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ALPHA RADIOACTIVITY IN THE MEDIUM HEAVY ELEMENTS

J. O. Rasmussen, S. G. Thompson and A. Ghiorso

September 20, 1951

Berkeley, California

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ABSTRACT

In a survey for alpha activity among cyclotron-produced neutron-deficient nuclides of the medium heavy elements alpha activity was found in a number of rare earth nuclides with atomic number greater than 62 (samarium) and in a gold and a mercury nuclide. The experimental results of studies of the various alpha activities discovered in this investigation are presented.

Experimental values of alpha decay rates are compared with calculations from four different alpha decay rate formulas. The trends of the rare earth alpha decay energies are noted and their interpretation as a consequence of a decrease in neutron binding energies just beyond the closed shell of 82 neutrons proposed.

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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. For the initial survey it seemed advisable to bombard with deuterons or protons of reasonably high energy (~200 Mev) and to examine the target material for alpha radioactivity as soon after bombardment as possible in an argon-filled ionization chamber connected to a 48-channel differential pulse analyzer.[12] Thus, in addition to detection of alpha radioactivity an alpha particle energy measurement by comparison with alpha-emitting standards of known energy was obtained. Such an energy measurement usually sufficed to distinguish the sought-for alpha activities from those alpha activities produced from heavy element impurities in the target material. The fact that the coulombic barrier for alpha particles is less for medium heavy element nuclei than for heavy element contaminant nuclei insures that the alpha radioactivity from a medium heavy element is of lower energy than

the heavy element alpha radioactivities of comparable alpha decay half-lives.

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[13] 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[14] 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.[15] 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.[12] 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 the appendix.

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 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 I.3. The eluting agent for the 82° 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[13] 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 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 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[13] 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.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 3.156 Mev from the plot in Figure 12. An earlier determination using fewer standards gave 3.18 Mev. The value 3.16 ± 0.1 Mev was selected, with conservatively large error limits selected because the alpha energy standards used were of much higher energy.

The 3.16 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,[13] 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 3 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 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 mgm 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 mgm 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 the 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 (3.95 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. 3.95 Mev Tb^{149}. 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 $\sim$ 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 $\pm$ 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 3.42 Mev. From an average of several measurements the energy was set at 3.44 ± 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 $Eu^{151}(\alpha,8n)Tb^{149}$ reaction. In the 60 Mev bombardment the $Eu^{151}(\alpha,6n)Tb^{149}$ reaction was probably proceeding just above its energetic threshold and is 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 4.21, 4.06, and 3.61 Mev energies, it is to be noted that all three are produced in good yield by 100 Mev protons on terbium. The 4.06 and 3.61 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 $153 \geq A \geq 149$ 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 4.21 ± 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. 4.06 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 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 4.06 ± 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 ± 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 3.61 ± 0.08 Mev.

Figure 13 is a semilog plot of the decay through two half-life periods of the alpha counts under the 3.61 Mev peak on the pulse analyzer. The half-life is determined from the slope to be 2.3 ± 0.2 hours.

E. 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 Isotopes (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.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 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^{-4} 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 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.3) 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 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

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

$$\lambda = \lambda_o (E)^{1/2} P,$$

where λ is the alpha decay constant in second^{-1}, E is the alpha decay energy in Mev, P is the exponential barrier penetration factor, and λ_o 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 is 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 λ_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 determined from the work of Jungermann[36] on Th^{232} (alpha, fission) cross sections

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

(A = mass number of daughter nucleus) was used. The additive constant 1.48×10^{-3} cm for the alpha particle radius was used by Weisskopf.[37]

The second formula is the form of the one-body alpha decay expression used by Perlman and Ypsilantis[30] with $\lambda_0(E)^{1/2}$ 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 $\lambda_0 = 7.7 \times 10^{20}$ seconds^{-1} for the rare earth studies. They found good agreement in their calculations using a simple radius expression

$r = 1.48\ A^{1/3} \times 10^{-13}$ cm., 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 λ_0 value of 5.03×10^{18} seconds^{-1} was used. The simple radius formula

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

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

The fourth formula is that used recently by Kaplan[32] and derived by Preston.[31] The values tabulated in Table III were calculated by taking,

Table III

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

Nuclide	Alpha particle energy (Mev)	E_{recoil} (Mev)	E_{sc} (Mev)	E_{α} nuclear energy (Mev)	Partial alpha half-life (Exp)	Est Error limit factor	Calc. half-life exp. radius Cohen	Factor calc. $(T_{1/2})$ exp. $(T_{1/2})$	Calc. half-life one body Perlman-Ypsilantis	Factor calc. $(T_{1/2})$ exp. $(T_{1/2})$	Calc. half-life many body Bethe	Factor calc. $(T_{1/2})$ exp. $(T_{1/2})$	Calc. half-life one body Preston-Kaplan	Factor calc. $(T_{1/2})$ exp. $(T_{1/2})$
Sm^{147}	2.18	0.06	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	2.7×10^{13}y	190
Eu^{147}	2.88	0.08	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	4.8×10^{5}y	80
Gd^{148}	3.16	0.09	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	4.0×10^{3}y	29
Gd^{149}	3.0	0.08	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	0.07	0.02	2.8			8.8×10^{6}y		$4.0_8 \times 10^{6}$y					
Tb^{149}	3.95	0.11	0.02	4.07	>4.1h		9.2h		5.5h		7.0d		9.9d	
$Tb^{(151)}$	3.44	0.09	0.02	3.55	>19h		3.9y		1.95					
$Dy^{(151)}$	4.21	0.12	0.02	4.34	>8m		42m		57m					
$Dy^{(151)}$	4.06	0.11	0.02	4.19	>19m		5.2h		7.6h					
$Dy^{(151)}$	3.61	0.10	0.02	3.72	>2.3h		0.83y		1.15y					
$Au^{(188)}$	5.07	0.11	0.03	5.21	>4.3m (~30d)	?	17h		8.0h					
$Hg^{(189)}$	5.60	0.13	0.03	5.76	>0.7m		33m		3.6m					

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

as Kaplan has 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.

$$r = r_o{}^*A^{1/3}(1 + 0.8\ (A - 2Z)^{2A^{-2}} - 0.3A^{-1/3} + 0.010\ Z^2\ A^{-4/3})$$

with $r_o{}^* = 1.50 \times 10^{-13}$ cm, the value found by Kaplan[32] from 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 as

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

It is felt that the inclusion of both corrections is not proper for reasons set forth 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 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 a 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 that in the Perlman-Ypsilantis treatment, a radius formula without additive constant is adequate, but with formulas employing lower frequency factors, as do the others, 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.[42]) 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[41] 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.

These comparisons with rare earth alpha decay rate data do not rule out the validity of any of the four treatments. The comparisons suggest

that the use of a nuclear radius formula with an additive constant for the alpha particle is needed in all but the Perlman-Ypsilantis one body formula. Nothing can be concluded about the best value for the kinetic energy of the alpha particle within the nucleus, determined by the value assumed for the "internal potential" for the alpha particle.

