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Berkeley, California

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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 relationship between alpha decay rates and energies in the rare earth region is examined. Calculations of the "effective nuclear radius for alpha particles" were made using five different alpha decay rate formulas.

The trends of the rare earth alpha decay energies are discussed, particularly with respect to the discontinuity in neutron binding energies at the closed shell of 82 neutrons.

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*Most of the material in this article is presented in greater detail in University of California Radiation Laboratory Report UCRL-1473 (1951) and Ph.D. Dissertation, J. O. Rasmussen, Jr., University of California (1952).

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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

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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 semiempirical 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 (contant 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.

A survey was initiated in which targets of various medium heavy elements were 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[10] 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.

II. EXPERIMENTAL RESULTS

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.

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. 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 Isotope (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 to observe its alpha decay branching. Following bombardment of separated isotope $_{62}Sm^{147}$ oxide with 8.5 Mev protons and of natural samarium oxide with 19 Mev deuterons, there was observed a small amount of alpha activity which decayed with about a three week half-life.

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	$3.1_6 \pm 0.1$	>5 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	$3.9_5 \pm 0.04$ 3.97 ± 0.02*	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 hr ± 1	--	--	--	$Gd(p,xn)100$ Mev $Eu^{151}(\alpha,xn)60$ Mev
Dy	66	$149 \leq A \leq 153$	$4.2_1 \pm 0.06$	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

NOTE: No alpha decay fine structure has been detected for any of the alpha emitters listed above. Alpha decay energies are from ion chamber pulse analysis unless otherwise noted.

*Energy determination in magnetic alpha ray spectrograph by Asaro and Rasmussen.[11]

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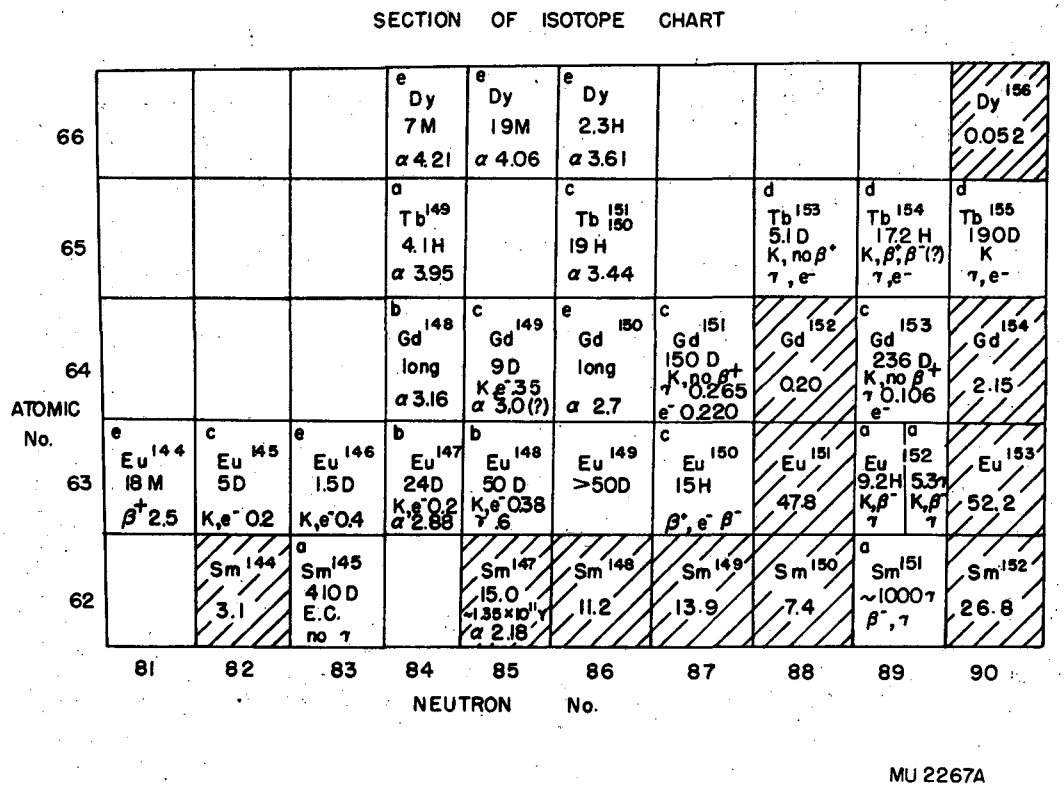


Fig. 1

Section of isotope chart showing the rare earth alpha emitters. Mass numbers of dysprosium alpha emitters are not known.

From the decay curve 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[12] has been recently made by proton and deuteron bombardments of isotopically enriched samarium oxide[10] 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 observed alpha activity in europium is evidently due to alpha decay branching of Eu^{147}. Comparable amounts of Eu^{145}, Eu^{148}, and Eu^{149} have not shown detectable alpha activity and are, thus presumed to have partial alpha decay rates less by at least a factor of five than that of Eu^{147}.

After these bombardments europium activities were separated from the samarium target material by means of the standard cation exchange column elutions described in Appendix I.E. 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, as mentioned. Further proof that it is an europium isotope will be given presently.

Energy determination for the europium alpha activity was made by pulse analysis[13] of a thin vaporized sample, with Gd^{148} and Sm^{147} serving as alpha energy standards. 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.[14]

The energy for the standard Gd^{148} is taken as 3.16 Mev, the determination of which is described in the following section, II 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 $\pm$0.1 Mev.

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.E). 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).

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 has not been determined yet and the counting yield of the Geiger-Mueller counter is not known. 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[15]) operated at 5 kev. The counting yield of the Nucleometer for electron 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 could 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 particles 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. The uncertainty of this partial alpha half-life estimate is largely due to the counting yield assumption for EC, but the value 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^{10} 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, which procedures are described in Appendix I.E. 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 to be 3.16 ± 0.1 Mev by pulse analysis and comparison with many heavy element alpha emitting standards of known energy. The

conservatively large error limits are given because the alpha energy standards were of much higher energy.

