S wave alpha particles to give the same residual nuclide as the alpha transition being considered; that is, D should be the level spacing between nuclear states of I = 0, even. On the case of ordinary alpha emission from unexcited nuclei D would be just the excitation energy of the first I = 0, even, excited state. Now, alpha emission from low lying excited states of nuclei is experimentally generally unobservable due to the rapid competing transitions by gamma emission. However, the spacing between alpha fine structure groups of the alpha decay parent of the alpha emitter under consideration should give information concerning D. In determining average level spacing from alpha fine structure one should admit only groups of S-wave alpha particles, those giving rise to alpha decay daughter states with zero total angular momentum ("spin") and even parity, like the ground states of the even-even nuclei. Unfortunately, little is now known of the spectroscopic assignments of the nuclear excited states following alpha emission in the even-even alpha emitters. Beling, Feld, and Halpern[69] by a study of the alpha-gamma angular correlation in decay of Th^{228} find evidence that the shorter range alpha particle emission gives rise to an intermediate state of total angular momentum 2. In fact, for any even-even fine structure group for which a single gamma emission to the ground state is observed it can be stated that the alpha group going to the excited state is not S wave, for the $0 \longrightarrow 0$ single gamma transition is strictly forbidden. Thus, we know that most of the excited states arising from alpha decay fine structure of even-even type alpha emitters do not have I = 0, even parity. Whether any of the excited states now known have I = 0, even, is an unanswered question, as yet.

The fact that alpha decay groups leading to spin zero levels are not observed allows a rough lower limit of 300 kev to be set on the energies of first excited spin zero, even parity states of even-even alpha emitters.

Let us apply the new decay formula (5.47) to the alpha decay calculations of Table VII, based on Jungermann's radius value for Th^{232}. In the last two columns of Table VII the energy dependent part in the coefficient of decay formula (5.47) is divided out of f. The last column should be proportional only to D*. The average of log γ_0 in the last column for the 23 alpha emitters with N > 126 is 19.10 ± 0.14, corresponding to a value for D* of 13.1 kev, where we define γ_0 by the equation

$$\gamma_0 = \frac{2D^*}{\pi\hbar} \tag{5.48}$$

The average D* of 13.1 kev is much smaller than the experimental lower limit of about 300 kev. In agreement with the experimental limit is the 400 kev value of D* corresponding to the higher frequency factor of Preston's one-body formula.

Thus, the new decay formula (a many-body treatment) favors the one-body decay expressions over the older many body expressions. The disagreement with the correlation here based on the Jungermann radius may mean the new decay formula is not applicable,* or more likely it may mean the effective nuclear radius for the low energy alpha particles of alpha decay is less by around 6% than the radius for the alpha particles of about the energy

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* The disagreement between D* and D may be fundamental, indicating that the function y(W) defined in equation (5.24), although necessarily a monotonically increasing function of W, increases much more rapidly in the energy region near the ground state than its average increase between the ground state and the first excited state with I = 0, even. Equation (5.47) is formally correct, but its usefulness rests on the knowledge or assumption of a value for D* or a relation between D and D*.

of the top of the potential barrier. The nuclear radius effective for these higher energy alpha particles is measured by the excitation function. The radius difference is certainly not unreasonable, for nuclear forces are not of infinitely short range as in the idealized square well potential assumed for the calculations.

It is not yet possible to judge which of the several alpha decay expressions in the literature most nearly represent the physical truth, but it is to be hoped that further information on spins and parities of excited states of alpha emitters will afford a real opportunity to determine what, if any, relationship there is between alpha decay rate and level spacings. Perhaps, through this new avenue of approach the true nature of alpha decay can more fully be understood.

Since the "probability of formation" of the alpha particle does not enter into the FPW type formulation, one might hope to extend this type of treatment to the odd nucleon number nuclides with their irregularly hindered alpha decay. Roughly, one might associate the hindrance in odd nucleon types with the greater density of levels, or smaller level spacing. A quantitative formulation will probably be quite involved, in general, for nuclides with $I \neq 0$. At present not enough spectroscopic state assignments of excited levels have been made to provide a test of such a theory. Its formulation will not be attempted here.

Other many body processes may also eventually find useful expression by FPW type treatments giving decay rates proportional to level spacings. In particular, the spontaneous fission process and alpha emission by excited nuclear states formed by thermal neutron capture might find better expression by such treatment than by the simple liquid drop or the simple one body formulations.

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APPENDIX I.

Methods of Investigation of Properties of Alpha Active Nuclides

1. Alpha Particle Energy--Alpha particle energies were obtained by measuring the amount of ionization produced in an ionization chamber filled with purified argon. Pulse height measurements were made by using the 48-channel differential pulse height analyzer described by Ghiorso *et al.*[15] The pulse height of the unknown alpha activity was measured, and alpha activity standards of known alpha energy were also measured for comparison. The unknown alpha energy was determined by linear interpolation or extrapolation from the standards. The linear dependence of total ionization in pure argon gas on alpha particle energy over a wide range of energy has been shown by Jesse, Forstat, and Sadauskis.[70]

Figure 12 illustrates the use of the pulse generator comparison technique for determination of alpha energies. The pulse heights of unknown and energy standard alpha emitters were matched with the output of a pulse generator and the output potentiometer setting on the pulse generator recorded for each. The height of pulses from the generator was known to be exactly proportional to the potentiometer setting. The plot of potentiometer readings against alpha particle energies was made using the values from the alpha energy standards, and a straight line through these points and the origin was drawn. The unknown alpha particle energies could be read opposite their potentiometer settings from the straight line plot.

For obtaining the best possible alpha energy measurements thin, uniform samples were often prepared by vaporization in vacuo of the alpha active material from a tungsten filament at white heat to a nearby platinum plate.

2. Half-life of Activity--Half-life measurements were usually made by the conventional method of plotting the logarithm of the counting rate against time, with the half-life being determined from the slope of the straight line plot, after subtraction of contributions from other activities and from general background. This method was used in this work for the determination of half-lives ranging from 4 minutes to 24 days. The method is of course, not applicable for the determination of extremely long half-lives. The best determinations of half-lives of alpha activities, where more than one activity was present in a sample, were made by plotting on the decay curve the integrated counting rates as determined from the total counts in a peak on the pulse analysis curves. Thus, with a complete or partial resolution of one activity from the others in the same sample, less uncertainty was introduced into the half-life determination by the subtraction of background activities.

In the work reported here it is not possible to rule out from experimental evidence alone the possibility that some of the activities are of the delayed alpha emitter type, examples of which are the familiar beta emitter Li^8 and the positron emitters B^8, N^{12}, and Na^{20}, discovered by Alvarez.[71] Theoretical considerations concerning alpha decay rates as a function of decay energy make it highly unlikely that any of the medium heavy element alpha emitters to be reported in this paper are of the delayed alpha emitter type. The observed half-lives in decay of these activities are thus presumed to be the true half-lives of the alpha active nuclides and not those of parent activities.

3. Atomic Number of Alpha Active Nuclide.--The determinations of atomic number were made by chemical means in the case of all activities

where chemical separations could be made in a length of time comparable to the half-lives. For activities with half-lives too short to permit chemical element assignments it was necessary to resort to the physical arguments already given concerning the appearance or nonappearance of the activity in bombardments of different target materials.

Element assignments by chemical means were accomplished by making chemical separations on the bombarded material and observing in which chemical fraction the activity of interest appeared. By making adequate chemical separations the activity could be assigned to a single chemical element.

It should be mentioned that such chemical assignments of atomic number do not necessarily determine the atomic number of the alpha active nuclide. The possibility always exists as mentioned in Appendix I.2 that the activity of interest is a short-lived activity in equilibrium with a longer-lived parent activity. In this case the activity of interest would appear in the chemical fraction of the longer-lived parent. Again it can only be said that theoretical alpha decay half-life considerations make it unlikely that such be the situation with any of the alpha emitters to be reported in this paper.

The rare earth elements (lanthanum through lutetium) present a special problem in chemical separation from one another, since they are very similar in chemical behavior.

The necessity of using thin samples for alpha counting and alpha energy measurement places a restriction on the amount of carrier material which may be added for the purpose of facilitating radiochemical separations. The cation exchange resin column separations to be mentioned below easily meet these requirements since no added carrier is required.

The separation of the individual rare earth elements in small amounts in the work reported here was usually made by elution from cation exchange resin columns. Dowex-50 resin[72] (spherical fines) in the ammonium form was used exclusively in the columns. The eluting solutions of citric acid-ammonium citrate were always 0.25 M in total citrate with 1 g/l. phenol added to prevent the growth of mold. Most of the separations were made in a jacketed column maintained at 78° C, 83°C, or 87° C. The apparatus and techniques employed in the column separations at elevated temperatures have been described in detail in a recent article.[14] Where speed in separation was unnecessary, the elutions were sometimes made at room temperature at a slower flow rate as described by Wilkinson and Hicks.[73]

Before several column separations small amounts of various rare earth elements were added, in order that positive chemical identification of elution peaks could later be made by spectrographic analysis.

Europium and gadolinium are not satisfactorily separated from one another by the cation exchange separations of the type described above. A sodium amalgam reduction procedure was employed as a means of effecting their separation. The zinc amalgam procedure described by Wilkinson and Hicks[73] for europium reduction was found to give extremely low yields unless europium carrier material was added in amounts too large for good alpha counting. However, the sodium amalgam procedure described here was found to give a good yield of carrier free europium activity. The reduction potential is sufficient to reduce samarium into the amalgam phase in low yield, and thus cannot be used effectively as the basis of a separation of europium from samarium. This procedure is essentially that used by Newton and Ballou for separation of samarium and europium fission products from other rare earths.[74]

Amalgam Procedure: Dissolve the europium oxide or hydroxide (not more than 10 mgm) in a minimum of hydrochloric acid. Dilute the solution to 15 ml with water, and add ten drops of glacial acetic acid. To the solution in a separatory funnel add 4 ml of 0.5 percent sodium amalgam. Shake for five seconds and transfer the amalgam layer to a second separatory funnel containing 30 ml of water. Again shake and transfer to a third funnel containing 30 ml of water. Finally, shake and transfer to a fourth funnel containing 20 ml 2 N HCl. After the evolution of hydrogen has ceased, the europium fraction can be recovered by hydroxide precipitation of the aqueous phase in the fourth funnel, while the gadolinium can be recovered from the aqueous phase in the first funnel.

The radioactive mercury fraction was separated from the bombarded gold leaf by volatilization. Within a special steel chamber the gold was heated strongly under a platinum plate, which was cooled on the back by circulating water. The mercury condensed on the platinum plate.

A gold fraction was isolated from the gold or platinum targets by dissolving these metal targets in hot, concentrated aqua regia, diluting the solutions to about 6 N in acid and extracting the gold chloride into ethyl acetate, which was then washed once or twice with 2 N HCl. The gold was recovered by evaporating the ethyl acetate solution on a platinum plate.

4. Mass Number--Determinations of mass number of the alpha active nuclides studied in this investigation were made by two methods: (a) Deduction from production yields of activity as a function of energy of bombardment (excitation functions), and (b) mass spectrographic isotope separation with detection of alpha activity by nuclear emulsion transfer

plate.[57] A third method of mass assignment, the deduction from observation of daughter activities of known mass number, did not facilitate any mass assignments of the alpha activities reported here.

For the alpha active isotopes within a few mass numbers of the naturally occurring isotopes the first method proved most convenient.

Alpha particle bombardment excitation functions were determined by 60-inch cyclotron bombardments using a "stacked foil" technique. A series of 0.00025 inch platinum envelopes, each containing 0.1 mgm of samarium oxide were bombarded, one behind the other in the special water cooled interceptor referred to in section II.A. The alpha particle energy bombarding each oxide sample was calculated from the initial energy and the thickness of absorbing material before the sample. The nuclear reaction by which a given activity is formed can often be determined by estimation of the energetic threshold. The Gd^{148} mass assignment was made in this manner (cf. Section III B1).