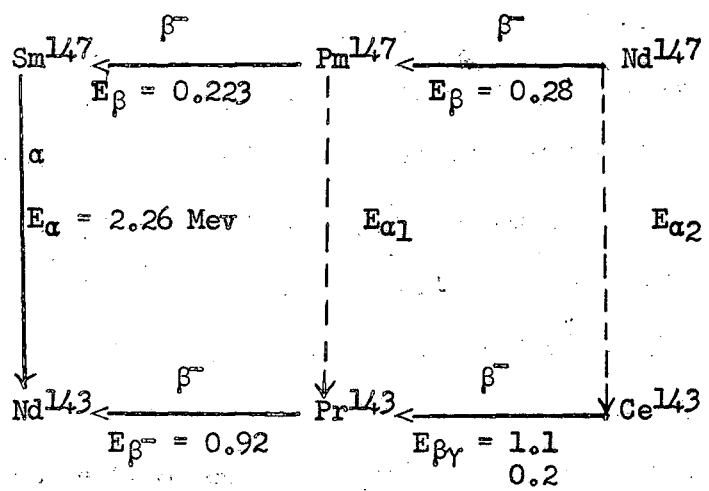
While the physical truth of the alpha decay process is probably represented by a many body theory, the many body correction to the frequency factor and the alpha particle radius correction to the effective nuclear radius nearly compensate each other with the result that the one body model still gives good agreement with experimental data. Present[39] points out in his discussion of the nuclear radius concept that the same compensation occurs in alpha decay treatments in the heavy element region.

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[42] 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[43] and by Rasmussen et al.,[25] and the alpha particle energy 2.18 Mev as determined in an argon-filled ionization chamber by Jesse and Sadauskis[16] is used.)



Conservation of energy requires that

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

$$E_{\alpha 2} = E_{\alpha 1} + 0.82 - 1.1 - 0.2 = 1.1 \text{ Mev}$$

(Beta decay energies from NBS table.[44]) 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.[45] 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.

Table V

Alpha Decay Energies of Rare Earth Nuclides

Nuclide	N	E_α (Exp.) Mev	"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	1.1 (calc)	-1.28	2.3
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.

It is evident from Figure 15 that a maximum in alpha decay energy should occur 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.

The normal trend for alpha decay energy to increase with decreasing neutron number is seen from the family of parallel lines in Figure 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 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 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[46] 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.[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[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[44] have been used, except for the positron decay energy of Pr^{140}, which was recently redetermined[48] 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 Figure 16, 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.

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	<u>9.5</u>	8.40	+1.1
Ba^{139}	56	83	<u>5.2</u>	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	<u>7.4</u>	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]

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 Figure 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. 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 with 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[46] 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.

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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.[12] 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.[49]

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.[50] 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 alphaactive 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[51] (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 or 87° C. The apparatus and techniques employed in the column separations at elevated temperatures have been described in detail in a recent article.[15] 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.[52]

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[52] 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.[53]

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.[25] 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 by two of the authors in collaboration with Reynolds[25] 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.

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[54] model ($W = 20.83\ Z^{7/3}$ ev[55]). 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[56] 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.

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

(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 present authors agree 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 equation (1). That the addition of both these corrections[57] amounts virtually to correcting twice for the same effect has been recently demonstrated in unpublished work of Serber and Snyder.

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.[58] 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 was estimated for mercury ($Z = 80$) using the relativistic expression of Racah[59] 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 (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 (1) is nearly equal to the seemingly unrelated 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 time of traversal of the electronic cloud by the alpha particle were very long compared to the mean time for rearrangement of the electronic wave functions, then presumably the electron wave functions would smoothly rearrange (adiabatically) 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 time for traversal of the electron cloud by the alpha particle is very short with respect to the time of rearrangement of the electron wave functions, 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 that that if the electron cloud was absent. The alpha decay energy of the nuclear process must then 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 of the above two approximations 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[60] 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.

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

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SECTION OF ISOTOPE CHART

ATOMIC No.	81	82	83	84	85	86	87	88	89	90
66				e Dy 7M α 4.21	e Dy 19M α 4.06	e Dy 2.3H α 3.61				Dy 156 0.052
65				a Tb 149 4.1H α 3.95		c Tb 151 150 19 H α 3.44		d Tb 153 5.1 D K, no β+ γ, e-	d Tb 154 17.2 H K, β+, β-(?) γ, e-	d Tb 155 190D K γ, e-
64				b Gd 148 long α 3.16	c Gd 149 9D K e- 35 α 3.0 (?)	e Gd 150 long α 2.7	c Gd 151 150 D K, no β+ γ 0.265 e- 0.220	Gd 152 0.20	c Gd 153 236 D K, no β+ γ 0.106 e-	Gd 154 2.15
63	e Eu 144 18 M β+ 2.5	c Eu 145 5D K, e- 0.2	e Eu 146 1.5D K, e- 0.4	b Eu 147 24D K, e- 0.2 α 2.88	b Eu 148 50 D K, e- 0.38 γ .6	Eu 149 >50D	c Eu 150 15H β+, e- β-	Eu 151 47.8	a Eu 152 9.2H K, β- γ / a 152 5.3γ K, β- γ	Eu 153 52.2
62		Sm 144 3.1	a Sm 145 410 D E.C. no γ		Sm 147 15.0 ~1.35×10^11 γ α 2.18	Sm 148 11.2	Sm 149 13.9	Sm 150 7.4	a Sm 151 ~1000γ β-, γ	Sm 152 26.8

NEUTRON No.

MU 2267A

Fig. 1

SECTION OF ISOTOPE CHART SHOWING THE RARE EARTH ALPHA EMITTERS.

MASS NUMBERS OF DYSPROSIUM ALPHA EMITTERS ARE NOT KNOWN.

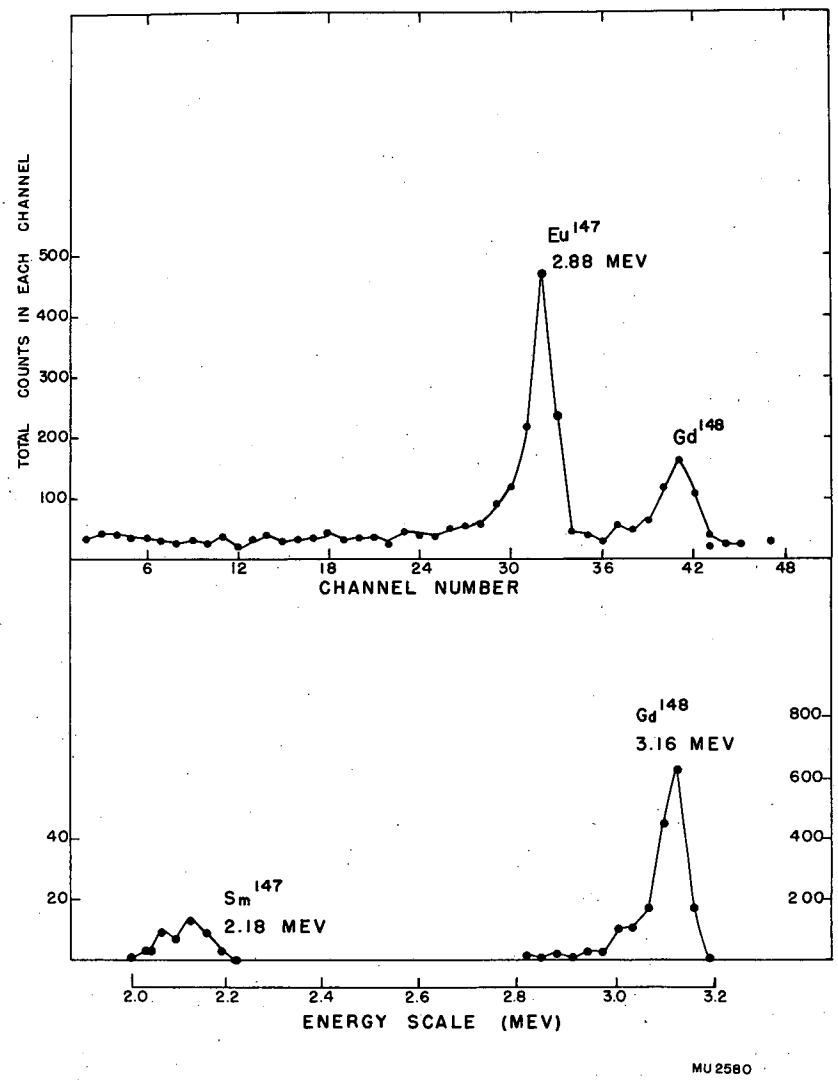

Fig. 2

ALPHA PULSE ANALYSIS CURVE OF EU^{147} AND COMPARISON CURVES OF SM^{147} AND GD^{148} STANDARDS.