Counting data of the 3.16 Mev alpha activity over a two year period 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, and by proton bombardment of natural europium. The alpha particle excitation function, shown in Figure 2, was determined by bombardment in the 60-inch cyclotron and the proton function by 31 Mev proton bombardment in the linear accelerator. 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 uncertainty in the ordinates of the points in Figure 2 may be about 30 percent arising from uncertainties in chemical yield and self absorption of alpha particles in the relatively thick samples.

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 2 with reaction threshold 28-30 Mev is that of an α,n, $\alpha,2n$, $\alpha,3n$, or $\alpha,4n$ 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

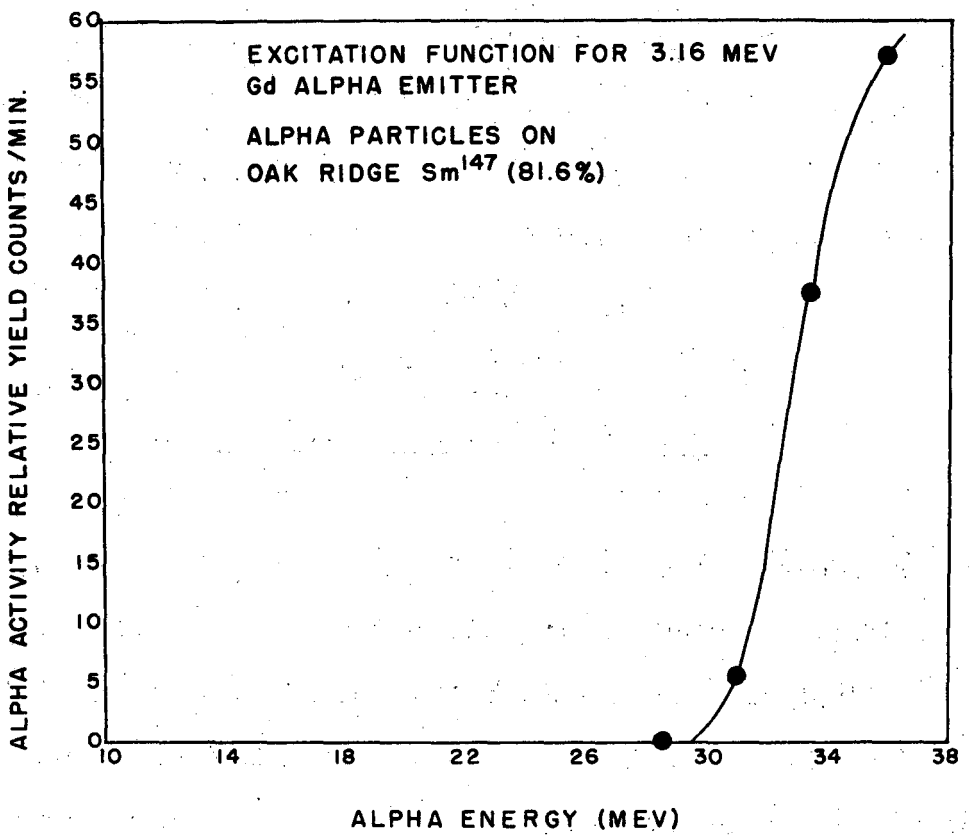


Fig. 2

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

were made by calculation from masses in the atomic mass table of Metropolis and Reitwiesner[16] (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[17]). 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 considerable energy available for electron capture decay and consequently a much shorter half-life than 35 years.

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,

$$T_{1/2\alpha} / \sigma(\alpha,3n) \approx 1.4 \times 10^2 \text{ years barn}^{-1}.$$

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,[18] the estimate of the partial alpha decay half-life of Gd^{148} would be 1.4×10^2 years. This estimate is probably 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. 3.0 Mev Gd^{149}.--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.E) 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.[12] 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 half-life of about 4×10^3 years.

3. 2.7 Mev Gd^{150}.--It has been suggested by Kohman[19] 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 an intense irradiation 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 6 N HCl, the rare earth hydroxides were precipitated with ammonia, and after redissolving with a minimum of 6 N HCl a chemical separation of europium (and partially samarium) from gadolinium was made by the sodium amalgam procedure (Appendix 1.E).

A thin uniform sample of part of the gadolinium fraction on a platinum plate was prepared by the volatilization technique (Appendix 1.E). This sample exhibited 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,[20] 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.[21]

The mass assignment of this alpha activity to Gd^{150} should be considered tentative, since the assignment is based principally on semiempirical considerations of the probable expected alpha decay energy for Gd^{150}. Decay measurements over a one year period show the activity to have a half-life of greater than two years, ruling out the possibility that this activity could arise from 155 day[22] Gd^{151}.

Sun et al.[23] 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 it has not been possible here to detect any rare earth alpha activity of half-life near seven hours following intense irradiations of Sm_2O_3 with 30 Mev alpha particles. 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 $Bi^{209}(\alpha,2n)At^{211}$. 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 (3.95 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. 3.97 Mev Tb^{149}.--This alpha emitter of 4.1 ± 0.1 hour half-life was the first of the artificial rare earth alpha emitters to be observed, 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.E. The 4.1 hour alpha activity was always observed exclusively in the terbium chemical fractions.

The alpha particle energy was determined by pulse analysis to be 3.95 ± 0.04 Mev. A recent energy measurement by Asaro and Rasmussen,[11] using a magnetic alpha ray spectrograph gave the more accurate result 3.97 ± 0.02 Mev.

The mass number 149 was assigned to this activity on the basis of a mass spectrographic determination previously reported[24] by the authors in collaboration with F. L. Reynolds.

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 there were present in all samples of Tb^{149} many other isotopes undergoing decay by electron capture and positron emission. A lower limit on the alpha decay to electron capture branching ratio of about one percent was previously given.[2]

2. 3.4 Mev Tb^{151}.--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 ± 1 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.E. 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 average of several pulse analysis energy measurements gave an alpha particle energy of 3.44 ± 0.1 Mev.

Only a tentative mass assignment of the 19 hour activity can be made at present. The alpha particle excitation work on europium oxide by Rollier and Rasmussen[25] indicated a probable mass assignment to 151, with 150 a possibility.