For isotopes many mass numbers away from beta stability, mass assignments cannot usually be made with certainty on the basis of excitation functions alone, although mass number limits can be set.

A mass spectrographic technique was used for the mass assignment of the Tb^{149} alpha activity. This technique is described in the report[27] by the author in collaboration with Reynolds and Thompson on the mass assignment of the Tb^{149} and Sm^{147} alpha activities. The mass spectrograhic technique as applied to Tb^{149} is limited to activities of at least several hours half-life that can be produced in relatively large amounts.

5. Other Modes of Decay.--Since the alpha active nuclides studied here all lie to the neutron-deficient side of beta stability, it seems

certain that all (with the exception of Gd^{150} and possibly Gd^{148}) will undergo decay by orbital electron capture, and some of the nuclides may exhibit positron emission also. These decay processes give rise to electron and electromagnetic radiation, referred to collectively as beta-gamma activity.

The beta-gamma radiations were studied by standard techniques using argon-filled or xenon-filled Geiger counters, a windowless methane-filled proportional counter, and a small beta ray spectrometer of low resolution. Counting through various absorbers was done to give information on energies and relative abundances of various components of beta-gamma radiations.

APPENDIX II

Correction to Alpha Decay Energy from Electron Screening

Recently Ambrosino and Piatier[40] have pointed out that it is not quite correct to use the observed alpha decay energy (alpha particle energy plus recoil energy) in the barrier penetration expressions relating alpha decay energies with decay rates. The presence of the electron cloud about the nuclei undergoing alpha decay necessitates the application of correction terms.

Firstly, they reason, one should use in decay expressions the energy release of the nuclear alpha emission process itself. The conservation of energy demands that the nuclear process must have a decay energy exceeding that observed in the laboratory (by alpha particle energy measurements or by calorimetric measurements) by at least the difference in total binding energy of the orbital electrons to the parent and to the daughter nuclei. This correction could be calculated by adding the term values for all the orbital electrons in the two nuclei and taking the difference, or it could be estimated from the expression for total electron binding energy calculated from the Fermi-Thomas[75] model ($W = 20.83\ Z^{7/3}$ ev[76]). The correction amounts to about 40 kev in the case of the uranium - thorium difference. (36 kev by the estimate of Ambrosino and Piatier.)

In addition to this correction, Ambrosino and Piatier point out that a further correction to the decay energy must be made by considering the amount by which the effective nuclear potential barrier for alpha particles is lowered by the presence of the cloud of negatively charged orbital electrons. For estimation of this effect they use the Fermi-Thomas model series approximation[77] to the electrostatic potential of a nucleus surrounded

by its electron cloud and consider the lowering of the potential at the nuclear radius to constitute a second correction to the alpha decay energy to be added to the first. The magnitude of this correction is estimated for the uranium - thorium difference to be 38 kev, the value of the second term in the series expansion of the potential energy of a doubly charged particle in the electrostatic field of a nuclide.

$$V(r) = 2e\frac{\gamma}{x}\left(1 + Bx + \frac{4}{3}x^{3/2} - \frac{2}{5}Bx^{5/2} + \ldots\right) \qquad (A2.1)$$

(energy units)

where $x = r/\mu$, $\mu = 3^{2/3}\, h^2/2^{13/3}\pi^{4/3}\, me^2 Z^{1/3}$,

$\gamma = 2^{13/3}\pi^{4/3}\, me^3 Z^{4/3}/3^{2/3}\, h^2$, and $B = -1.589$.

The terms after the second were found to be negligible at the nuclear radius distance. (The first term is the uncorrected pure coulombic nuclear potential. The second term $2eB\gamma = 97.4Z^{4/3}$ kev.)

While the author agrees that a correction term at least as large as the first-mentioned correction must certainly be made, it does not seem permissible to add to this the correction from the second term in the series expansion of Eqn. (A2.1). That the addition of both these corrections amounts virtually to correcting twice for the same effect has been recently demonstrated in unpublished work of Serber and Snyder.[78]

The duplication of the two corrections is also evident from Foldy's special application of a theorem due to Feynman.[80]

That it would be incorrect to make both corrections can be seen from the following arguments: the energy release of the nuclear process itself can be calculated by simple consideration of the conservation of energy as outlined for the first correction. Strictly speaking, the difference in total electron binding energies before and after alpha decay should

be added to the total energy released in the laboratory to obtain the energy release of the nuclear process. This total energy would comprise the alpha particle energy, recoil energy, and any excitation energy of the electron cloud about the residual nucleus.

The problem of ionization of atoms in alpha decay has been treated theoretically by Migdal.[81] From his results it seems unlikely that the average energy of electronic excitation following alpha decay will be great enough to affect significantly the alpha decay calculations.

Now, the question is asked: how much is the effective nuclear potential barrier lowered by the presence of the electron cloud? Clearly, any charge density lying outside the nucleus cannot affect the potential barrier effective for the escaping alpha particle except insofar as that external charge density might deviate from spherical symmetry and induce a polarization of the nuclear charge. Such a polarization induced by the electron cloud is certainly negligible in this problem. Any electron charge density lying within the nuclear volume would, on the other hand, lower the nuclear potential barrier by decreasing the effective nuclear charge. The magnitude of this latter effect can be estimated for mercury ($Z = 80$) using the relativistic expression of Racah[82] for the K electron density near the nucleus. The probability of a K electron being within the nucleus is about 10^{-8}. Considering all the orbital electrons the reduction in nuclear charge will be of the order of 10^{-7} electronic charge, which will lower the barrier for alpha particles by only 0.03 ev, a negligible amount.

The second term in equation (A2.1) thus does not represent an estimate of the barrier lowering due to electronic charge within the nucleus but is

a constant potential term independent of r, numerically fixed by the arbitrary assignment of zero for the electrostatic potential at infinite distance. The lowering of the potential barrier by electronic charge within the nucleus would be estimated by the Fermi-Thomas model from the series terms following the second, a negligible amount at the nuclear radius.

It is interesting, though, to note that it is no coincidence that the second term of equation (A2.1) is nearly equal to the first correction. This relation is most readily seen by considering the correction term in the two extreme cases of orbital electron interaction in the alpha decay process.

First, if the velocity of the alpha particle were much less than the average velocities of the orbital electrons, then presumably the electron wave functions would smoothly rearrange (adiabatic case) as the alpha particle was escaping, the electrons of the residual nuclide would be left unexcited, and the alpha particle and recoil would carry off the energy of the nuclear process less the difference in total electron binding energies as kinetic energy. Ambrosino and Piatier's first correction would be appropriate.

On the other hand, if the velocity of the alpha particle were much greater than the average velocities of the orbital electrons, it would be appropriate to use the sudden approximation, i.e., that the alpha particle completely escapes before any rearrangement of electronic wave functions occurs. In this case the final kinetic energy of the alpha particle and recoil nuclide would be that of the nuclear process less the difference in electron binding energies less some amount of electronic excitation energy of the recoil nuclide. The decay energy of the nuclear process could then be calculated from the observed alpha particle energy by noting that the alpha decay took place within the electron cloud of the parent nucleus in

a region at a potential 38 kev (for uranium) lower than that in the absence of the electron cloud. The alpha decay energy of the nuclear process must than have been just 38 kev greater than the energy of alpha particle plus recoil nuclide.

The work required to decrease the total binding energy of the electrons (as considered in the first correction term) is done by the alpha particle in moving from the center of the electron cloud to infinity against the attractive coulombic forces between electrons and alpha particle (as considered in the second correction term).

Neither the adiabatic nor the sudden approximation is entirely applicable to alpha decay, for the average velocities of the more tightly bound electrons in a heavy nucleus are greater than the velocity of emission of the alpha particle. Since alpha particle energies are generally determined by estimating the maximum energy of emission rather than the average, the electronic excitation energy should be of less concern. The correction to be added to the maximum alpha decay energy observed should be very nearly the first correction, the change in total electron binding energy in the alpha decay process.

In view of the above considerations, the first of the two corrections has alone been used for the calculations of alpha decay rate in this paper. The correction made was estimated from the total electron binding energy given by the Fermi-Thomas model but with the change that the coefficient 15.6 from experimental data[83] rather than the theoretical 20.83 value was used. The total binding energy is then $W = 15.6\ Z^{7/3}$ kev. The screening correction term used is twice the derivative of W with respect to Z evaluated at Z = 1.

$$Esc = 72.8\ (Z - 1)^{4/3}. \qquad \text{Eqn. (4.6)}$$

Since the time that the alpha decay rate calculations of Tables III and VII were made, using the energy correction of equation (4.6), there have come to the author's attention the calculations by Dickinson[84] of the potential at the nucleus due to orbital electrons. It will be shown below that the difference in total electron binding energies between a nucleus of charge Z and one of Z - 2 is very nearly equal to the potential energy due to orbital electrons of a doubly charged particle at the center of the electron cloud.

Foldy's[79] equation (2) states

$$E_B(Z) - E_B(Z - 1) = \int_{Z-1}^{Z} eV(Z)dZ + I_Z,$$

where I_Z is the first ionization potential of the atom of atomic number Z. V(Z) is the potential due to the electrons, considered as a smooth function of Z.

E_B is the total electron binding energy. Z is considered a continuous variable.

From the nonrelativistic Hartree calculations of Dickinson, Foldy finds the relation

$$eV = (12/5)Z^{7/5}\ R \qquad \text{(A 2.2)}$$

where R is the Rydberg constant, corresponding to an energy of 13.6 ev. Making an approximate relativistic correction in the manner suggested by Dickinson,[84] the expression

$$eV = (12/5)Z^{7/5}(1 + 0.006\ Z^2/29^2)R \qquad \text{(A 2.3)}$$

is obtained.

To derive the desired quantity

$$E_B(Z) - E_B(Z - 2)$$

write from Foldy's equation (2) $\Delta E = E_B(Z) - E_B(Z-2) - I_Z - I_{Z-1}$

$$= \int_{Z-2}^{Z} eV(z)\, dz \qquad \text{(A 2.4)}$$

then $\int_0^Z eV(z)dz$ and $\int_0^{Z-2} eV(z)dz$ are expanded in a Taylor series about $Z - 1$ and subtracted to give

$$\Delta E = 2\left[\frac{d}{dZ}\int_0^Z eV\,dz\right]_{Z=Z-1} + \frac{2}{3!}\left[\frac{d^3}{dZ^3}\int_0^Z eV\,dz\right]_{Z=Z-1} + \dots \qquad \text{(A 2.5)}$$

but differentiation with respect to the upper limit of the definite integral gives

$$\frac{d}{dZ}\int_0^Z eV\,dz = eV(Z)$$

$$\text{and} \quad \frac{d^3}{dZ^3}\int_0^Z eV\,dz = \frac{d^2}{dZ^2}\, eV(Z) \qquad \text{(A 2.6)}$$

Substituting (A 2.6) into (A 2.5)

$$\frac{1}{2}\Delta E \approx eV(Z-1) + \frac{1}{6}\left[\frac{d^2}{dz^2}\, eV(z)\right]_{z=Z-1} \qquad \text{(A 2.7)}$$

The second term of (A 2.7) is evaluated using (A 2.4)

$$\frac{d^2}{dz^2}\, eV(z) = \frac{12}{5}\left[\frac{7}{5}\cdot\frac{2}{5}\, z^{-\frac{3}{5}} + 7.15\cdot 10^{-6}\,\frac{17}{5}\cdot\frac{12}{5}\, z^{\frac{7}{5}}\right] R \qquad \text{(A 2.8)}$$

$$\left[\frac{d^2}{dz^2}\, eV(z)\right]_{Z-1} = 18\,(Z-1)^{-\frac{3}{5}} + 0.0019\,(Z-1)^{\frac{7}{5}}$$

(ev)

Finally, combining (A 2.4), (A 2.7) and (A 2.8), we write (A 2.9)

$$E_B(Z) - E_B(Z-2) \approx 2eV(Z-1) + I_Z + I_{Z-1} + 36(Z-1)^{-\frac{3}{5}} + 0.0038(Z-1)^{7/5},$$

I_Z and I_{Z-1} are of the order of 5 ev each. The last two terms in (A 2.9) for uranium with $Z = 92$ are equal to 2.4 ev and 2.1 ev, respectively. Then it is proved that for the alpha screening correction the terms after the first are negligible, being about 15 ev as compared to the first term of 38 kev (for uranium).