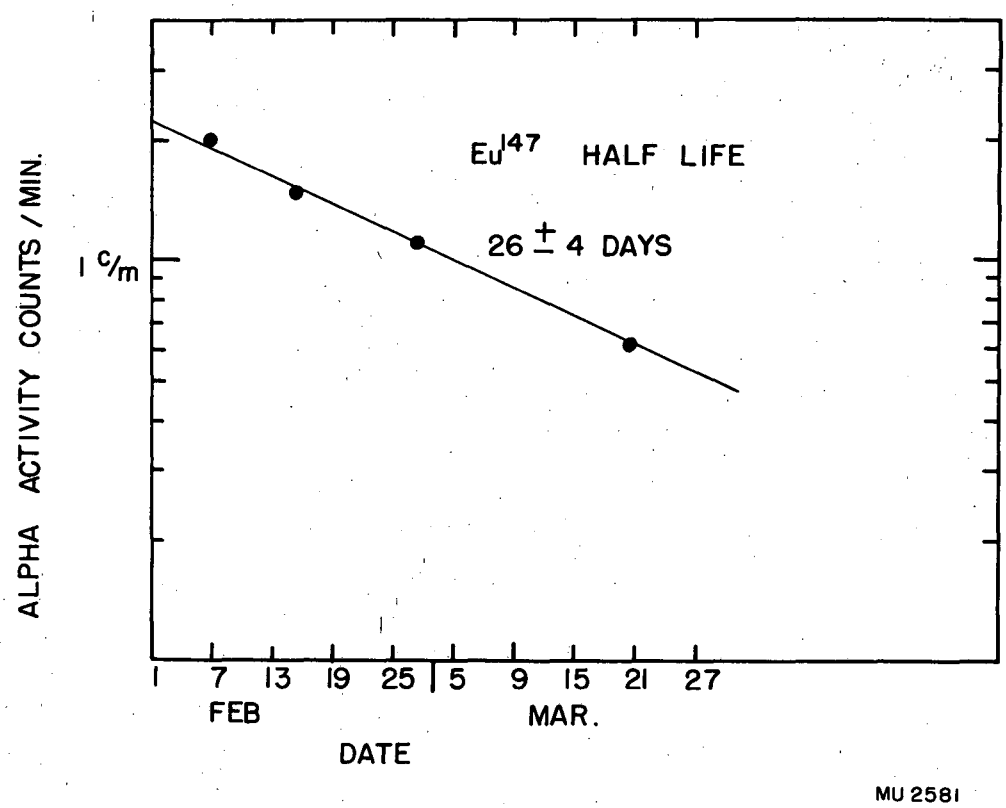

Fig. 3

DECAY CURVE OF EU^{147} ALPHA ACTIVITY FROM PULSE ANALYSIS COUNTING DATA. AN ESTIMATED BACKGROUND CORRECTION WAS MADE BEFORE PLOTTING.

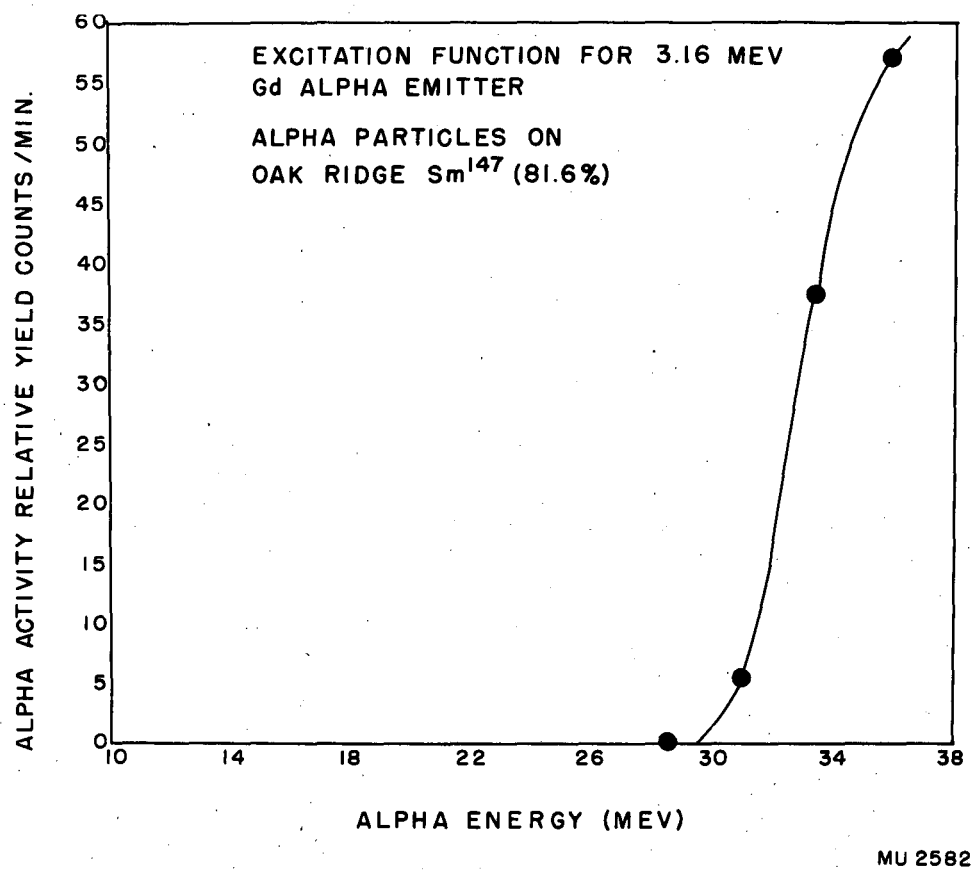

Fig. 4

YIELD OF 3.16 MEV GADOLINIUM ALPHA ACTIVITY VERSUS ENERGY OF ALPHA PARTICLES BOMBARDING SM^{147}.