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,[26] 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, although the lower limit $\alpha/EC > 4 \times 10^{-6}$ (probably much too low) can be set.[25]

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 similar to that outlined by Fermi[27] 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 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 $153 \geq A \geq 149$ to be set on the mass numbers. Further studies of bombardment yields of these alpha emitters in the proton energy range between 70 and 120 Mev, should 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[28] 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, or 120 Mev. Hence, the seven minute activity is attributed to a dysprosium nuclide (Z = 66).

The seven minute alpha activity is seen on the first two pulse analysis curves of Figure 3. From these curves the alpha particle energy of the seven minute activity was determined as 4.21 ± 0.06 Mev. The counting rates of this activity were calculated by the summation of the counts under the pulse analysis peak. The half-life of 7 ± 2 minutes was determined from the slope of the decay curve. Other half-life determinations agree within these limits.

2. 4.06 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, 90, and 120 Mev. The alpha particle energy of the 19 minute dysprosium activity is determined as 4.06 ± 0.04 Mev by pulse analysis.

From several decay curves of this alpha activity the half-life of 19 ± 4 minutes was determined.

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

The element assignment to dysprosium was 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.E.

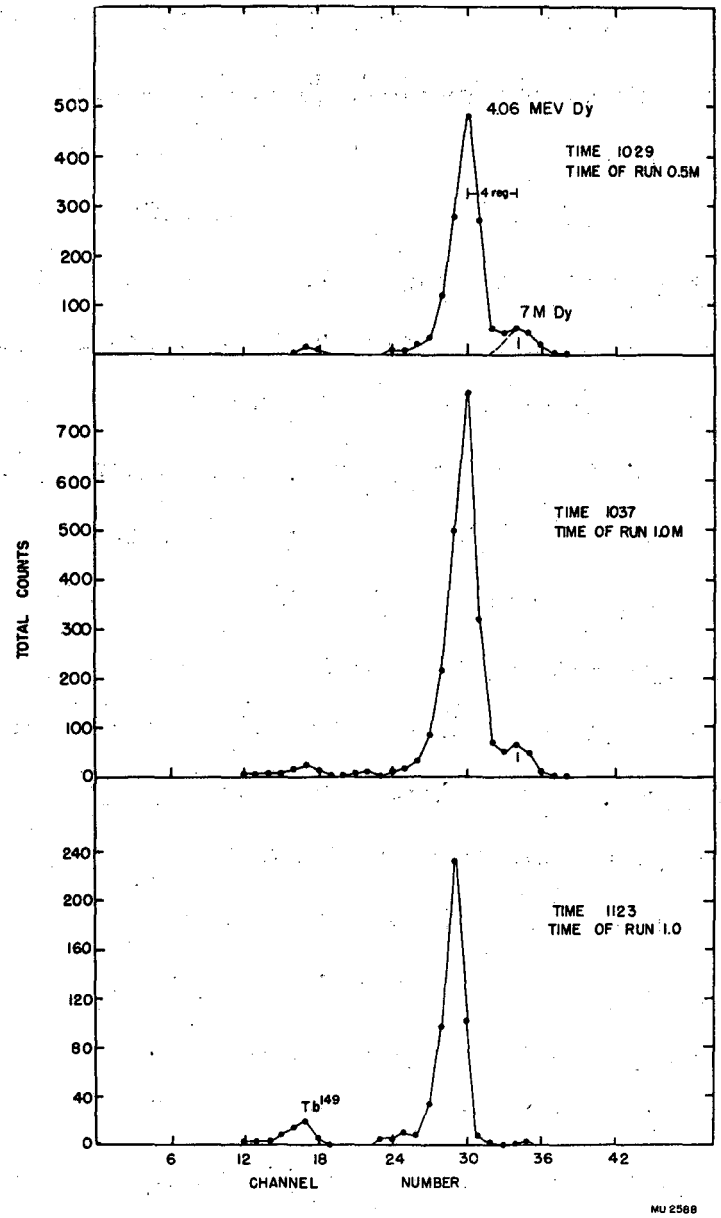


Fig. 3

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

The alpha particle energy was determined by pulse analysis to be $3.6_1 \pm 0.08$ Mev. The half-life was determined from decay curves 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 II, C 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 3.5 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 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 various decay processes 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.

It appears that no appreciable alpha branching is exhibited by neutron deficient nuclides of atomic numbers 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, so significant upper limits on alpha half-lives cannot be set.

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[19] but is missing in nature.

F. Gold Isotope (A = 79)

1. 5.1 Mev Gold.--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.E. 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 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 $\pm$ 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 $\pm$ 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^{-4} as reported in the 1949 paper.[2] This branching ratio should be regarded as uncertain.

G. Mercury Isotopes (Z = 80)

1. 5.6 Mev Mercury.--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.E) from the gold foil. Its alpha particle energy 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 besides the original 190 Mev deuteron bombardments; hence, no significant limits on the mass number can be set at this time.

III. 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 alpha decay rate well determined. The alpha decay rates of Eu^{147}, Gd^{149}, and Gd^{148} are known approximately. Only upper limits can presently be set on the alpha decay rates of the terbium and dysprosium alpha emitters, since their alpha to electron capture decay branching ratios have not yet been determined experimentally.

The correlations of alpha decay rates and decay energies among the heavy element alpha emitters show great regularity if nuclides of even-even nuclear type are alone included. The odd nucleon number alpha emitters exhibit decay rates lower than the correlations of the even-even type nuclides would predict by factors as great as 1000.

