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RADIOACTIVITIES PRODUCED IN EUROPIUM AND GADOLINIUM

by

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**A Dissertation Submitted to the
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INTRODUCTION

The study of radioactivities produced in gadolinium and europium was undertaken with two objectives in mind. The first objective was the determination of half-lives, energy of emitted radiations, mode of decay and decay schemes of gadolinium activities, and the assignment where possible of these activities to the proper isotope and element. The first aim is then fulfilled in that these tracers when catalogued may be used in rare earth separations as an identification of the gadolinium fraction.

Secondly, the present studies were designed to clear up some debated points in regard to europium activities. It was hoped that some assignment of radiations and half-lives could be made by the comparison of deuteron and neutron produced europium activities.

It should be pointed out at this time that primarily these studies deal with long-lived activities although two short-lived activities were investigated. As the samples were irradiated either at Argonne National Laboratory or Oak Ridge, half-lives of the order of a few hours or less could not be conveniently studied because of the time interval between the end of bombardment and the receiving of the sample.

When this investigation was started, the information regarding the neutron induced activities was somewhat meager. A long-lived neutron-induced gadolinium activity had been assigned to Gd^{153} on the basis of mass spectrographic measurements. A gadolinium activity produced in deuteron bombarded europium had also been tentatively assigned to Gd^{153} as this activity was long-lived. Neither the mode of decay nor energy of emitted particles of the long-lived neutron-induced gadolinium activity had been studied. This long-lived gadolinium activity would be useful as a tracer if its radiation characteristics were known. Various half-lives, energies and assignments had been reported for the short-lived gadolinium activities. The europium activities had been studied to some extent. It was known that both Eu^{152} and Eu^{154} were long-lived activities and their radiation spectra were complex. The short-lived 9.2 hour Eu^{152} was well known.

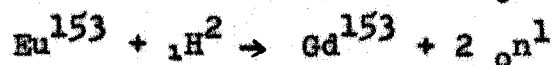
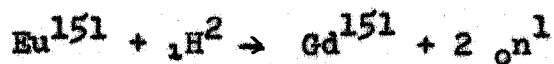
The problem thus involves the study of gadolinium activities, their half-lives, energies, mode of decay, decay schemes and assignments. Table I lists the stable isotopes of samarium, europium, gadolinium and terbium. In a slow neutron bombardment of gadolinium, the nuclides 153, 159, and 161 could be produced. Somewhat less likely is the production of an active isomer of a stable gadolinium

TABLE I

Stable Isotopes of Samarium,
Europium, Gadolinium and Terbium.

Tb				159
Gd		152	154-158	160
Eu		151	153	
Sm	144	147-150	152	154

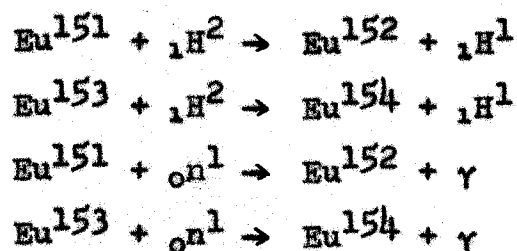
isotope. Fast neutron activation could give Gd^{151} by an (n, 2n) reaction on Gd^{152} . Deuteron bombarded europium could give the following gadolinium isotopes with the (d, 2n) processes the more probable for deuterons with the energy of those used, 16 Mev.



Gd^{152} and Gd^{154} would decay from the activated state to the ground state of the stable isotope by emission of one or more gamma rays. If conversion of a gamma ray occurred, the characteristic x-ray which would then be present would identify the activity as an isomer of a stable gadolinium isotope. Gd^{151} or Gd^{153} must decay to the corresponding stable europium isotope either by electron capture or

positron emission or a combination of these. Positrons are readily detected by magnetic deflection experiments so the mode of decay could be established. As a long-lived neutron induced gadolinium activity was assigned to Gd^{153} , a comparison of the long-lived gadolinium activity produced in europium is of some interest. If the activities were not similar, an assignment probably could be made on the basis of radiations emitted by the activity.