Figure 18 is a log-log plot which enables one to read the screening energy correction to be made to the alpha decay energy of a nuclide of atomic number Z. The nonrelativistic curve plots energy values just twice those in Dickinson's[84] Table II. The upper curve makes the approximate relativistic correction in the manner suggested by Dickinson. The upper curve is the best present estimate for the screening correction. These values of the correction are somewhat higher than the values from equation (4.9) used for the calculations of Tables III and VII, but the difference is less than the usual experimental error in determination of alpha particle energies.

Appendix III.

Table of $\gamma(y)$ for Alpha Decay Rate Calculations

An exponential factor appears in the alpha particle penetration factor expressions of Bethe[56] (by the WKB method) and Preston[31] (derived by a method of steepest descents). This exponential factor is, in the notation of Bethe,[56]

$$P = e^{-2g\,\gamma(x)} \qquad \text{(A 3.1)}$$

where

$$g = (4MZe^2R)^{1/2}/ \quad \text{, independent of decay energy,} \qquad \text{(A 3.2)}$$

$$\gamma(x) = \frac{\text{arc cos } x^{1/2}}{x^{1/2}} - (1 - x)^{1/2} \qquad \text{(A 3.3)}$$

$$x = E/B \qquad \text{(A 3.4)}$$

E = Total nuclear alpha decay energy

B = The "potential barrier height" = $\frac{2Ze^2}{R}$

M = Reduced mass of alpha particle

R = Effective nuclear penetration radius

e = The electrostatic unit charge

Bethe[56] presents a graph of the function $\gamma(x)$ vs. x. For precise alpha decay calculations it is desirable to have a table of this function giving γ to greater accuracy than it can be read from the graph. Hence, the following table was prepared.

A change of variable has been made to facilitate calculation. If the substitution

$$x = 1 - y^2 \qquad \text{(A 3.5)}$$

is made in (A 3.3), the following expression is obtained:

$$\gamma(y) = \frac{\text{arc sin } y}{(1 - y^2)^{1/2}} - y \qquad \text{(A 3.6)}$$

For the compilation of the table values of arc sin y were taken from the Table of arc sin x of the New York WPA[85] and values of $(1 - y^2)^{1/2}$ were taken (as $P_1^1(x)$) from the Tables of Legendre Associated Functions of Mursi.[86]

$\gamma(y)$ is given to five decimal places for the range in y of 0.750 (.001) 0.946, and values of the first difference are also tabulated.

TABLE OF $\gamma(y)$

$$y = \sqrt{1 - x}$$

where $x = E/B$

y	$\gamma(y)$	δ_1	y	$\gamma(y)$	δ_1
0.750	0.53215	0.00350	0.790	0.69556	0.00480
1	0.53565	352	1	0.70036	485
2	0.53917	355	2	0.70521	488
3	0.54272	357	3	0.71009	493
4	0.54629	361	4	0.71502	497
5	0.54990	363	5	0.71999	501
6	0.55353	366	6	0.72500	506
7	0.55719	368	7	0.73006	510
8	0.56087	372	8	0.73516	514
9	0.56459	374	9	0.74030	519
0.760	0.56833	377	0.800	0.74549	524
1	0.57210	380	1	0.75073	528
2	0.57590	383	2	0.75601	533
3	0.57973	386	3	0.76134	537
4	0.58359	389	4	0.76671	543
5	0.58748	392	5	0.77214	547
6	0.59140	396	6	0.77761	552
7	0.59536	398	7	0.78313	557
8	0.59934	401	8	0.78870	563
9	0.60335	405	9	0.79433	567
0.770	0.60740	408	0.810	0.80000	572
1	0.61148	411	1	0.80572	578
2	0.61559	414	2	0.81150	584
3	0.61973	418	3	0.81734	588
4	0.62391	421	4	0.82322	594
5	0.62812	424	5	0.82916	600
6	0.63236	428	6	0.83516	605
7	0.63664	431	7	0.84121	611
8	0.64095	435	8	0.84732	617
9	0.64530	438	9	0.85349	623
0.780	0.64968	442	0.820	0.85972	629
1	0.65410	446	1	0.86601	635
2	0.65856	449	2	0.87236	640
3	0.66305	453	3	0.87876	647
4	0.66758	457	4	0.88523	654
5	0.67215	460	5	0.89177	660
6	0.67675	465	6	0.89837	666
7	0.68140	468	7	0.90503	673
8	0.68608	472	8	0.91176	679
9	0.69080	476	9	0.91855	687

y	Y(y)	δ_1	y	Y(y)	δ_1
0.830	0.92542		0.870	1.27014	
1	0.93235	0.00693	1	1.28098	0.01084
2	0.93935	700	2	1.29196	098
3	0.94642	707	3	1.30308	112
4	0.95357	715	4	1.31435	127
5	0.96079	722	5	1.32575	140
6	0.96808	729	6	1.33731	156
7	0.97545	737	7	1.34902	171
8	0.98289	744	8	1.36089	187
9	0.99042	753	9	1.37291	202
0.840	0.99802	760	0.880	1.38510	219
1	1.00570	768	1	1.39745	235
2	1.01346	776	2	1.40997	252
3	1.02131	785	3	1.42266	269
4	1.02924	793	4	1.43553	287
5	1.03725	801	5	1.44858	305
6	1.04535	810	6	1.46181	323
7	1.05354	819	7	1.47523	342
8	1.06182	828	8	1.48885	362
9	1.07020	838	9	1.50266	381
0.850	1.07866	846	0.890	1.51667	401
1	1.08722	856	1	1.53088	421
2	1.09587	865	2	1.54531	443
3	1.10463	876	3	1.55995	464
4	1.11348	885	4	1.57482	487
5	1.12243	895	5	1.58990	508
6	1.13148	905	6	1.60522	532
7	1.14064	916	7	1.62078	556
8	1.14991	927	8	1.63658	580
9	1.15929	938	9	1.65262	604
0.860	1.16877	948	0.900	1.66893	631
1	1.17837	959	1	1.68549	656
2	1.18808	971	2	1.70232	683
3	1.19791	983	3	1.71943	711
4	1.20785	994	4	1.73682	739
5	1.21792	0.01007	5	1.75449	767
6	1.22811	019	6	1.77247	798
7	1.23842	031	7	1.79075	828
8	1.24887	045	8	1.80934	859
9	1.25944	057	9	1.82826	892
0.870	1.27014	070	0.910	1.84750	924
		0.01084			959

y	γ(y)	δ_1
0.910	1.84750	
1	1.86709	0.01959
2	1.88703	994
3	1.90732	0.02029
4	1.92799	067
5	1.94903	104
6	1.97048	145
7	1.99233	185
8	2.01459	226
9	2.03728	269
0.920	2.06042	314
1	2.08401	359
2	2.10808	407
3	2.13263	455
4	2.15768	505
5	2.18326	558
6	2.20937	611
7	2.23604	667
8	2.26328	724
9	2.29112	784
0.930	2.31958	846
1	2.34867	911
2	2.37843	976
3	2.40887	0.03044
4	2.44003	116
5	2.47193	190
6	2.50459	266
7	2.53806	347
8	2.57236	430
9	2.60753	517
0.940	2.64359	606
1	2.68060	701
2	2.71859	799
3	2.75760	901
4	2.79768	0.04008
5	2.83888	120
6	2.88125	237
		359

REFERENCES

1. G. Hevesy and M. Pahl, Nature 130, 846 (1932).

2. S. G. Thompson, A. Ghiorso, J. O. Rasmussen, and G. T. Seaborg, Phys. Rev. 76, 1406 (1949).

3. G. Gamow, Proc. Roy. Soc. London A126, 632 (1930), Constitution of Atomic Nuclei and Radioactivity (Oxford University Press, London, 1931), 15-21, Zeits. f. Physik 89, 592 (1934), Structure of Atomic Nuclei and Nuclear Transformations (Oxford University Press, London, 1937), 7, 38-45, 48-52; W. Heisenberg, Zeits. f. Physik. 77, 1 (1932); A. Lande, Phys. Rev. 43, 620, 624 (1933); E. D. Eastman, Phys. Rev. 46, 1, 238 (1934); H. A. Bethe and R. F. Bacher, Revs. Modern Phys. 8, 82 (1936), section 8, 30; C. F. v. Weizsacker, Die Atomkerne (Akademische Verlagsgesellschaft, Leipzig, 1937), 34-35, 101-102; A. J. Dempster, Phys. Rev. 53, 869 (1938); Hahn, Flugge, and Mattauch, Physik. Zeits. 41, 1 (1940).

4. G. Gamow, Zeits. f. Physik 52, 510 (1929).

5. E. U. Condon and R. W. Gurney, Phys. Rev. 33, 127 (1929).

6. T. P. Kohman, Phys. Rev. 76, 448 (1949).

7. N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939).

8. E. Feenberg, Revs. Modern Phys. 19, 239 (1947).

9. I. Perlman, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 77, 26 (1950).

10. G. T. Seaborg, B. B. Cunningham, H. H. Hopkins, M. Lindner, D. Miller, P. R. O'Conner, I. Perlman, and S. G. Thompson, Phys. Rev. 72, 739 (1947).

11. D. H. Templeton, Atomic Energy Commission Declassified Document AECD-1525 (1947).

12. Electromagnetically enriched stable isotope. On loan from Oak Ridge Isotope Research and Development Division.

13. J. O. Miller, J. G. Hamilton, T. M. Putnam, H. R. Haymond, and G. B. Rossi, Phys. Rev. 80, 486 (1950).

14. S. G. Thompson, A. Ghiorso, and G. T. Seaborg, Phys. Rev. 80, 782 (1950).

15. A. Ghiorso, A. H. Jaffey, H. P. Robinson, and B. B . Weissbourd, National Nuclear Energy Series, Plutonium Project Record, Volume 14B, "The Transuranium Elements: Research Papers, "Paper No. 16.8 (McGraw-Hill Co., Inc., New York, 1949).

16. W. P. Jesse and J. Sadauskis, Phys. Rev. 78, 1 (1950).

17. R. W. Hoff, J. O. Rasmussen, and S. G. Thompson, Phys. Rev. 83, 1068 (1951).

18. Manufactured by Radiation Counter Laboratories, Chicago, Illinois.

19. N. Metropolis and G. Reitwiesner, Table of Atomic Masses, NP-1980 (March, 1950) USAEC Document.

20. J. A. Harvey, Phys. Rev. 81, 353 (1951).

21. E. L. Kelly and E. Segrè, Phys. Rev. 75, 999 (1949).

22. T. P. Kohman, Phys. Rev. 73, 20 (1948).

23. D. C. Dunlavey, Unpublished results.

24. D. J. Rotblat, Nature 165, 387 (1950).

25. R. E. Hein and A. F. Voigt, Phys. Rev. 79, 783 (1950).

26. K. H. Sun, F. A. Pecjak, B. Jennings, A. J. Allen, and J. F. Nechaj, Bulletin Am. Phys. Soc. 26, 16, No. 2 (March 8, 1951).