If one knew which of several alpha decay rate formulas that have been proposed most nearly represented the true physical situation, it would be possible by using the experimental alpha decay data of $_{64}Gd^{148}$ to calculate the value of the "effective nuclear radius for alpha particles" or "alpha particle penetration radius" for the daughter nuclide Sm^{144}. Alpha decay rate formulas may generally be considered as the product of two factors, as $\lambda = f \cdot P.$, where λ is the alpha decay constant in seconds^{-1}. P represents the penetrability factor of the coulombic potential barrier, and f, the hypothetical "decay constant in the absence of the potential barrier." The potential which the alpha particle experiences is generally idealized as a pure coulombic potential at distances of the alpha particle from the nucleus greater than an "effective nuclear radius" R, at which point there is assumed a sharp cutoff of the coulombic potential to some low value. There is wide variation in the values estimated for f, the decay constant without barrier, depending upon the particular nuclear model chosen. The one body model as applied by Biswas and Patro[29] and by Perlman and Ypsilantis[30] leads to f values of the order of 2×10^{21} sec^{-1}, while at the other extreme the many body model proposed by Bethe[31] leads to f values of the order of 1×10^{15} sec^{-1}. As a result, radius values determined from the experimental data on alpha decay in the heavy region by the latter model exceed by 45 percent those determined by the former model. Other alpha decay formulas lead to results between these extremes.

In view of the considerable variation in f estimates and some other details in various alpha decay rate formulas, the R values (Sm^{144} daughter nucleus) were calculated from Gd^{148} data by five separate rate expressions. The first is the one body formula used by Biswas and Patro[29] and by Perlman and Ypsilantis.[30] The second is a formula derived by one of the authors (J.O.R.).[32] The third is an approximate form of the one body formula derived and applied by Preston[33] and also applied by Kaplan.[34] The fourth is a many body expression of the same form as the second, but based on the procedure suggested by Cohen,[35] which uses alpha penetration radii determined from alpha particle bombardment excitation functions. The radii used were based on the (α,fission) cross section measurements on Th^{232} by Jungermann.[36] The fifth decay expression used is the form of the many body decay model proposed by Bethe.[31]

The details of these five alpha decay formulas can be found in Appendix II.

R values (Em^{220} daughter nucleus) from Ra^{224} data were also calculated by these expressions and are presented in Table III. From these R values in the two regions constants "a" and "b" were determined to give agreement with the effective nuclear radius formula of type

$$R = (aA^{1/3} + b) \times 10^{-13} \text{ cm},$$

where A is the mass number. The first term in the expression may be associated with the radius of the nucleus proper and the second term with the "radius of the alpha particle" plus the range of nuclear forces. A radius expression in which the constant term might have greater physical significance was felt to be the following, employing Present's[37] nuclear radius formula which takes into account a finite compressibility of nuclear matter:

$$R = \left\{a'A^{1/3}\left[1 + 0.8(A-2Z)^2/A^2 - 0.3/A^{1/3} + 0.010\ Z^2/A^{4/3}\right] + b'\right\} \times 10^{-13} \text{ cm}$$

The constants a, b, a', and b' are given in Table III with the five sets of R values.

Table III

Alpha Particle Penetration Radius Values Calculated from Alpha Decay Rate Data and Corresponding Constants for Radius Expressions

Rate formula	Penetration radii (in 10^{-13} cm)		Constants in Radius Expressions			
			$aA^{1/3} + b$		Present's Eqn.[37]	
	Ra^{224}	Gd^{148}	a	b	a'	b'
1. BP and PY	8.70	7.55	1.46	-0.10	1.085	1.79
2. R	9.01	7.84	1.48	0.07	1.10	2.01
3. PK	9.25	8.00	1.58	-0.30	1.18	1.75
4. RCJ	10.16	9.10	1.34	2.07	1.00	3.80
5. BMB	12.60	11.78	1.04	6.32	0.77	7.70

The b' values for formulas (1), (2), and (3) seem to be reasonable physically, while the b' values for (4) and (5) seem too large. This comparison would favor the decay formulas (1), (2), and (3) with the higher f values over those with lower values. It should be borne in mind in this regard that the uncertainties in the experimental alpha decay data for Gd^{148} suggest error limits on the calculated R values of about ± 5 percent. The constant terms b and b' in the radius expressions are very sensitive to the difference between the R values for Ra^{224} and Gd^{148}. A change of the Gd^{148} R value by ± 5 percent would cause a change in b and b' values of about ± 3. Alpha decay formula (5) would still appear to be excluded by too large a value for b', but formula (4) could not be ruled out.

Using the R values for Gd^{148} the alpha decay rates of the other rare earth alpha emitters were calculated from the various decay expressions. Many of these alpha emitters are not even-even nuclear type. From observations among the heavy elements one would expect the calculated rates to be merely upper limits for nuclei not of even-even type. Lower rates would not be surprising in view of the common occurrence of hindrance with odd nucleon type alpha emitters. The calculations for the rare earth alpha emitters are summarized in Table IV, as the logarithms to the base ten of the alpha decay mean life τ in seconds ($\tau = 1/\lambda$).

The decay rates calculated for each alpha emitter by the various decay formulas are seen to differ very little. The different decay formulas evidently give about the same dependence of decay rate on decay energy, since no significant systematic differences from formula to formula are evident. As to the agreement with the experimental decay rates, it is seen that no disagreement occurs with the lower limits in the terbium and dysprosium nuclides, for which good α:EC branching ratios are not known. Note, however, that Gd^{149}, Eu^{147}, and Sm^{147} all exhibit progressively larger actual decay rates than are calculated. This discrepancy is of the opposite sign from the possible hindrance associated with odd nucleon nuclear types. Sm^{147} exhibits a decay rate ten times greater than predicted. The discrepancy may not be fundamental but may be due to experimental error in the alpha particle energy value of 2.18 Mev used for Sm^{147}. This value was determined by Jesse and Sadauskis[14] in argon filled ionization chamber. The assumption that total ionization in argon is strictly proportional to alpha particle energy was tested and used in their Sm^{147} energy determination. If this assumption is not strictly true, and there is an "ionization defect," the energy value 2.18 Mev for Sm^{147} is too low. So sensitive are the decay rate calculations in Sm^{147}