Because of the radiation complexities of the europium activities, the study was concerned more with a comparison of the activities produced by neutron and deuteron bombardment of europium. The europium activities are produced in the following way:



If in the neutron activation of europium, the process $Eu^{151}(n, \gamma) Eu^{152}$ had a large capture cross section and the $Eu^{153}(n, \gamma) Eu^{154}$ process a small probability, the Eu^{152} activity would of course be predominant. If also such a probability existed in the deuteron bombarded europium that Eu^{154} was the favored activity, comparison of these activities produced by the two different particles would be of value. In this idealized case it would then be possible

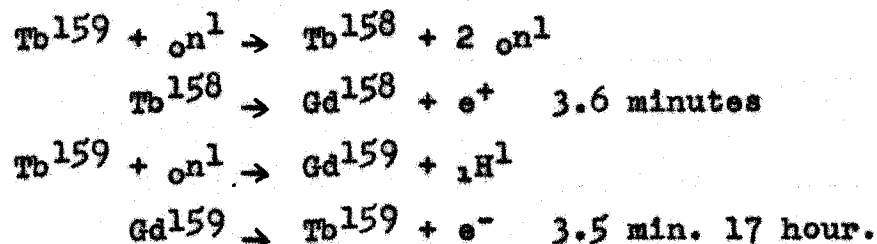
to study radiation, half-lives and decay schemes. However, mass assignments could not be made without some additional information. It was on this basis of comparison that the work on the europium activities was performed.

The bane of all radioactive studies is the possibility of active impurities. This is especially true in the rare earths which are difficult to separate. A rare earth contaminant in gadolinium may not be detected spectrographically but because of the possible large neutron capture cross section of this contaminant and some rare earths have unusually large cross sections, an activity which may be an appreciable percentage of the whole is introduced. Thus it is necessary to carefully purify the bombarded material so that extraneous activities do not distort the results. Perhaps the most convenient and best method for separating and purifying rare earths is the ion exchange resin technique. This method was employed frequently in the identification and separation of activities present in the neutron bombarded gadolinium. The final purity of the gadolinium activities is based on this method. The purification of europium is somewhat unique since advantage is taken of the divalent state to separate europium from most rare earths.

REVIEW OF LITERATURE

The presence of an 8 hour activity (1) in neutron bombarded gadolinium was first reported in 1935. A mixture of radium and beryllium provided the neutrons for the bombardment. In the same year Sugden (2) bombarded several rare earths which were purified by J. K. Marsh. Using neutrons from radon, in quantities up to 100 millicuries, sealed in small glass tubes with powdered beryllium no activity was produced in gadolinium. An intense 9.2 ± 0.1 hour europium activity was detected with a Geiger-Mueller counter. Also a 3.9 ± 0.1 hour activity was produced in terbium. These activities were reinvestigated with a stronger neutron source a short time later (3). Again no activity was detectable in the gadolinium. Also in 1935 another group of investigators (4) found a 6.4 ± 0.3 hour activity in gadolinium. In this experiment a 500 millicurie radium-beryllium neutron source bombarded 14 grams of gadolinium oxalate. The initial activity was 16 counts per minute. These early experiments lack conclusiveness because of low intensity of the activity and impurity of samples. The radium-beryllium sources provided a low neutron flux which was not capable of activating isotopes with a low capture cross section. Also the rapid separation of rare earth fractions after bombardment could not be accomplished so chemical identification of the activity was difficult.

After an interim of 2 or 3 years, the use of cyclotron techniques to obtain a high neutron flux revived the study of rare earth activities. Pool and Quill (5) did not detect an 8 hour activity in either fast or slow neutron irradiated gadolinium. The neutrons were produced by the lithium plus deuteron reaction. However, half-lives of 3.5 minutes and 17 hours were found in the gadolinium after both the fast and slow neutron bombardment. Both activities were assigned to Gd^{159} . This assignment was seemingly verified by fast neutron bombardment of Tb^{159} as activities with half-lives of 3.6 minutes and 17 hours were found. The reaction producing them would be the (n, p) process. The 3.6 minute activity decayed in part by emission of positrons. A slow neutron bombardment produced a 3.3 hour activity assigned to Tb^{160} . The following processes were involved according to the investigators.