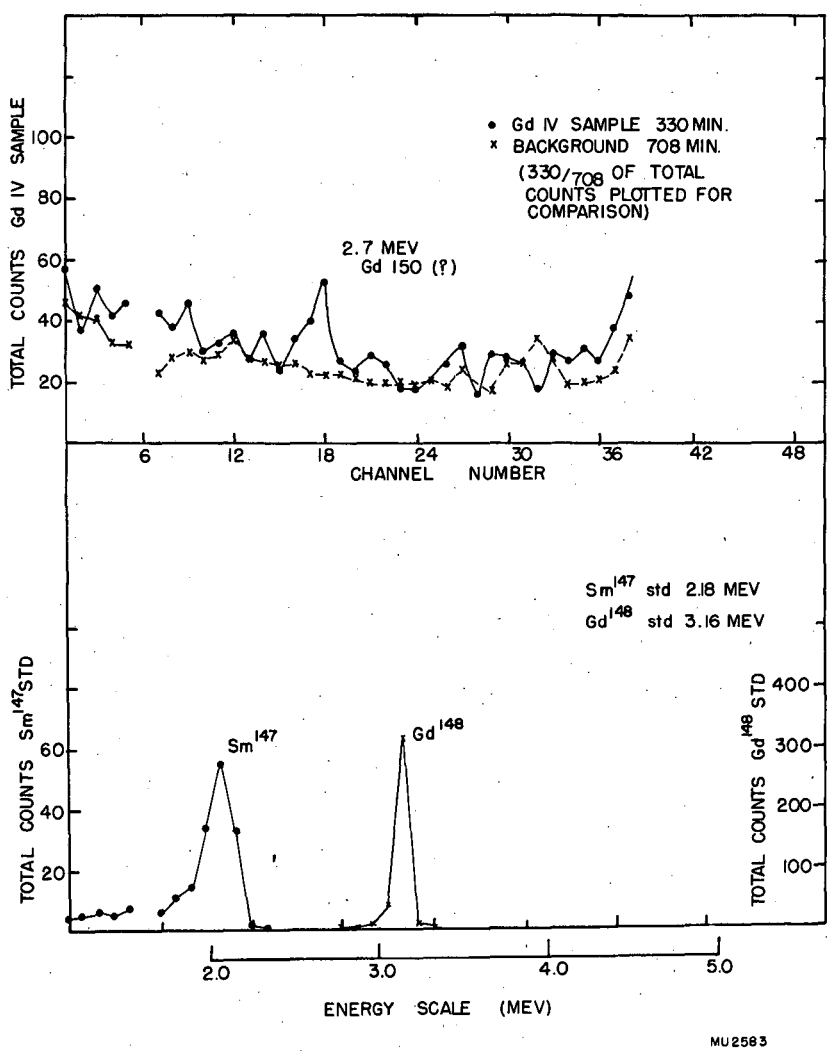

Fig. 5

ALPHA PULSE ANALYSIS CURVE OF A GADOLINIUM SAMPLE FROM 19 MEV DEUTRON 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.

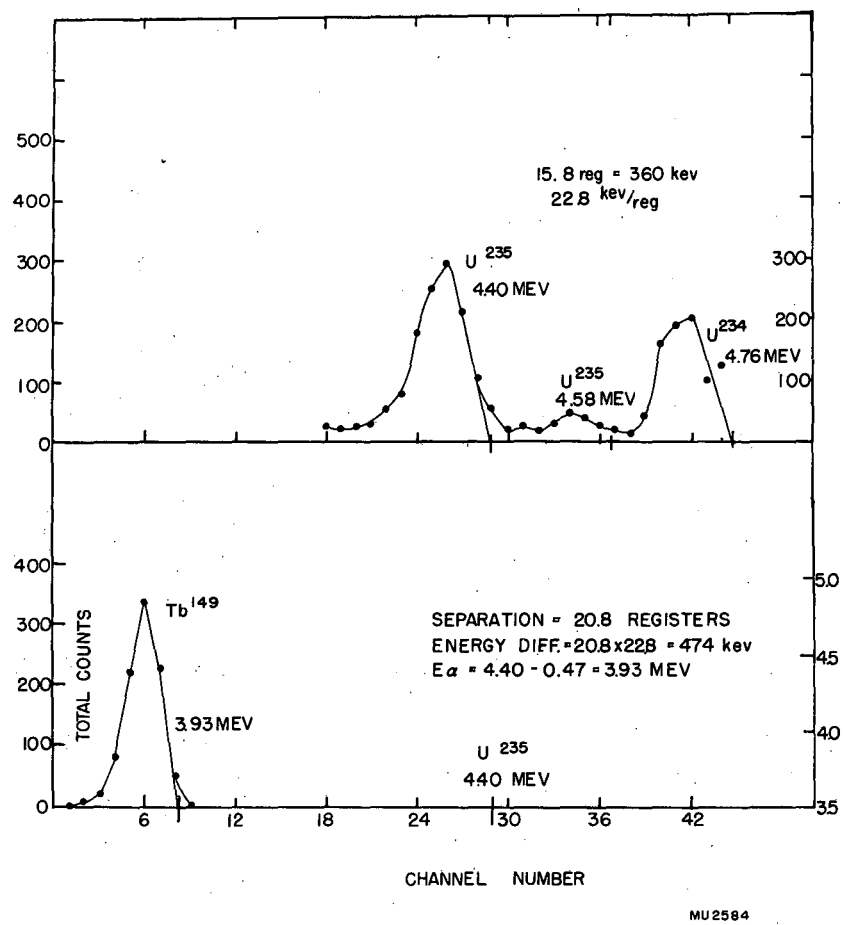

Fig. 6

PULSE ANALYSIS CURVES OF TB^{149} WITH COMPARISON ENERGY STANDARD U^{234} - U^{235}.

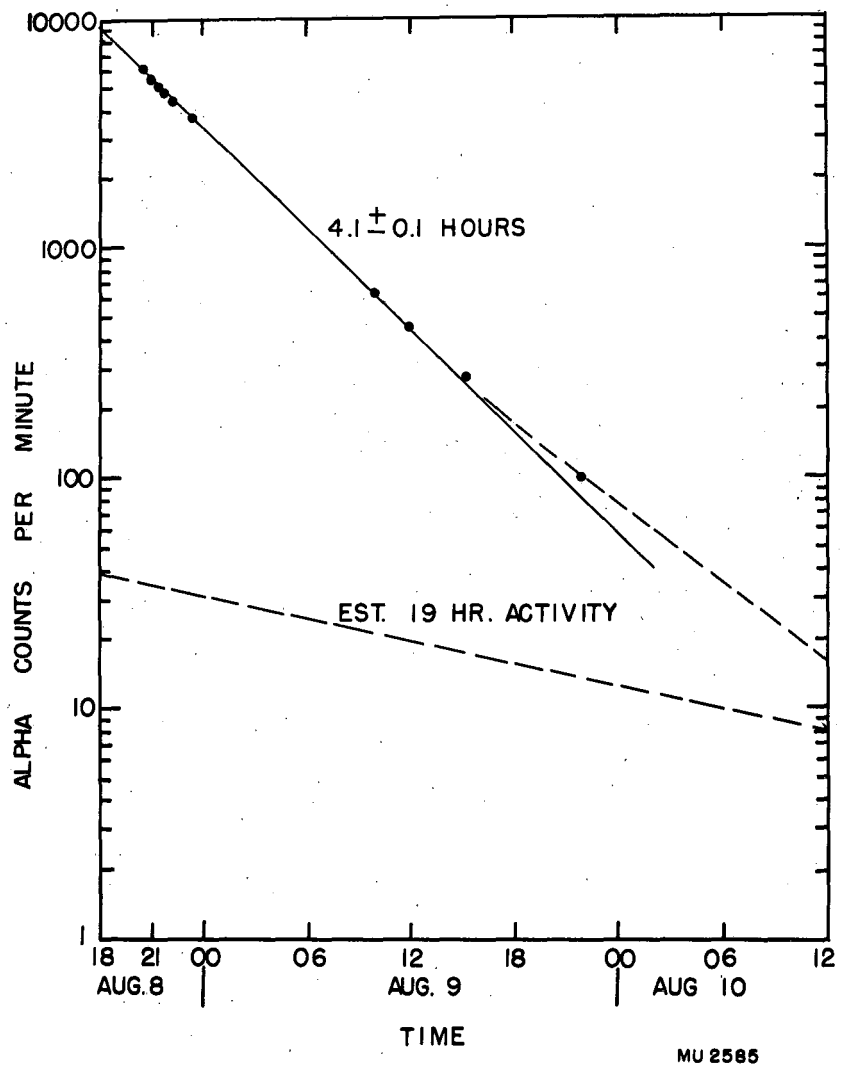

Fig. 7

DECAY CURVE OF GROSS TERBIUM ALPHA ACTIVITY IN ION CHAMBER ALPHA COUNTER.