27. J. O. Rasmussen, F. L. Reynolds, S. G. Thompson, and A. Ghiorso, Phys. Rev. 80, 475 (1950).

28. G. Wilkinson and H. G. Hicks, Phys. Rev. 79, 815 (1950).

29. E. Fermi, Nuclear Physics (The University of Chicago Press, 1950), p. 162.

30. I. Perlman and T. J. Ypsilantis, Phys. Rev. 79, 30 (1950).

31. M. A. Preston, Phys. Rev. 71, 865 (1947).

32. I. Kaplan, Phys. Rev. 81, 962 (1951).

33. B. L. Cohen, Phys. Rev. 80, 105 (1950).

34. G. M. Temmer, Phys. Rev. 76, 424 (1949).

35. S. N. Ghoshal, Phys. Rev. 80, 939 (1950).

36. J. Jungermann, Phys. Rev. 79, 640 (1950).

37. V. F. Weisskopf, Metallurgical Laboratory Report MDDC-1175 (1947), Lecture Series in Nuclear Physics, p. 105.

38. H. A. Bethe, Revs. Modern Phys. 9, 161 (1937).

39. R. Present, Phys. Rev. 60, 28 (1947).

40. G. Ambrosino and H. Piatier, Comptes Rendus Acad. Sci. Paris 232, 400 (1951).

41. Nuclear Data, U. S. National Bureau of Standards Circular 499 (1950).

42. J. A. Wheeler. Talk before Physics Journal Club, University of California, Berkeley, Fall, 1950.

43. M. O. Stern, Revs. Modern Phys. 21, 316 (1949). A. H. Wapstra, Physica 16, 33 (1950).

44. B. Weaver, Phys. Rev. 80, 301 (1950).

45. W. S. Emmerich and J. D. Kurbatov, Phys. Rev. 83, 40 (1951).

46. C. E. Mandeville and E. Shapiro, Proc. Nat. Inst. Sci. India 17, 45 (1951).

47. F. Bestenreiner and E. Broda. Nature 164, 658 (1949); E. Broda and K. Jenkner, Experientia 7, 121-127 (1951).

48. M. G. Mayer, Phys. Rev. 75, 1969 (1949).

49. Ref. 9, p. 38.

50. C. I. Browne, J. O. Rasmussen, J. P. Surls, and D. F. Martin, University of California Radiation Laboratory Report, UCRL 1469, (1951).

51. Ref. 38, p. 163, Eq. (591).

52. Wentzel, Zeits F. Physik 38, 518 (1926).
Kramers, Zeits. F. Physik 39, 828 (1926).
Brillouin, Comptes rendus 183, 24 (1926).

53. Ref. 38, p. 161, Ref. 30, Ref. 31.
S. Biswas, Ind. J. Phys. 23, 51 (1949).

54. Ref. 38, p. 163, Ref. 33.
S. Fluegge, Introduction to Nuclear Physics, Interscience Publishers, New York: 1946, p. 80.

55. Ref. 39, p. 30.

56. Ref. 38, p. 166, Eqns. (600), (600a), (600b).

57. N. Bohr and J. A. Wheeler, Phys. Rev. 56, 431, 434, (1939).

58. M. A. Preston, Phys. Rev. 83, 475 (1951).

59. Ref. 30, p. 32. S. Biswas and A. P. Patro, Ind. Journ. Phys. 22, 540 (1948).

60. S. Fluegge, Introduction to Nuclear Physics, Interscience Publishers, New York: 1946, p. 80.

61. Ref. 57, p. 435.

62. L. Rosenfeld, Nuclear Forces II, Interscience Publishers, New York: 1949, p. 250, Eqn (39).

63. H. Feshbach, D. C. Peaslee, and V. F. Weisskopf, Phys. Rev. 71, 150 (1947).

64. E. P. Wigner, Am. J. Phys. 17 , 104 (1949).

65. Ref. 63, p. 148.

66. Whittaker and Watson, Modern Analysis, Cambridge: 1927, pp. 339, 343.

67. Ref. 31, p. 870.

68. Ref. 38, p. 163, Eqn. (589).

69. J. K. Beling, B. T. Feld, and I. Halpern, Phys. Rev. 84, 155 (1951).

70. W. P. Jesse, H. Forstat, and J. Sadauskis, Phys. Rev. 77, 782 (1950).

71. L. Alvarez, Phys. Rev. 80, 519 (1950).

72. Dow Chemical Co., Midland, Michigan.

73. G. Wilkinson and H. G. Hicks, Phys. Rev. 75, 1370 (1949).

74. A. S. Newton and N. Ballou, Chemical Procedure 62-1, University of California Radiation Laboratory Report UCRL-432 (1949).

75. E. Fermi, Zeits, f. Physik 48, 73 (1928); L. H. Thomas, Proc. Camb. Phil. Soc. 23, 542 (1927).

76. E. U. Condon and G. H. Shortley, The Theory of Atomic Spectra (Cambridge University Press, Cambridge, (1935), p. 339.

77. E. B. Baker, Phys. Rev. 36, 630 (1930). V. Bush and S. H. Caldwell, Phys. Rev. 38, 1898 (1931).

78. R. Serber and H. S. Snyder. Private communication.

79. L. L. Foldy, Phys. Rev. 83, 397 (1951).

80. R. P. Feynman, Phys. Rev. 56, 340 (1939).

81. A. Migdal, Journal Exp. and Theo. Phys. (USSR) 11, 207 (1941). J. Phys. (USSR) 4, 449 (1941).

82. G. Racah, Nature 129, 723 (1932).

83. L. A. Young, Phys. Rev. 34, 1226 (1929).

84. W. C. Dickinson, Phys. Rev. 80, 563 (1950).

85. New York W.P.A. Table of Arc Sin X. Columbia University Press. New York: 1945.

86. Mursi, Z., Tables of Legendre Associated Functions. Fouad I. University (Abbasia, Cairo), Faculty of Science, No. 4.

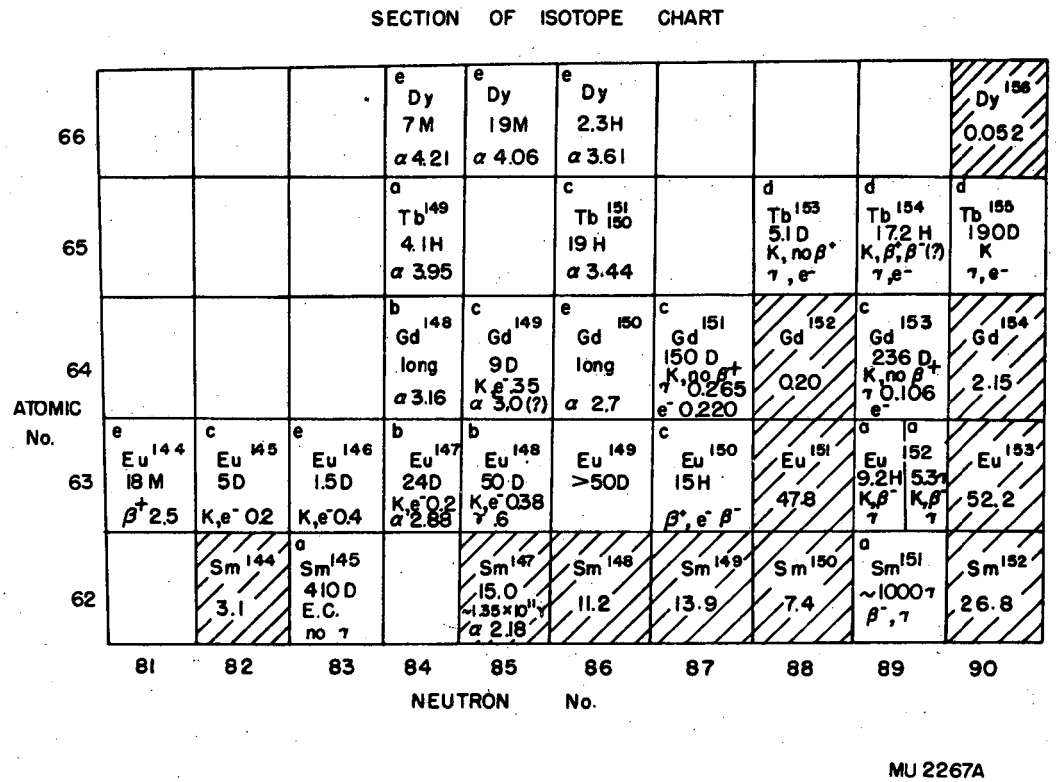

Fig. 1

Section of isotope chart showing the rare earth alpha emitters

Mass number of dysprosium alpha emitters are not known

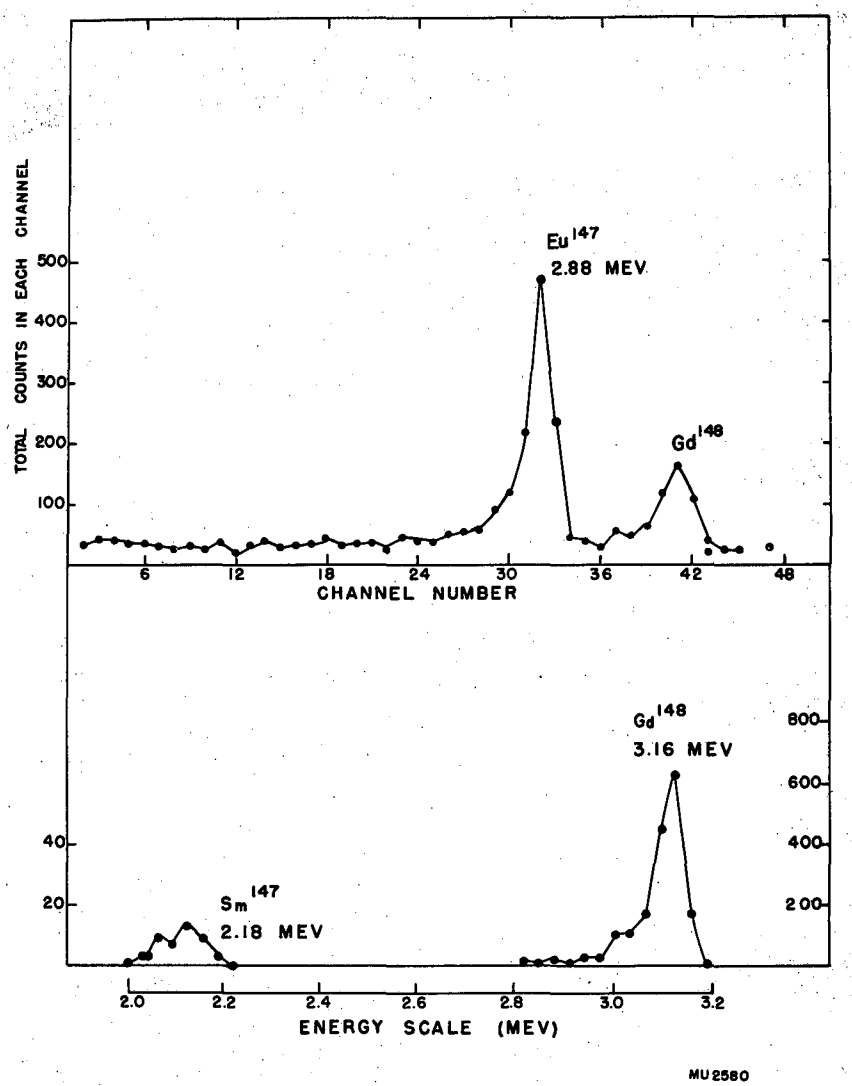

Fig. 2

Alpha pulse analysis curve of Eu^{147} and comparison curves of Sm^{147} and Gd^{148} standards