Table IV

Calculation of Decay Constants Using Nuclear Radii Fitting Gd^{148} Decay

Nuclide	Total* Nuclear Alpha Decay Energy (Mev)	$\log_{10} \tau$ (τ is the mean life for alpha decay in seconds. $\tau = 1/\lambda$)					
		Experimental	1 BP and PY	2 R	3 PK	4 RCJ	5 BMB
		Nuclear** Radius (in 10^{-13} cm) →	7.55	7.84	8.00	9.10	11.78
Sm^{147}	2.26	18.79	19.89	19.87	19.74	19.88	19.77
Eu^{147}	2.98	11.42	11.98	11.98	11.91	12.01	11.94
Gd^{148}**	3.27	9.98	--	--	--	--	--
Gd^{149}	3.1	11.26	11.56	11.55	11.50	11.56	11.51
Gd^{150}	2.8	--	14.66	14.65	14.58	14.60	14.50
Tb^{149}	4.08	>4.33	4.66	4.64	4.64	4.68	4.73
Tb^{151}	3.56	>5.00	8.32	8.30	8.27	8.31	8.28
Dy	4.35	>2.84	3.62	3.61	3.53	3.63	3.69
Dy	4.20	>3.21	4.54	4.50	4.51	4.50	4.58
Dy	3.73	>4.08	7.68	7.68	7.66	7.64	7.64

The following notes belong under Table IV.

*The "Total nuclear alpha decay energy" is the decay energy (alpha particle plus recoil) that the nucleus stripped of electrons would exhibit. It is calculated by adding to the observed alpha particle energy the recoil energy and an energy term equal to the electrostatic potential energy of the alpha particle at the nucleus due to the electron cloud (cf. reference 32, Appendix II). This last correction amounts to 30 kev for uranium, 26 kev for polonium, and about 20 kev in the rare earth region. A graph of the correction term applied is shown is Figure 4. This graph was plotted using the potential calculations of Dickinson.[38]

**Radius values are based on the experimental decay rate of Gd^{148}.

to decay energy that an increase in decay energy of only 70 kev would remove the factor of ten discrepancy in the values of Table IV. The smaller discrepancies in Gd^{149} and Eu^{147} would be nearly corrected, too, by such a change, since their decay energy values are based on linear interpolation of ionization chamber measurements between the energy standards Gd^{148} and Sm^{147}.

In Table V the $\log_{10}\tau$ values for formula (2) in Table IV have been converted to half-live values and are compared with the experimental half-lives.

Table V

Comparison of Calculated and Experimental Alpha Half-Lives

Nuclide	Total decay energy	Experimental alpha half-life	Alpha half-life (from formula 2)
Sm^{147}	2.26	1.35×10^{11} y	1.6×10^{12} y
Eu^{147}	2.98	$\sim 6 \times 10^{3}$ y	2.2×10^{4} y
Gd^{148}	3.27	$\sim 1.4 \times 10^{2}$ y	—
Gd^{149}	3.1	$\sim 4 \times 10^{3}$ y	7.8×10^{3} y
Gd^{150}	2.8	—	9.8×10^{6} y
Tb^{149}	4.08	>4.1 h	8.4 h
Tb^{151}	3.56	>>19 h	2.9 y
Dy	4.35	>8 m	42 m
Dy	4.20	>19 m	6.2 h
Dy	3.73	>2.3 h	256 d

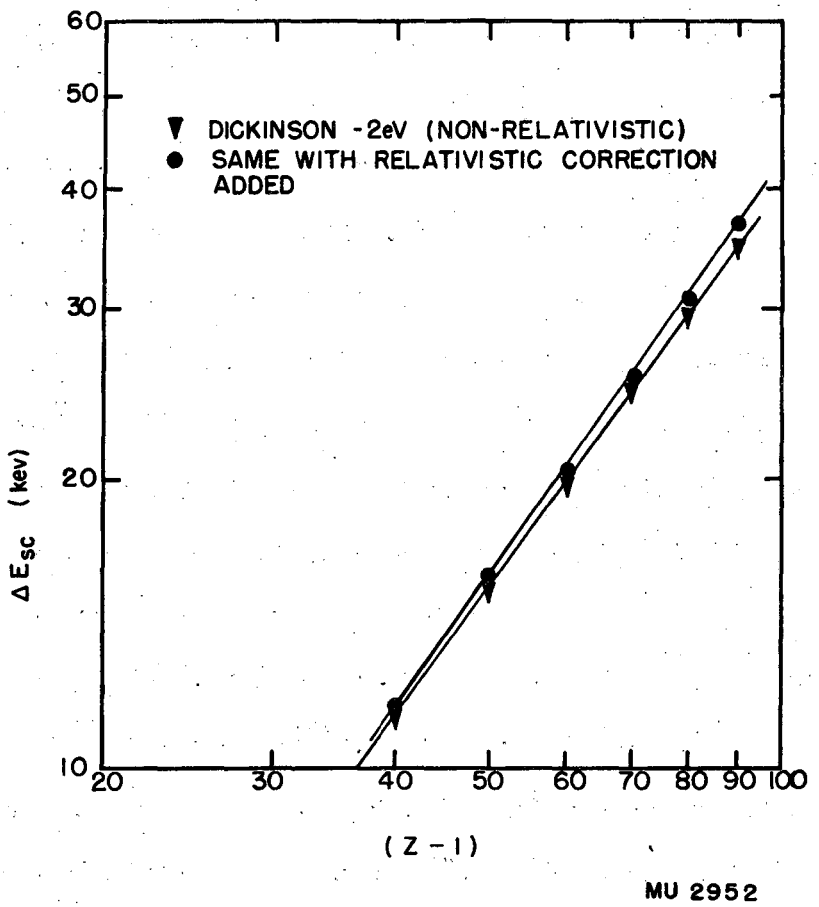


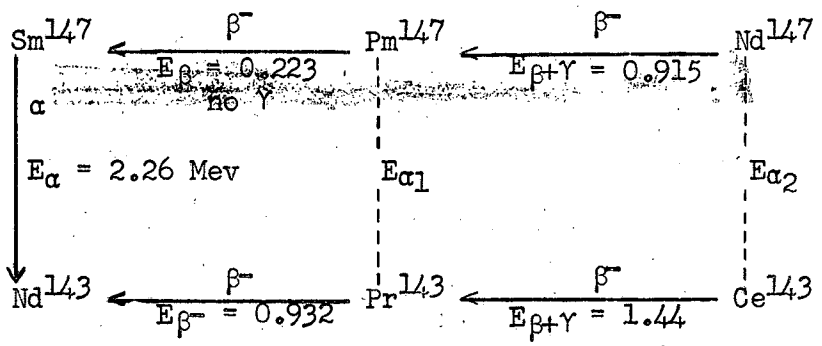
Fig. 4

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 versus the atomic number minus one for the nucleus undergoing alpha decay.