(PREDOMINANT ACTIVITY IS TB^{149}.)

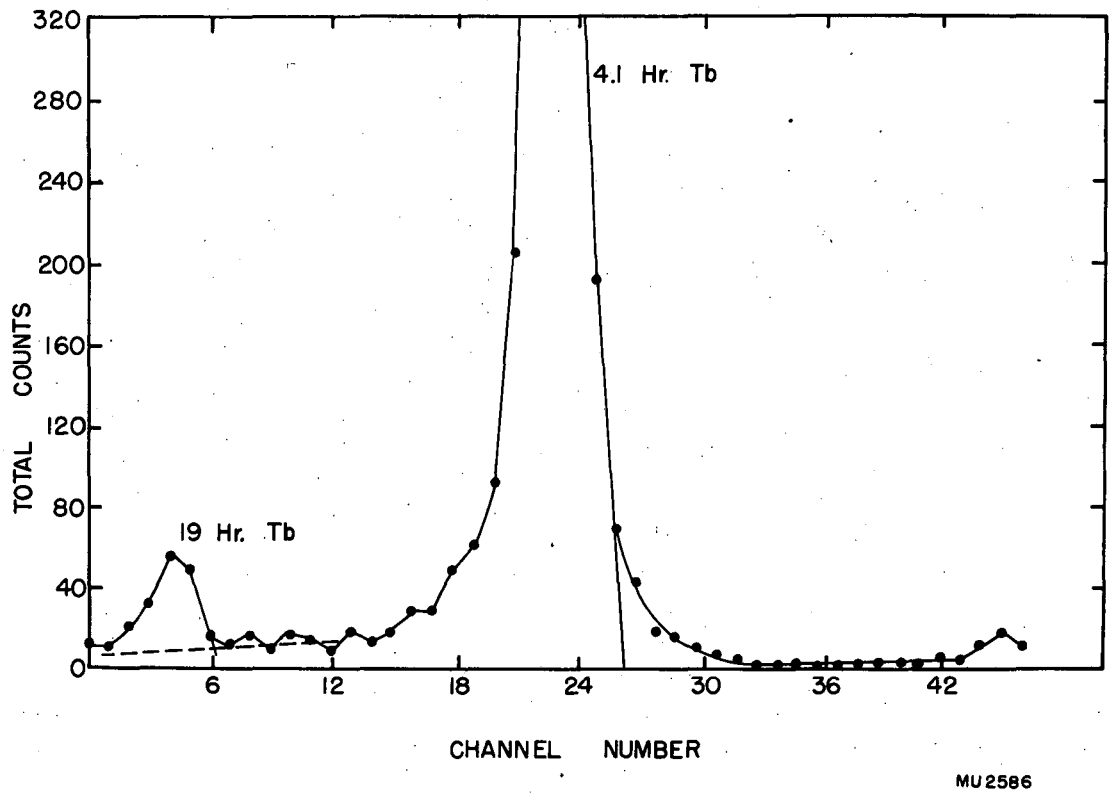

Fig. 8

ALPHA PULSE ANALYSIS CURVE OF TERBIUM ALPHA ACTIVITY SHOWING PEAKS CORRESPONDING TO THE 4.1 H AND 19 H ACTIVITIES

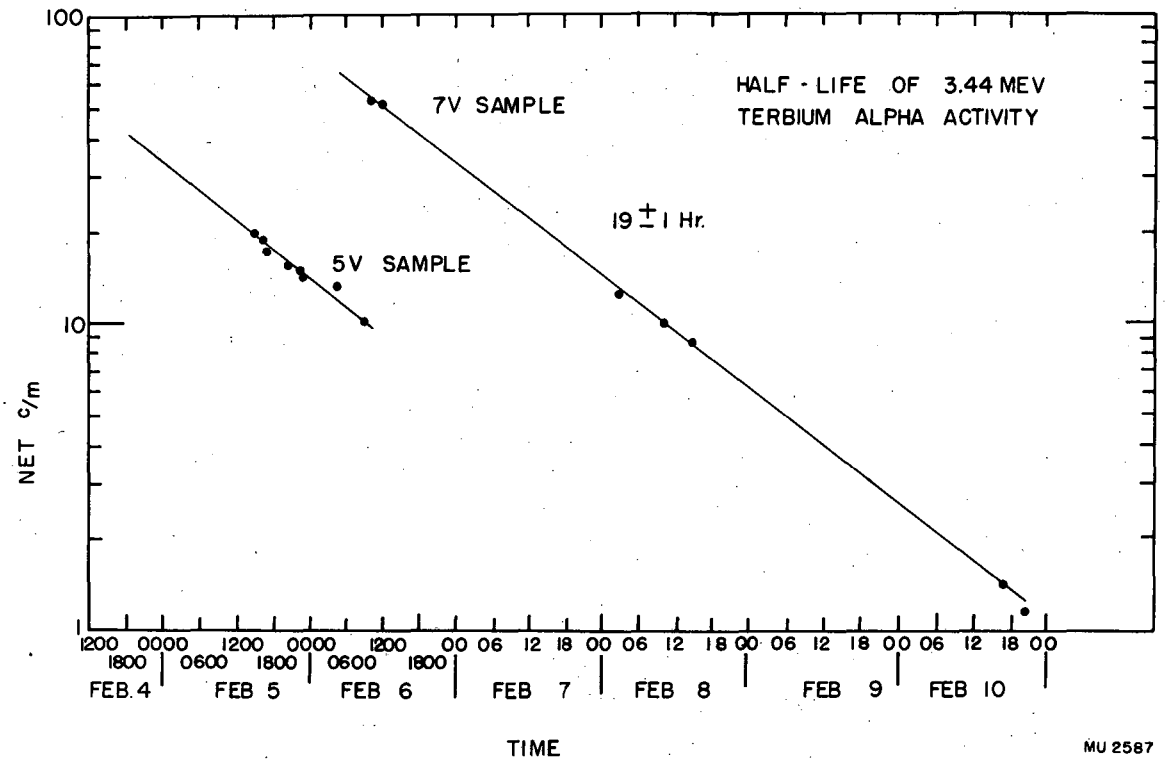

Fig. 9

DECAY CURVE OF TOTAL COUNTS IN THE 3.44 MEV TERBIUM ALPHA ACTIVITY PEAK ON PULSE ANALYSER COUNTS.