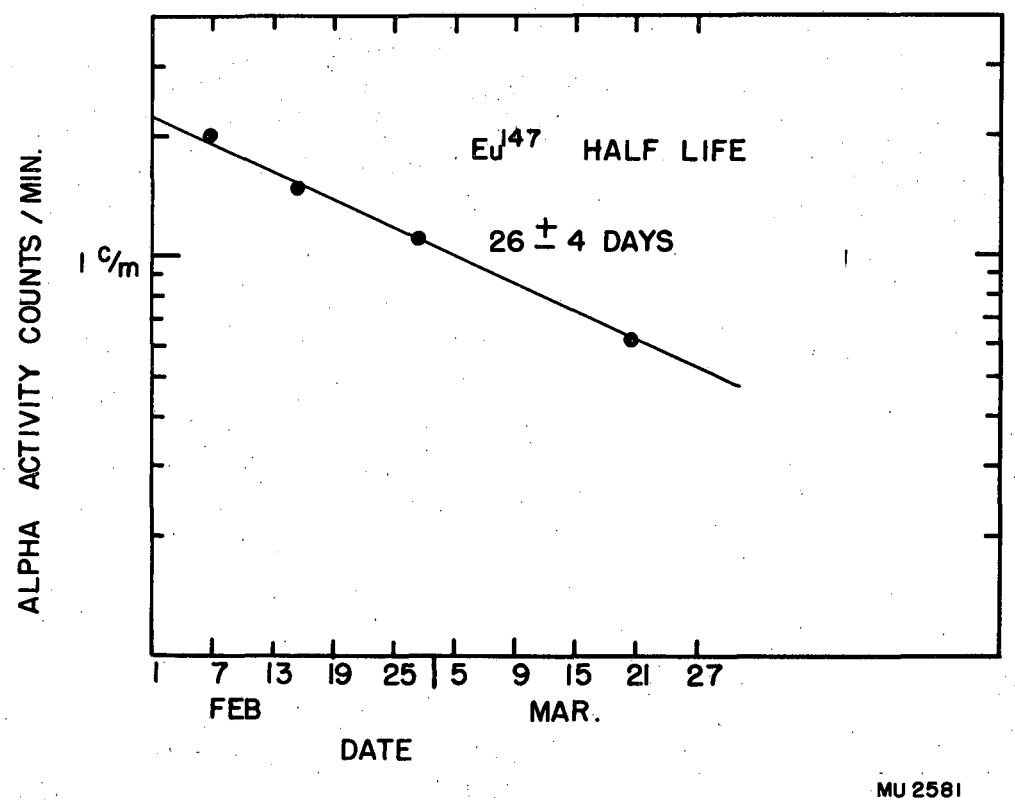

Fig. 3

Decay curve of Eu^{147} alpha activity from pulse analysis counting data. An estimated background correction was made before plotting.

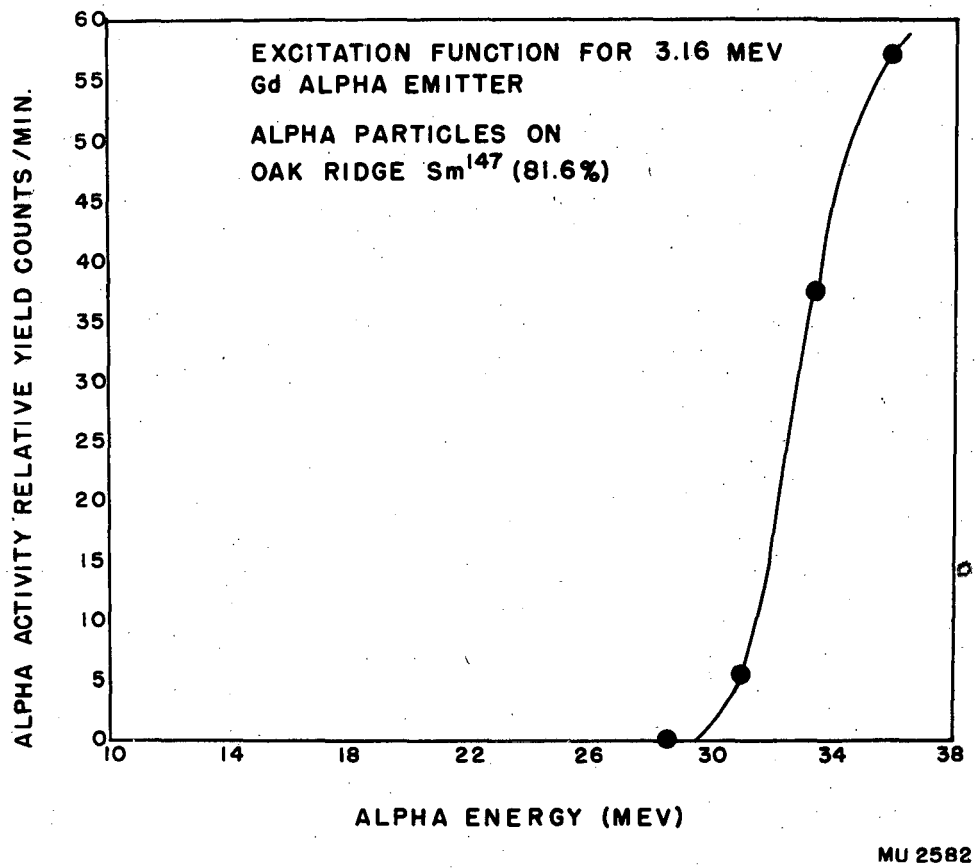

Fig. 4

Yield of 3.16 Mev Gadolinium alpha activity versus energy of alpha particles bombarding Sm^{147}

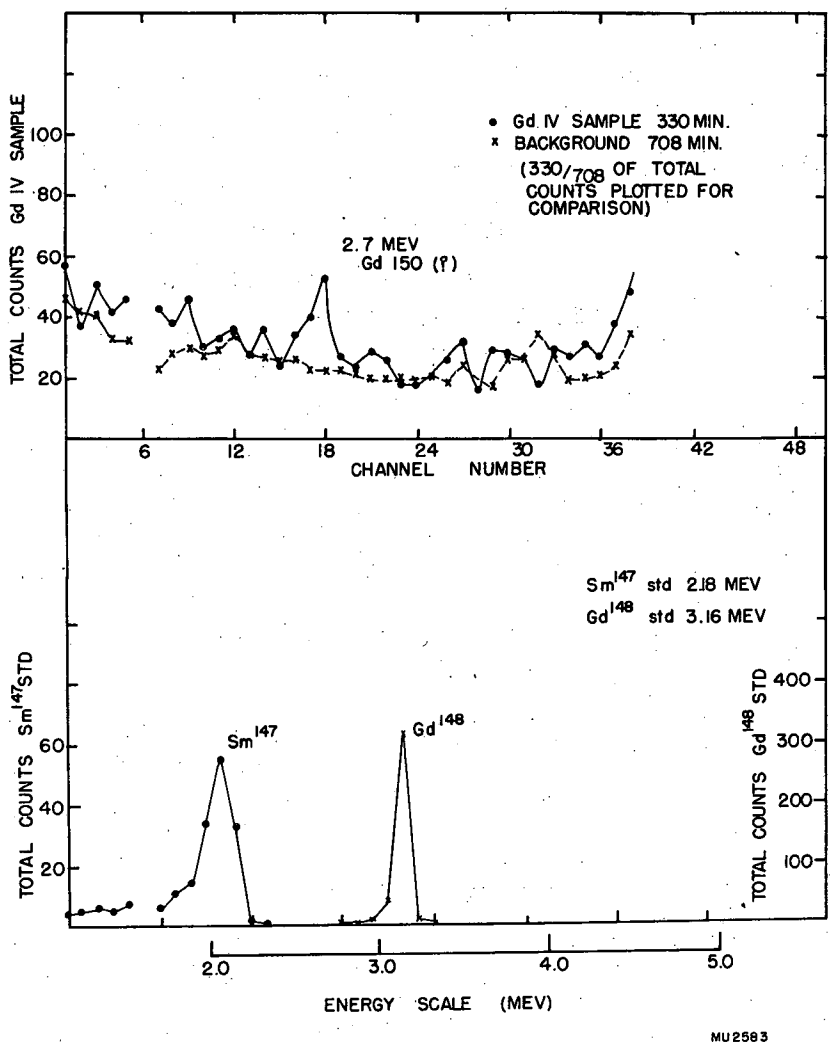

Fig. 5

Alpha pulse analysis curve of a gadolinium sample from 19 Mev deuteron bombardment of Europium oxide. Background count is plotted on the same scale. Pulse analysis curves for the energy standards Sm^{147} and Gd^{148} below.

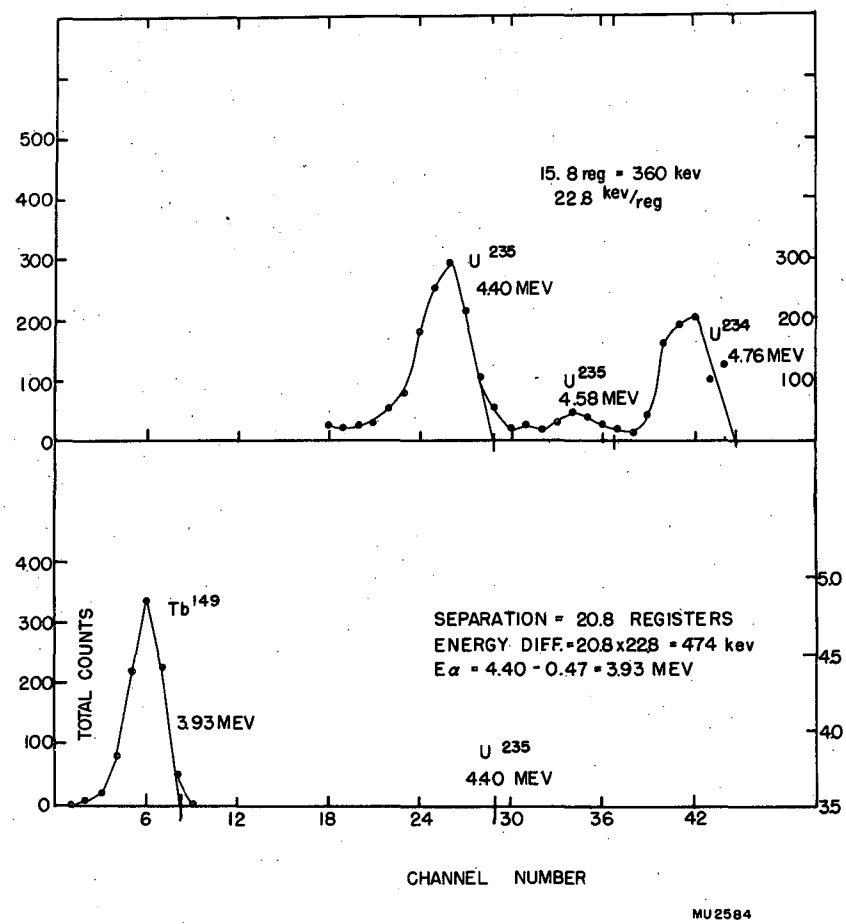

Fig. 6

Pulse analysis curves of Tb^{149} with comparison energy standard U^{234} - U^{235}