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[39] 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 generally 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[40] and by Rasmussen et al.,[24] and the alpha particle energy 2.18 Mev as determined in an argon filled ionization chamber by Jesse and Sadauskis[14] is used.)



Conservation of energy requires that

$E_{\alpha 1} = 2.26 + 0.223 - 0.932 = 1.56$ Mev

$E_{\alpha 2} = E_{\alpha 1} + 0.915 - 1.44 = 1.04$ 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,[42] and Ce^{143} energies are from the work of Mandeville and Shapiro.[43] 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 VI 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.[44] The lower energy limit in Table VI 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 VI also lists the alpha decay energies calculated from the semiempirical mass equation using the tables of Metropolis and Reitwiesner[16] and assuming a mass for He^{4} of 4.00390 atomic mass units and the mass energy conversion factor of 931.4 Mev = 1 amu.

In Figure 5 the energy data of Table VI are plotted against neutron number.

Table VI

Alpha Decay Energies of Rare Earth Nuclides

Nuclide	N	E_α (Exp.) (Mev) including screening correction	"Normal" E_α (Calc. from semiemp. 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.

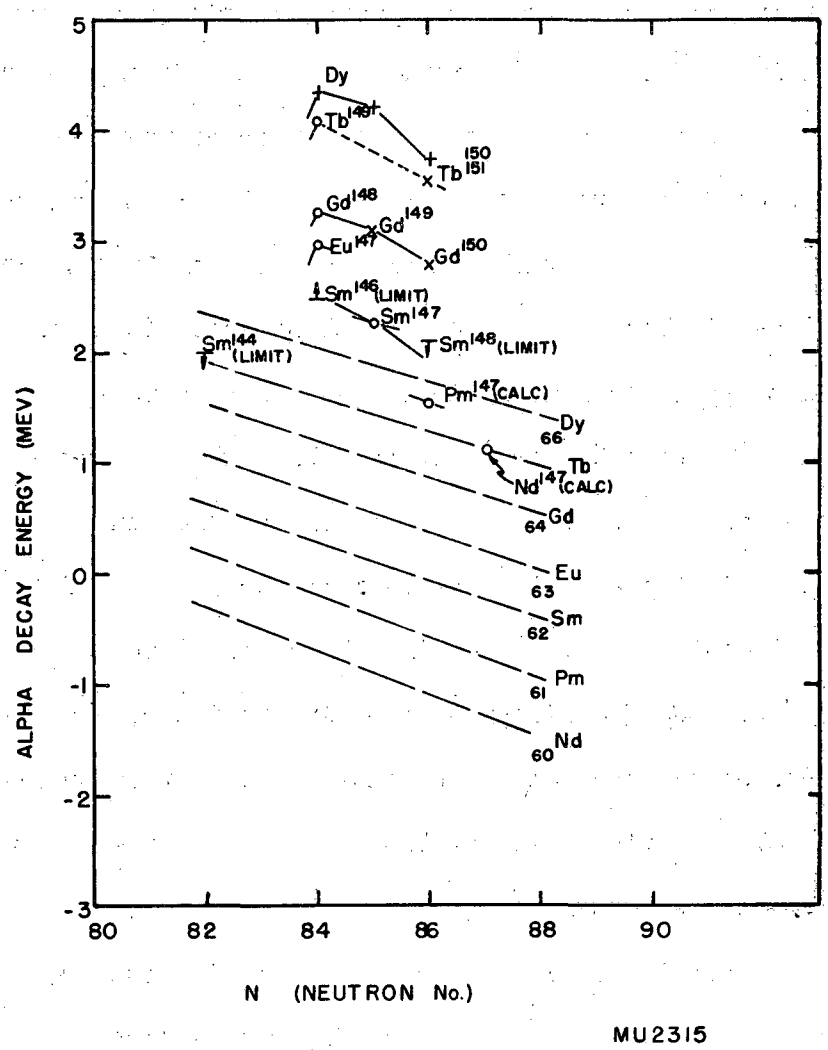


Fig. 5

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

The gadolinium isotopes constitute the only isotopic sequence for which the mass numbers are fairly certain. Mass assignments on the plot for the dysoprosium isotopes are guesses based on the expectation that the various curves of Figure 5 should be nearly parallel, as are those of Figure 6, a plot of alpha energies against mass number in the heavy region.

It is evident from Figure 5 that a maximum in alpha decay energy should occur for the samarium isotopes at 83 or 84 neutrons. In section II 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 occurs in the 84 neutron nuclides. Furthermore, no alpha activity ascribable to a nuclide with less than 84 neutrons has yet been observed.

The normal trend for alpha decay energy to increase with decreasing neutron number is seen from the family of parallel lines in Figure 5 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 structural 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