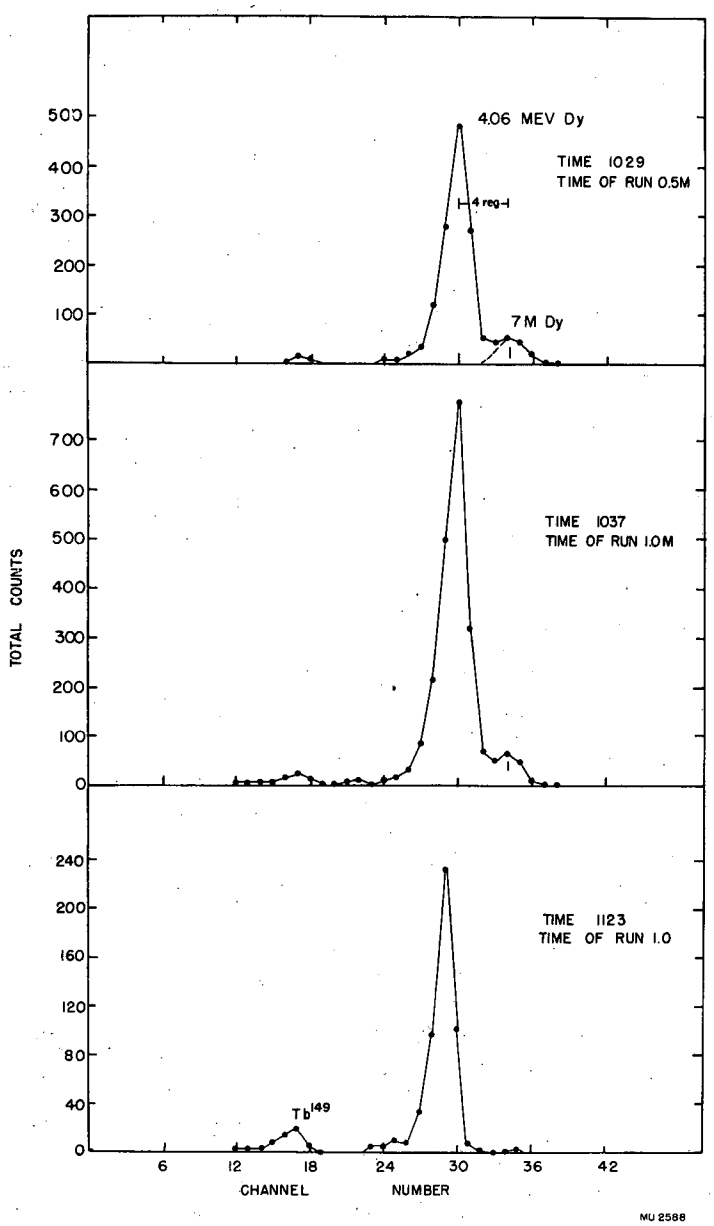

Fig. 10

ALPHA PULSE ANALYSIS CURVES OF A SAMPLE OF DYSPROSIUM ALPHA ACTIVITIES, SHOWING DECAY.

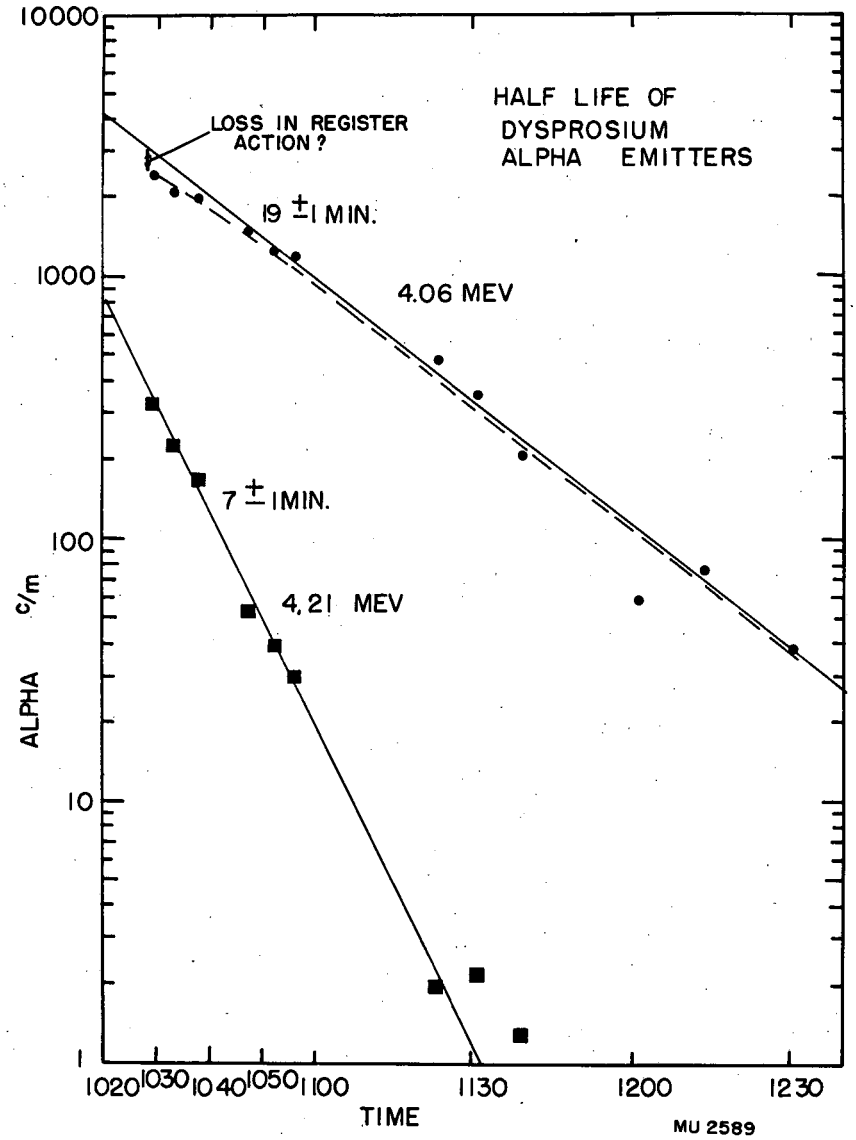

Fig. 11

DECAY CURVES OF DYSPROSIUM ALPHA EMITTERS

FROM PULSE ANALYZER COUNTS.

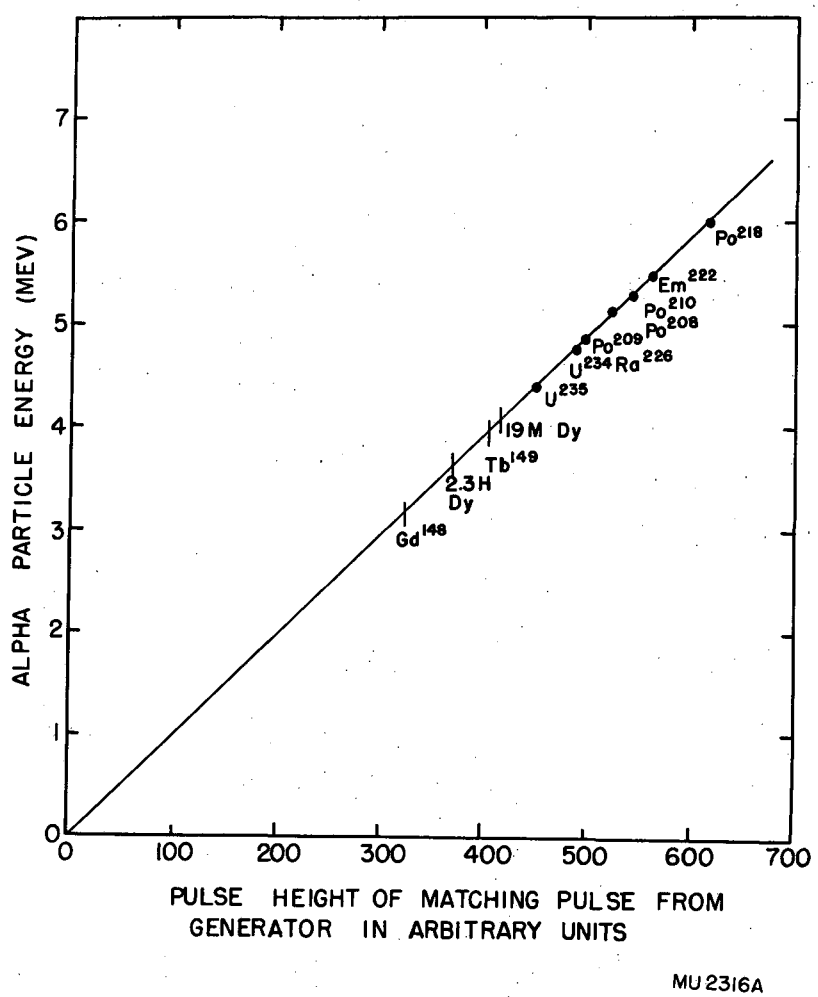

Fig. 12

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.