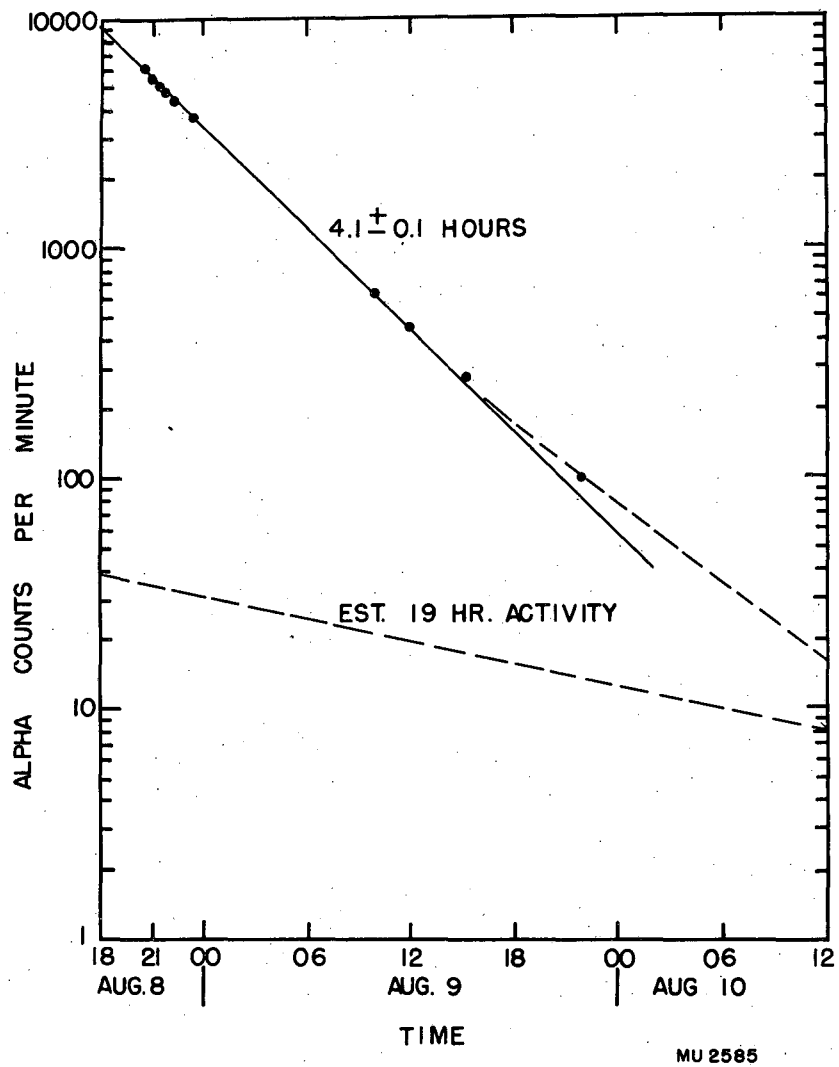

Fig. 7

Decay curve of gross terbium alpha activity in ion chamber alpha counter

(Predominant activity is Tb^{149}.)

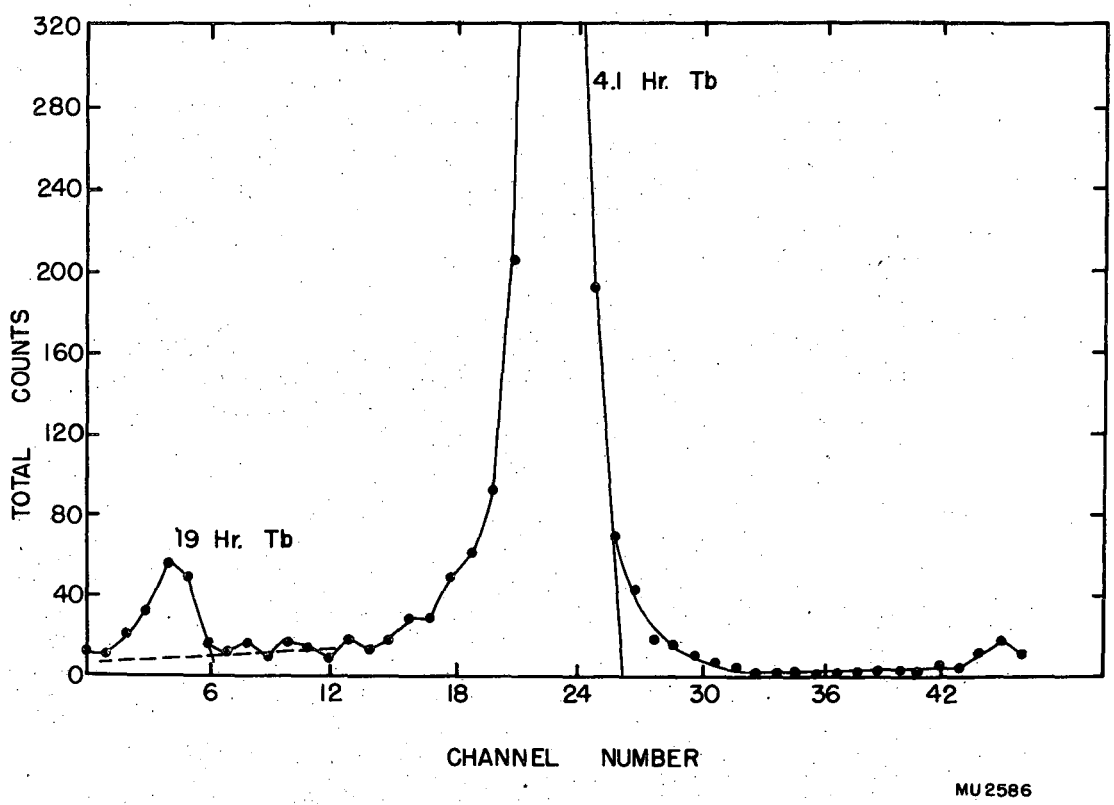

Fig. 8

Alpha pulse analysis curve of terbium alpha activity showing peaks corresponding to the 4.1 H and 19 H activities

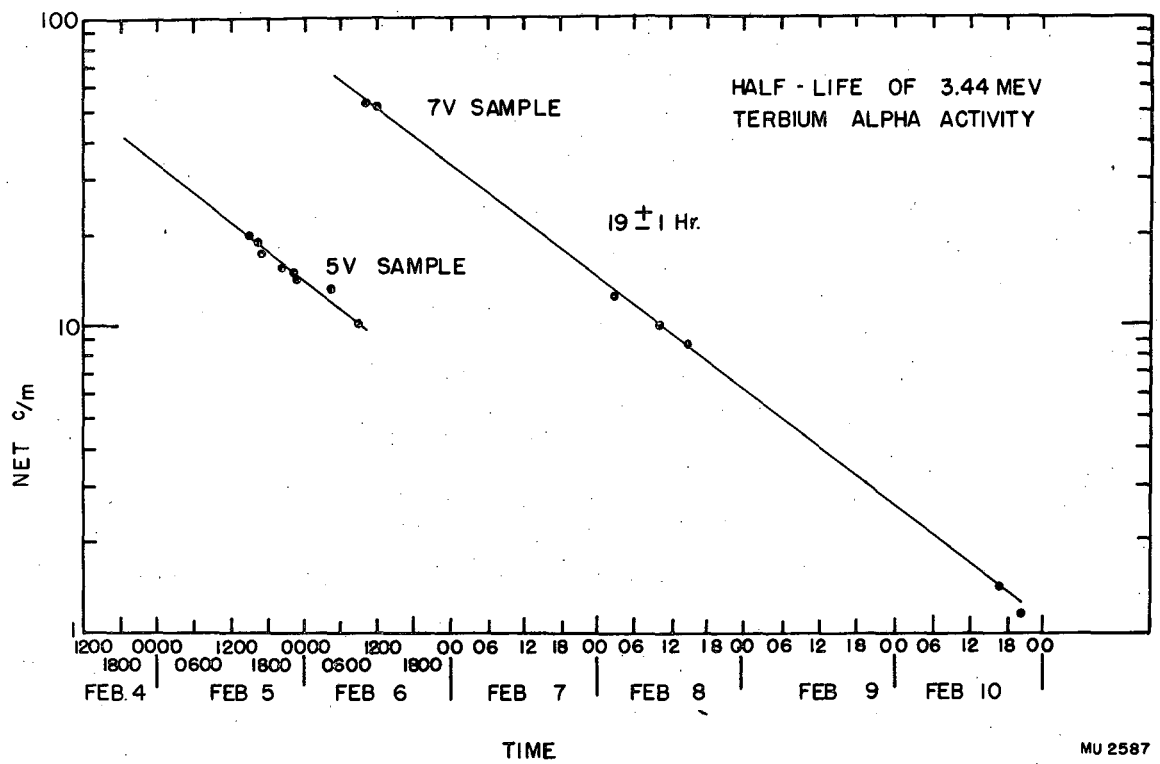

Fig. 9

Decay curve of total counts in the 3.4_4 Mev terbium alpha activity peak on pulse analyser counts

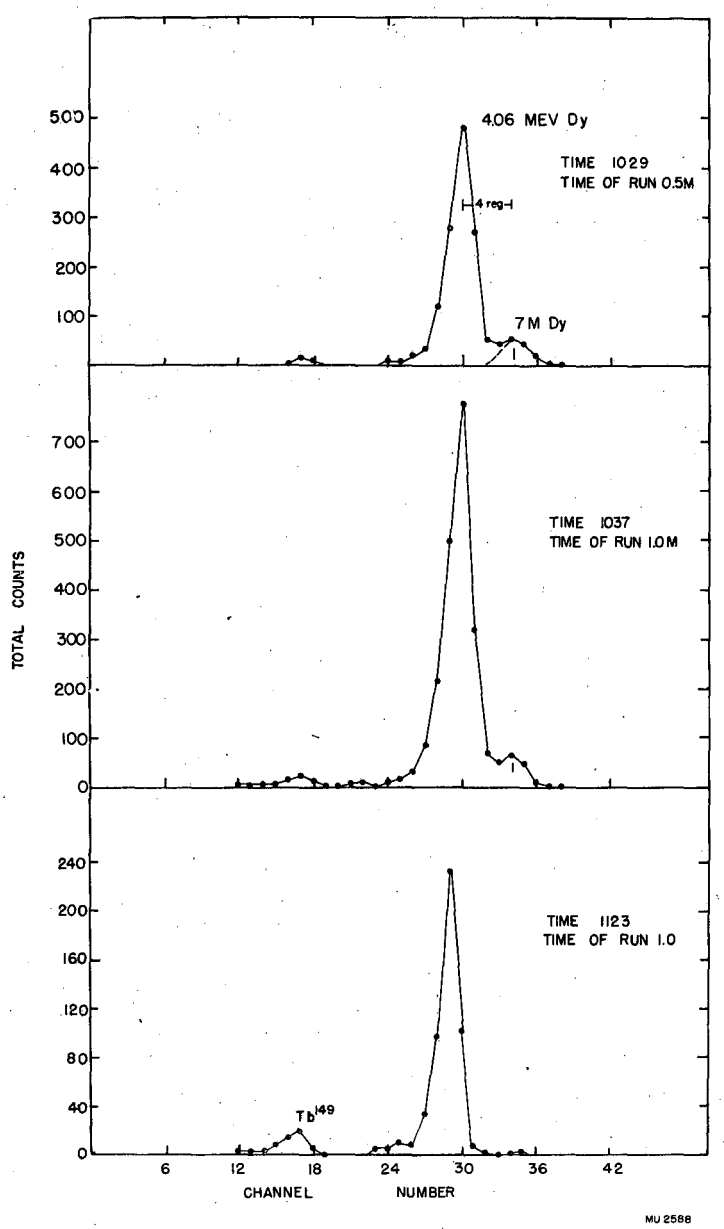

Fig. 10

Alpha pulse analysis curves of a sample of dysprosium alpha activities, showing decay