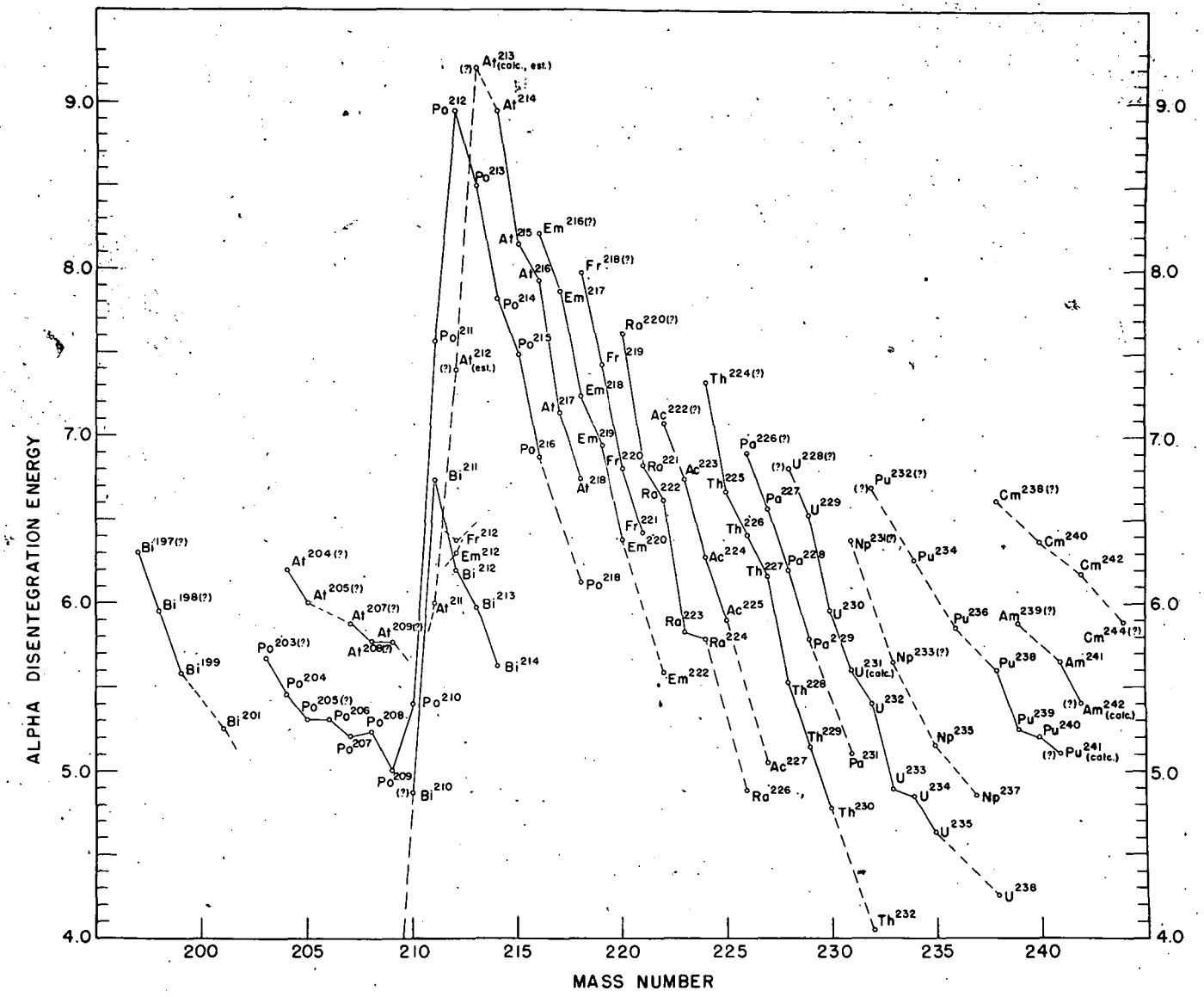


FIG. 6

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

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,[17] 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[45] 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, the relationship to the closed shell was discussed by Thompson et al.[2] and by Perlman et al.[9] 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[17] 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.

From small extrapolations of the curves in Figure 5 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. Since the magnitude of the discontinuity in neutron binding

energies at 82 neutrons is about 1.5 Mev,[17] 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 a few neutrons less than 84 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 for the 84 neutron nuclides 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 VI, 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[45] nuclear shell model could account for a slight extra stabilization of the 64 proton configuration.

IV. ACKNOWLEDGMENTS

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Appendix I. EXPERIMENTAL METHODS

A. Production of Alpha Active Isotopes

For the production of the alpha active isotopes 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 in platinum or aluminum envelopes 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.[46] 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 discs. The discs 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.[13] 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 nuclides than for heavy element nuclides, the alpha particles emitted by the rare earth alpha emitters have energies less than those of any heavy element alpha activities of comparable alpha decay rates by at least 1 Mev. Thus, from the observation of alpha particle energy and alpha decay rate alone, an estimate of the atomic number of the alpha active nuclide can be made with an uncertainty no greater than ten.

C. 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.[13] 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 to alpha particle energy over a wide range of energy has been shown by Jesse, Forstat, and Sadauskis.[47]

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.

D. 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. With long lived Gd^{148}, for example, an estimate of the partial alpha half-life had to be made from bombardment yields.

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, such as the beta emitter Li^{8}.

Theoretical considerations concerning alpha decay rates as a function of decay energy make it 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.

E. 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.D 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 is the situation with any of the

alpha emitters 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[48] (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°, 83°, or 87° C. The apparatus and techniques employed in the column separations at elevated temperatures have been described in detail in a recent article.[46] 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.[49]

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[49] 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 below was found to give a fair separation between europium and gadolinium. 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.[50]

Amalgam Procedure: Dissolve the europium oxide or hydroxide (not more than 10 mg) 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.

Following the deuteron bombardments of gold targets the radioactive mercury fraction was separated from the 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.

F. 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.[24] A third possible 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. 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 by the authors in collaboration with Reynolds[24] on the mass assignment of the Tb^{149} and Sm^{147} alpha activities. The mass spectrographic 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.

G. 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. ALPHA DECAY RATE FORMULAS

The five alpha decay rate formulas used for the calculations in III A will be discussed here in order of decreasing "frequency factor" f in the general rate expression, $\lambda = fP$.

The penetration factor P in all cases was calculated by the exponential expression (600)* of Bethe.[31] The reduced mass of the system and the total nuclear decay energy were used for the calculations.

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*A table has been prepared of Bethe's $\Upsilon(x)$ function to five decimal places as a function of the argument $y = (1 - x)^{1/2}$ over the range of y, 0.750(0.001)0.946. Copies of the table are available from the authors on request.

The various decay formulas differ in the factor f multiplying the exponential factor P.

The first formula, used by Biswas and Patro[29] and by Perlman and Ypsilantis,[30] sets $f = \frac{v}{R}$, where v is the velocity of alpha particle (center of mass system) and R is the nuclear radius.