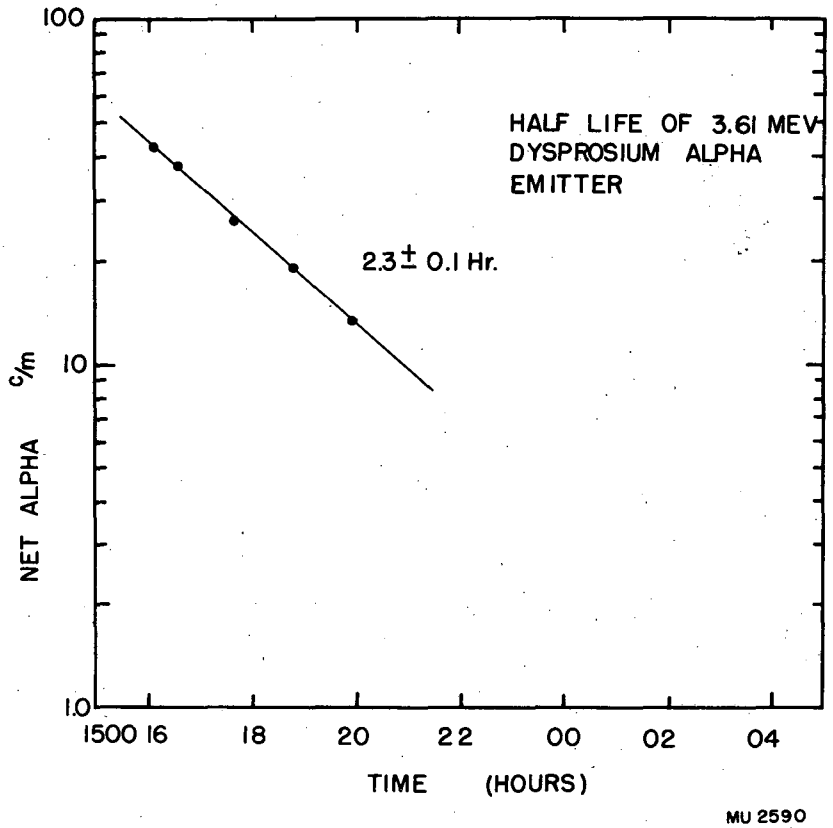

Fig. 13

DECAY CURVE OF 3.61 MEV DYSPROSIUM ALPHA ACTIVITY FROM PULSE ANALYZER COUNTS.

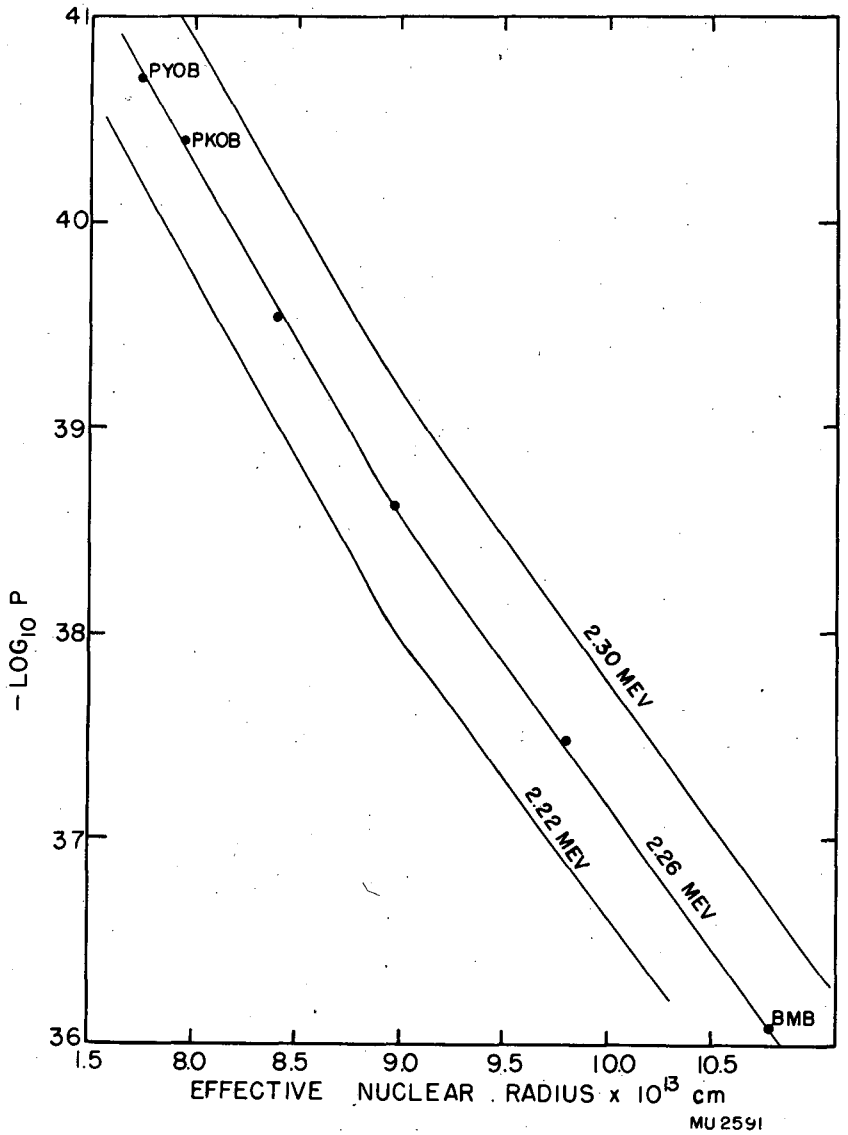

Fig. 14

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).