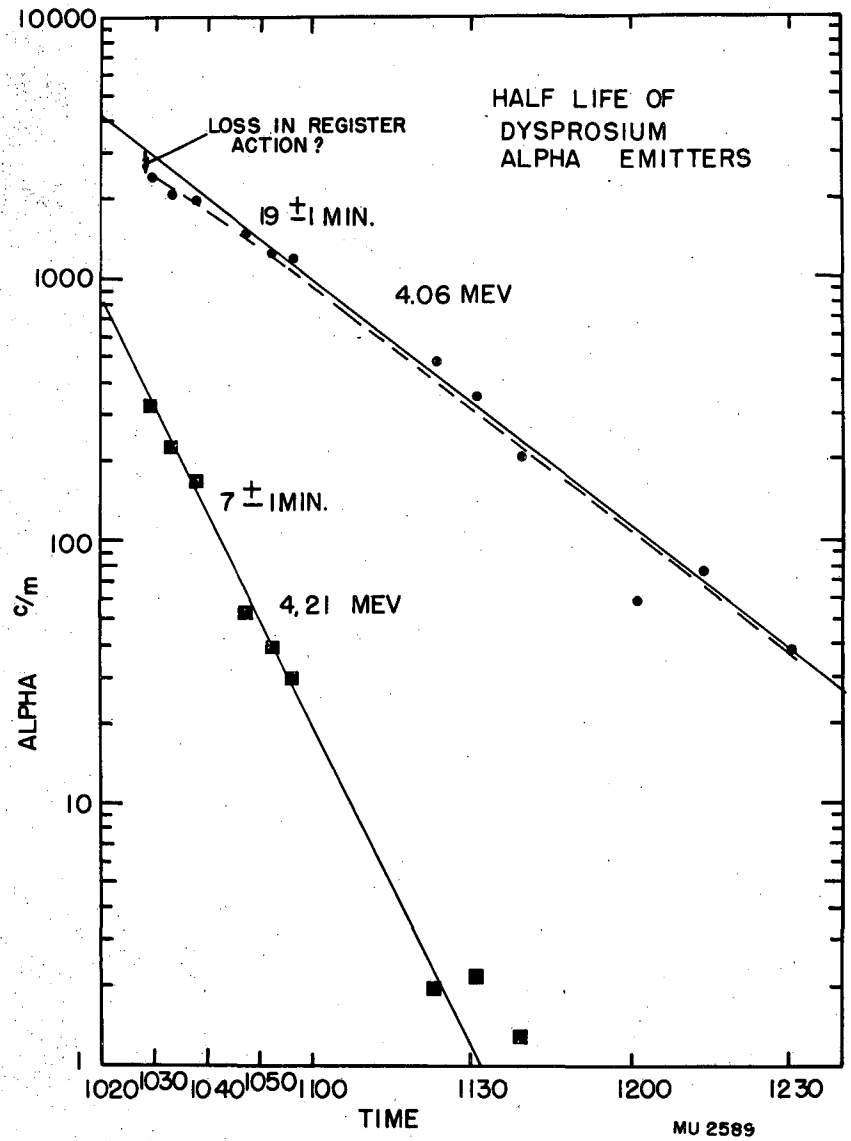

Fig. 11

Decay curves of dysprosium alpha emitters from pulse analyser counts.

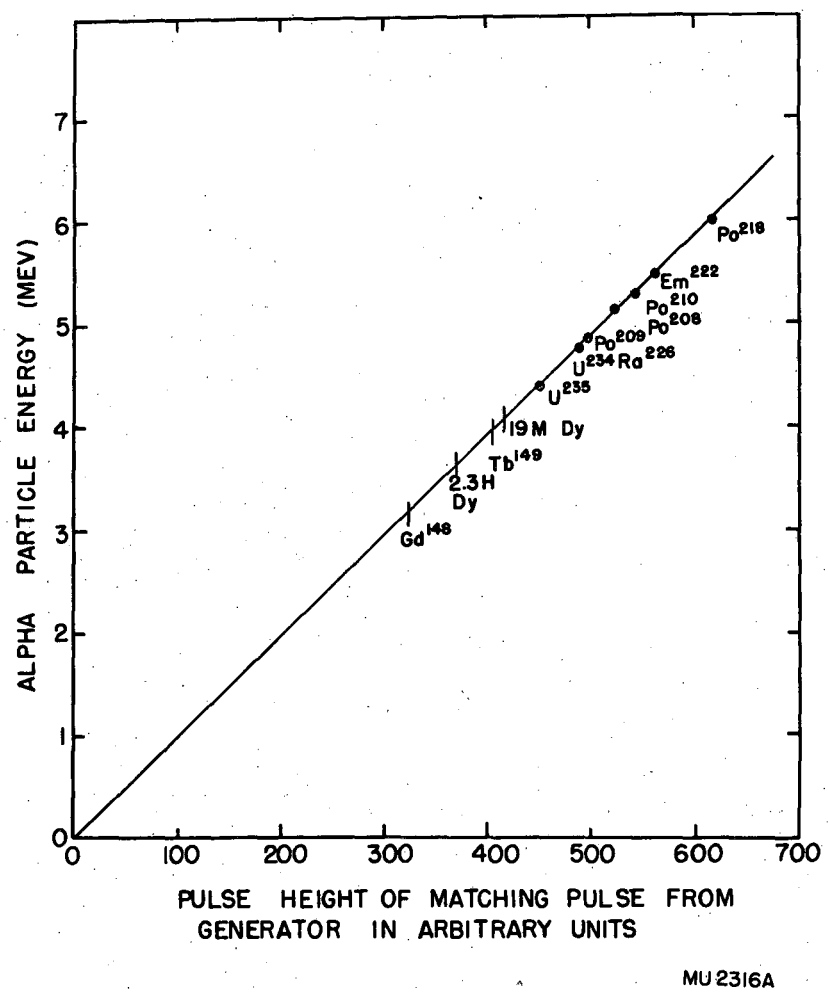

Fig. 12

Alpha particle energy versus pulse height of matching pulse from pulse generator. Energy standards are shown by dots. Unknown energies determined from plot by the ordinate of the intersection of plot line with vertical dashes.

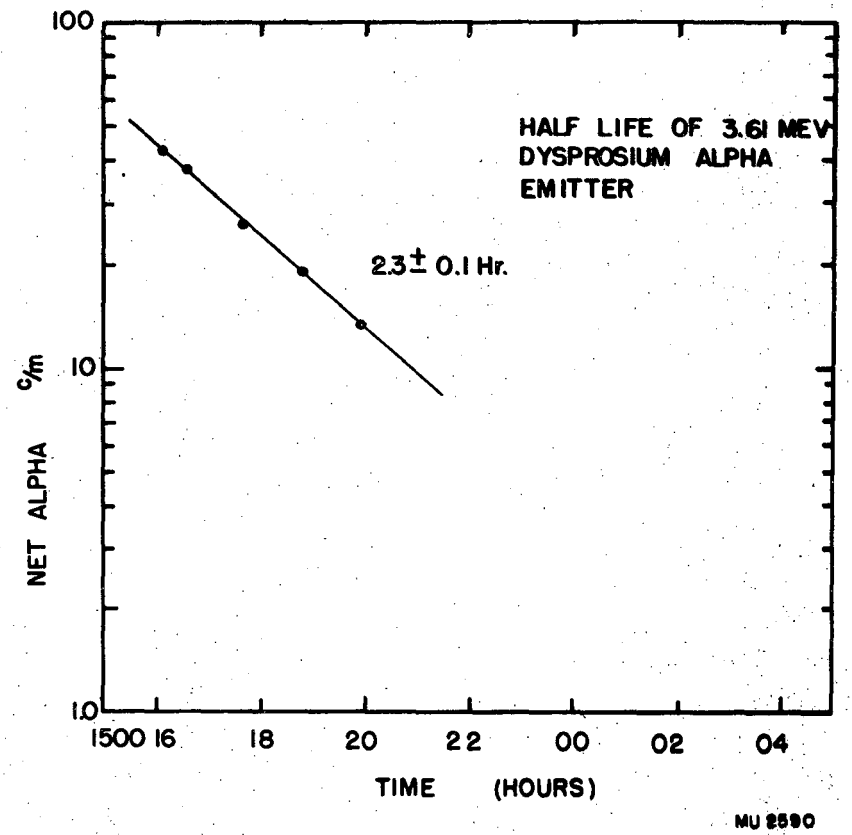

Fig. 13

Decay curve of 3.61 Mev dysprosium alpha activity from pulse analyzer counts

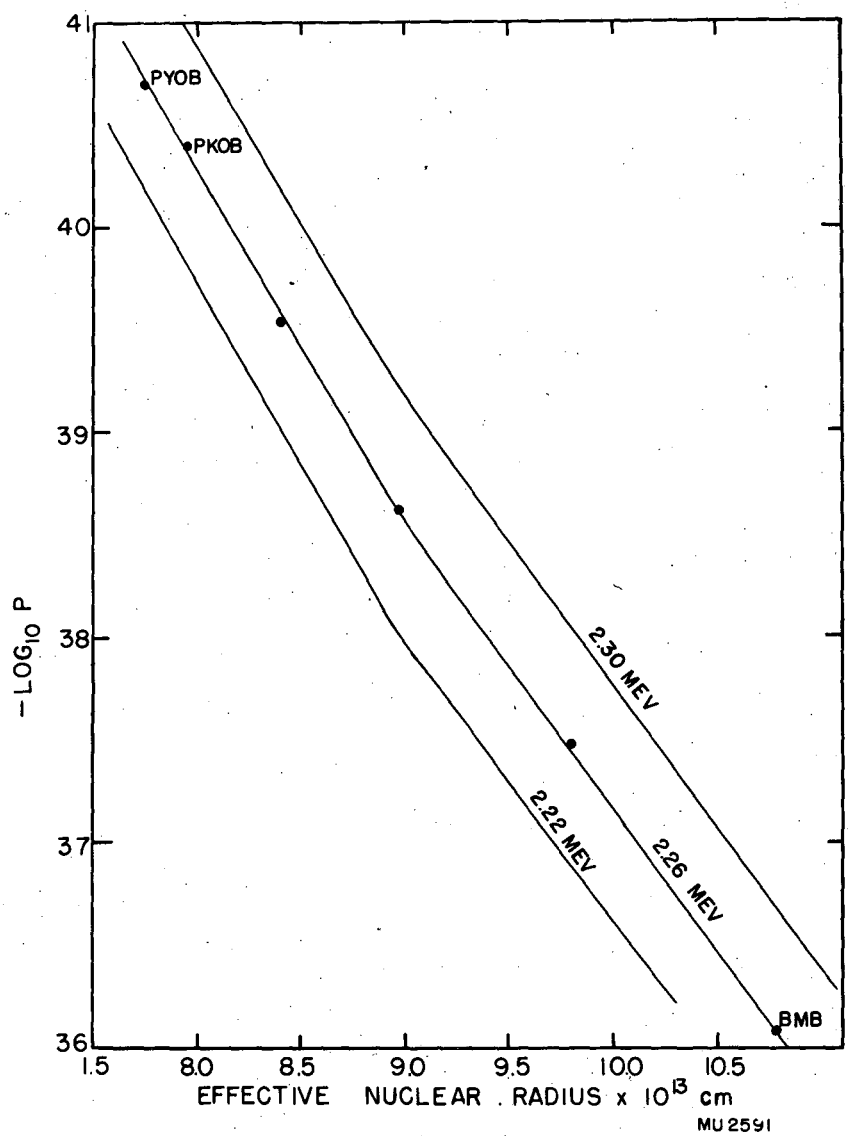

Fig. 14

Plot showing the calculated dependence of the exponential barrier penetration factor on values chosen for alpha decay energy and nuclear radius in Sm^{147} (Z = 62)