The second formula, derived by one of the authors (J.O.R.)[32] with a many body model relating f to nuclear level spacing, gives f as

$$f = \frac{2D^*}{\pi\hbar}\left(\frac{E - U}{B - E}\right)^{1/2},$$

where E is the alpha decay energy, B is the "barrier height" ($B = 2Ze^2/R$), U is the "potential energy of the alpha particle within the nucleus" (and is assumed equal to zero) and D* is an energy presumably of the same order of magnitude as the average level spacing between nuclear levels of the same spin and parity as the alpha emitting state.

In the case of even-even nuclides, whose ground states have zero spin and even parity, D* might be estimated by the energy of the first excited state of zero spin and even parity. Experimental evidence from alpha decay fine structure studies[51] indicates that this first excited $I = 0,+$ state in heavy nuclides must have energy greater than 300 kev.

Thus, in the absence of direct experimental evidence by which to estimate D*, the rough assumption was made that the first excited $I = 0,+$ state has an energy corresponding to the first excited level of the nuclear "liquid drop" oscillation of lowest order ($n = 2$). For this calculation the following equations of Bohr and Wheeler[52] were used:

$$1/2\hbar\omega_2 = A^{-1/2}\left\{4\pi r_o^2 0 \cdot 2(1 - x)\hbar^2/3M_p\, r_o^2\right\}^{1/2} \qquad \text{BW(26)}$$

where $4\pi r_o^2 0 \approx 14$ Mev BW(12)

and $x = \left(\frac{Z^2}{A}\right)\Big/\left(\frac{Z^2}{A}\right)_{lim} = \left(\frac{Z^2}{A}\right)\Big/ 47.8.$

The calculations give for Ra^{224} the quantum of energy,

$$\left(\hbar\omega_2\right)_{Ra^{224}} = 0.94 \text{ Mev}$$

and for Gd^{148},

$$\left(\hbar\omega_2\right)_{Gd^{148}} = 1.43 \text{ Mev}.$$

These values of $\hbar\omega_2$ were substituted for D* in the alpha decay rate calculations by the second formula. In effect, this means assigning the same nuclear "frequency factor" to alpha decay and to spontaneous fission. It is interesting in this connection to note Frenkel's discussion[53] of spontaneous fission and alpha emission as closely related processes.

The third alpha decay rate formula is an approximation to that derived by Preston[33] from a one body model using the steepest descents method of approximating the coulomb wave function. This formula was also used by Kaplan[34] in his correlation of even-even alpha emitter data.

The Preston formula is given as two simultaneous equations from which one can solve for both the nuclear radius, R, and the "internal potential energy," U, of the alpha particle within the nucleus.

$$\lambda = \frac{2v}{R}\,\frac{\mu^2 \tan\alpha_o}{\mu^2 + \tan^2\alpha_o}\,P$$

and

$$\mu = -\tan\alpha_o \tan(\mu kR)$$

where R and P have their usual meanings defined earlier in this section, v is the alpha particle velocity corresponding to the energy E, and k is its wave number.

$$\mu = \left(1 - \frac{U}{E}\right)^{1/2}$$

and

$$\alpha_o = \arccos\left(\frac{E}{B}\right)^{1/2} .$$

Now $\tan\alpha_o = \tan\arccos\left(\frac{E}{B}\right)^{1/2} = \left(\frac{B - E}{E}\right)^{1/2}$, and $v = (2E/m)^{1/2}$

so one obtains on substitution in the first equation,

$$\lambda = \frac{2^{3/2} E^{1/2}}{m^{1/2} R} \frac{\left(1 - \frac{U}{E}\right)\left(\frac{B - E}{E}\right)^{1/2}}{\left(1 - \frac{U}{E}\right) + \left(\frac{B - E}{E}\right)} P$$

$$\lambda = \frac{2^{3/2}}{m^{1/2} R} \frac{(E - U)(B - E)^{1/2}}{(E - U) + (B - E)} P \qquad (2.1)$$

The Preston equations can easily be put in a more convenient approximate form. Kaplan[34] showed from his correlation that

$$E - U \approx \pi^2 \hbar^2 / 2mR^2; \qquad (2.2)$$

that is, that the kinetic energy of the alpha particle within the nucleus in this one body model nearly equals the energy of the 1S state of the alpha particle in a spherical square potential well with radius R and with infinite walls. E - U is about 0.52 Mev for the heavy element alpha emitters correlated by Kaplan. Thus, if E - U be neglected with respect to B - E, one obtains from equation (2.1),

$$\lambda = \frac{2^{3/2}(E - U)}{m^{1/2} R (B - E)^{1/2}} P. \qquad (2.3)$$

Finally, substituting for E - U from (2.2) one obtains

$$\lambda = \frac{2^{3/2}\pi^2\hbar^2}{m^{3/2}R^3(B - E)^{1/2}} P \qquad (2.4)$$

the expression used as formula (3) in the calculations of section III A.

The fourth formula uses an f factor with the same energy dependence as the second formula.

$$F = f_o\left(\frac{E}{B - E}\right)^{1/2} \qquad (2.5)$$

However, the numerical calue of f_o was taken as the mean of f_o values calculated from heavy element even-even alpha emitter data using the radius expression

$$R = (1.43A^{1/3} + 1.48) \cdot 10^{-13} \text{ cm.} \qquad (2.6)$$

The average of log f_o = 19.10 from a correlation of Rasmussen.[54] This procedure follows Cohen's[35] correlation using penetration radius values determined from total nuclear absorption cross sections for bombarding alpha particles. The radius expression used above is taken from the (α,fission) excitation function work of Jungermann[36] on Th^{232}, compared with Weisskopf's[55] theoretical calculations.

The fourth decay expression then is as follows:

$$f = 1.26 \cdot 10^{19}\left(\frac{E}{B - E}\right)^{1/2} \qquad (2.7)$$

The fifth decay expression is the form of the many body decay model proposed by Bethe,[31] where,

$$f = 6.2 \cdot 10^{14} E^{1/2}$$

with E in Mev, f in sec^{-1}.

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