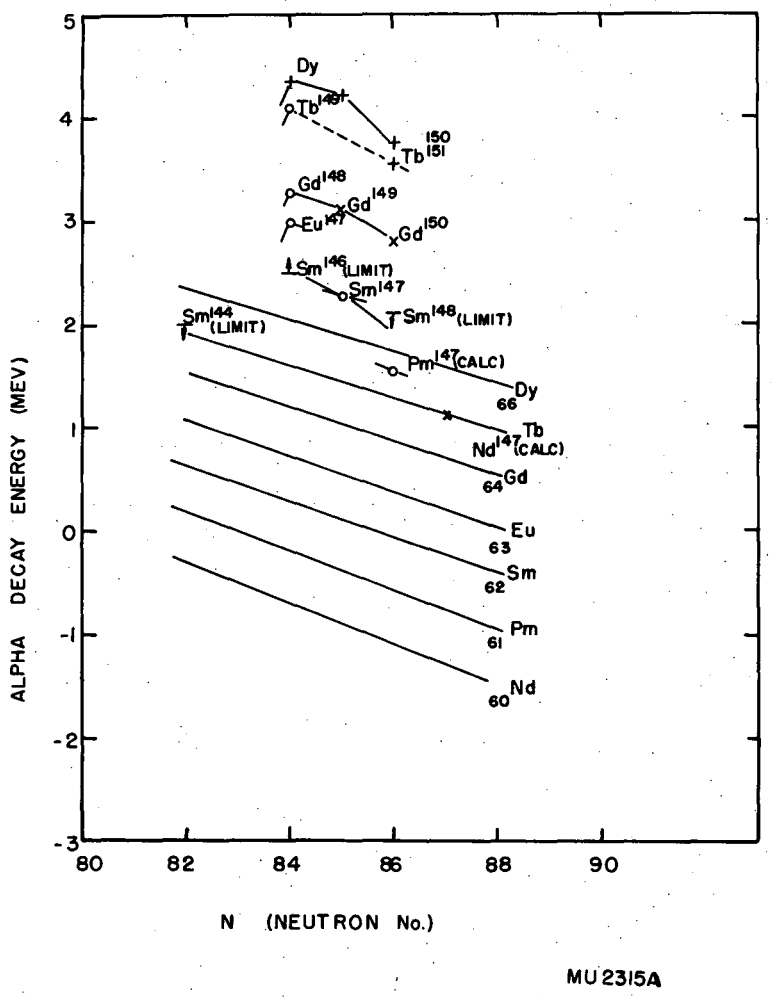

Fig. 15

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.

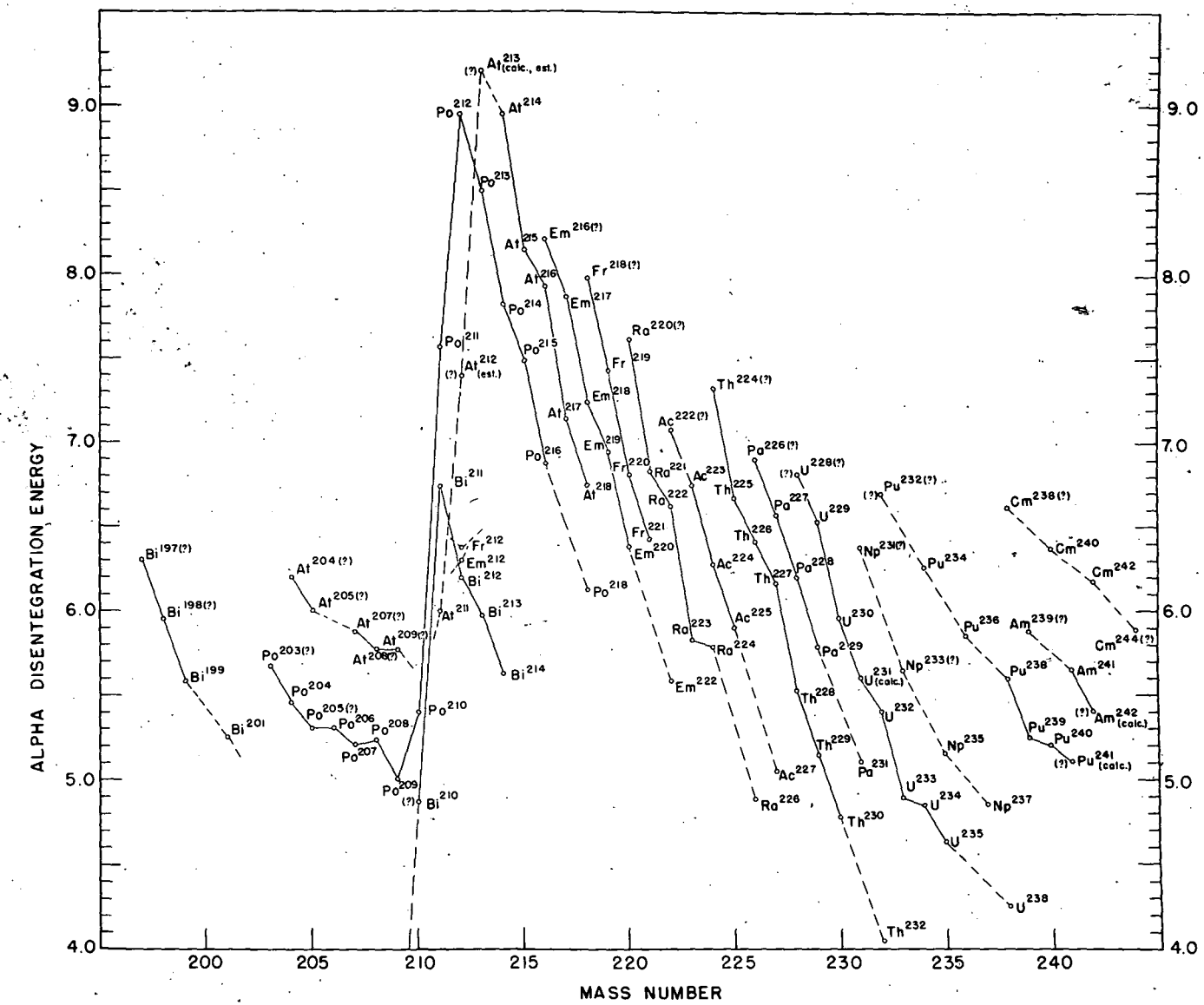

FIG. 16

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

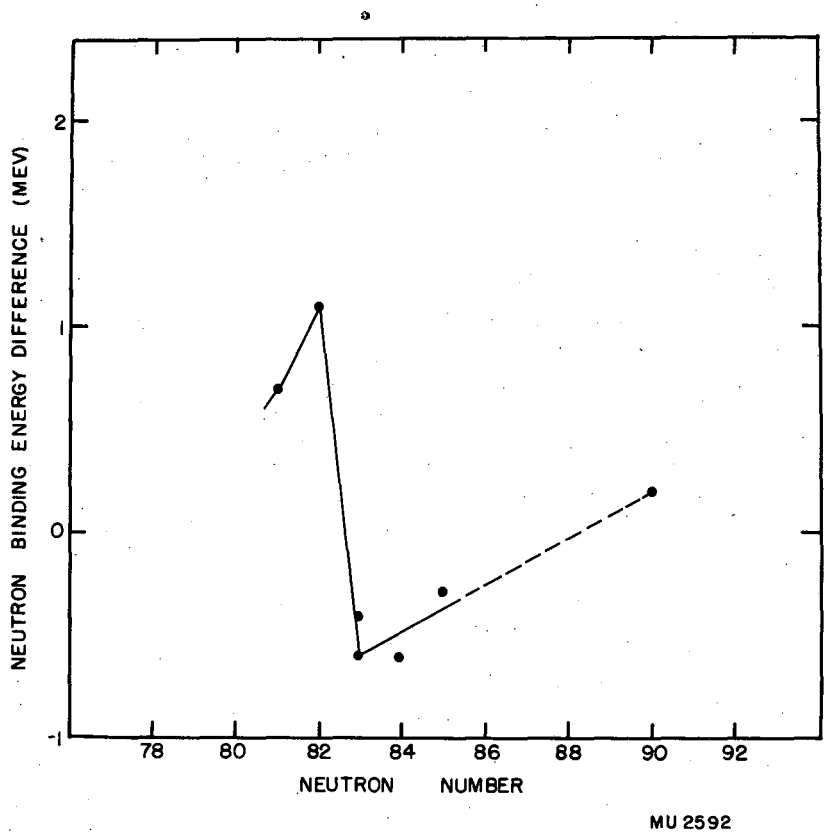

Fig. 17

NEUTRON BINDING ENERGY DIFFERENCES VS.
NEUTRON NUMBER NEAR 82.