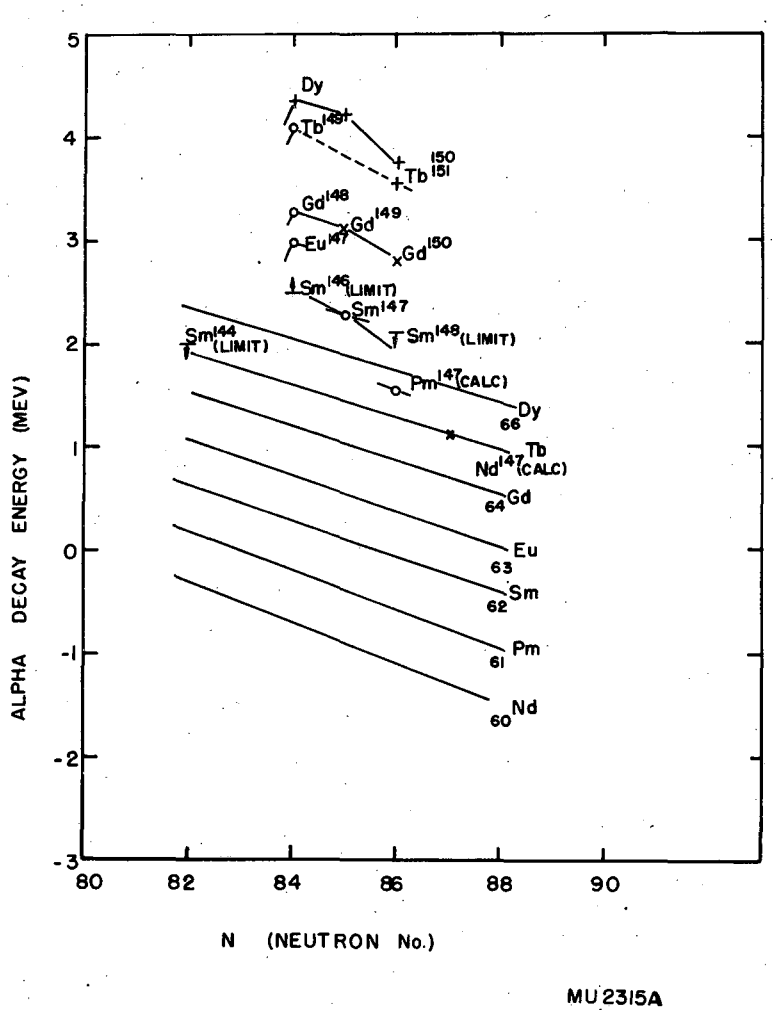

Fig. 15

Alpha decay energy versus neutron number relationships of rare earth nuclides. Lower family of lines defined by the calculated energies from semiempirical mass equation. -o- mass number certain. -x- mass number probable. -+- one of a few mass numbers.

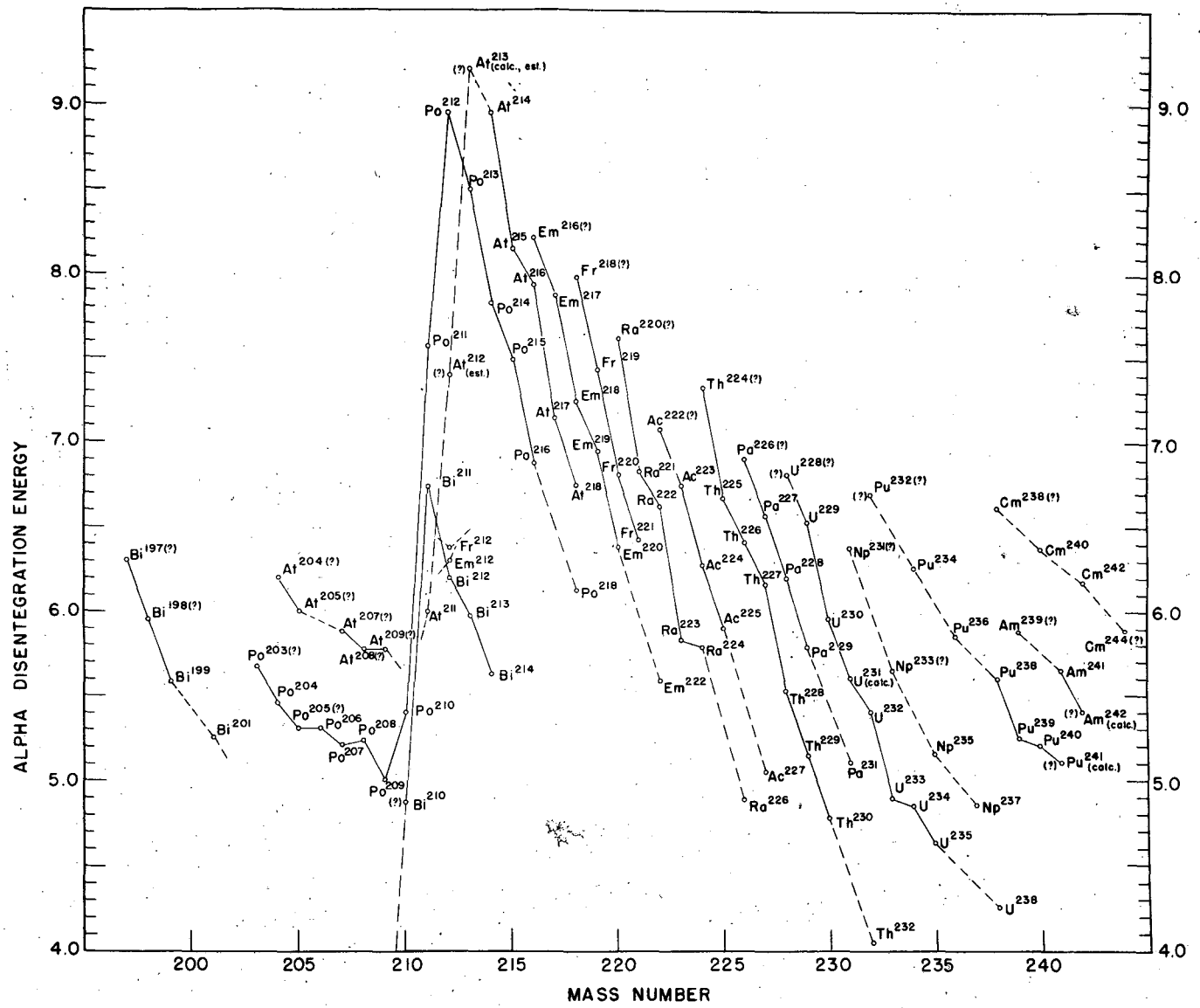

FIG. 16

ALPHA ENERGY VERSUS MASS NUMBER RELATIONSHIPS OF THE HEAVY NUCLIDES. FROM PERLMAN, GHIORSO AND SEABORG[9].

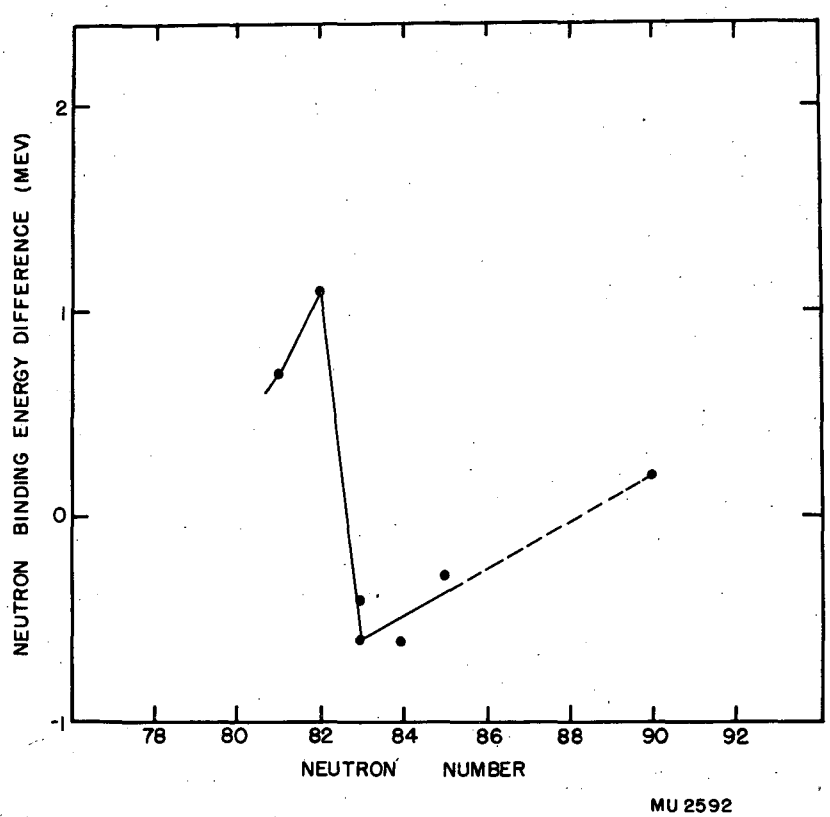

Fig. 17

Neutron binding energy differences vs.

neutron number near 82.

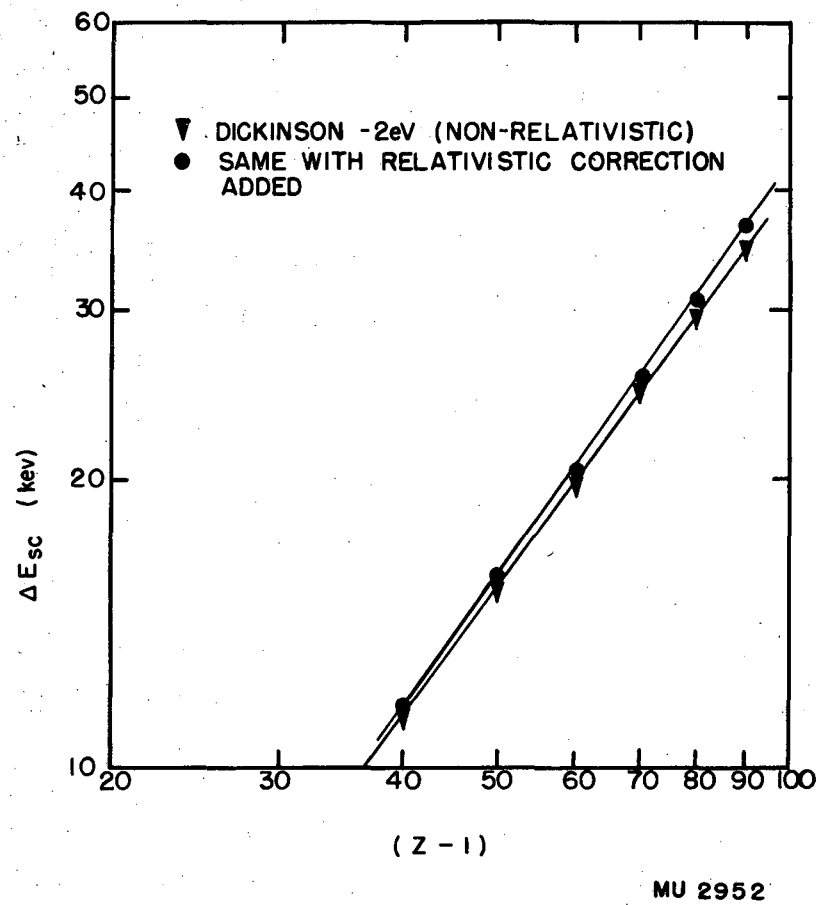

Fig. 18

The additive correction to the atomic alpha decay energy to obtain the nuclear alpha decay energy for decay rate calculations. The correction is in kev vs. the atomic number minus one for the nucleus undergoing alpha decay.