Evaluation of this work is somewhat difficult as later investigators (6) could not produce the above activities by fast neutron bombardment of terbium. However, at present activities with half-lives of 3.6 minutes and 18 hours are assigned to Gd^{161} and Gd^{159} respectively (7).

The possibility of gadolinium being present as an impurity in the terbium may be the answer.

Krisberg, Pool and Hibdon (6) reported no activity was detectable in gadolinium irradiated with a 500 milli-curie radon-beryllium source. However, slow neutrons produced by $\text{Li} + {}_1\text{H}^2$ and $\text{Be} + {}_1\text{H}^2$ gave 5.5 day and 18 hour activities. Both of these activities were tentatively assigned to Tb^{161} . There was not a parent-daughter relationship between the two based on saturation intensity ratios. Assignment of these activities to Gd^{159} was considered unlikely since fast neutron bombardment of Tb^{159} did not produce either activity. That the assignment of these activities was still in doubt is evidenced by a paper by these same investigators (8) in which the 18 hour activity is assigned to Gd^{161} and the 5.5 day to Tb^{161} . The radiations emitted by these activities are as follows:

Gd^{161} or Tb^{161}	18 hour	0.85 Mev β	0.3 Mev γ
Tb^{161}	5.5 days	0.5 Mev β	1.28 Mev γ

These same investigators bombarded gadolinium with deuterons producing 5.5 day and 72 day activities. The 5.5 day activity has already been discussed and was thought to be produced by a $\text{Gd}^{160} (\text{d}, \text{n}) \text{Tb}^{161}$ reaction. The 72 day Tb^{160} activity is well known and is produced by a $(\text{d}, 2\text{n})$ reaction on Gd^{160} . It should be noted that no long-lived neutron or deuteron induced gadolinium activities were reported.

Uranium reactors as neutron sources had entered the field by this time. After a long neutron bombardment in the uranium pile at Hanford, Washington, a sample of Sm_2O_3 on mass spectrographic analysis exhibited the activities listed in Table II (9). Europium, gadolinium and terbium were present in the samarium as impurities. The half-lives were estimated by comparing intensity of lines produced on photographic plates with an activity of a known half-life. The half-life of greater than 72 days for Gd^{153} was obtained by comparison with the 72 day Tb^{160} .

TABLE II

Neutron Induced Activities in Impure Sm_2O_3 .

Mass No.	Ion	Active Isotope	Half-life
145	Sm^{145+}	Sm^{145}	> 72 days
151	Sm^{151+}	Sm^{151}	~20 years
152	Eu^{152+}	Eu^{152}	~5 years
154	Eu^{154+}	Eu^{154}	~5 years
155	Eu^{155+}	Eu^{155}	2.3 years
156	Eu^{156+}	Eu^{156}	15.4 days
161	$\text{Sm}^{145}\text{O}^+$	Sm^{145}	> 72 days
167	$\text{Sm}^{151}\text{O}^+$	Sm^{151}	~20 years
169	$\text{Gd}^{153}\text{O}^+$	Gd^{153}	> 72 days
176	$\text{Tb}^{160}\text{O}^+$	Tb^{160}	72 days

Another extensive review of activities produced in pile neutron irradiated rare earths was reported a short time later (10). Oxides of lanthanum, samarium, europium, gadolinium and terbium were irradiated in the Oak Ridge pile for two months. The conclusions concerning activities in Gd_2O_3 are clouded because europium and terbium were also present and these activities were not separated from the gadolinium. However, a gamma ray of 102 Kev as measured with a magnetic spectrometer was assigned to Gd^{153} . This activity was given a rough half-life of 110 days.

After the investigation of gadolinium and europium activities had been started in this laboratory two other papers regarding bombardment of gadolinium with pile neutrons were reported. The first paper dealt with short-lived activities produced in Gd_2O_3 in the pile at Harwell, England (7). Table III gives the pertinent information from this report. The similarity between capture cross section for the 218 sec. and the 6.75 day activities suggested a parent-daughter relationship and assignment to Gd^{161} and Tb^{161} . Likewise the cross section value for the 18 hour activity placed this nuclide at Gd^{159} rather than Gd^{161} . Deuteron and neutron bombarded gadolinium did not give long-lived activities. The results discussed in the second paper (11) are in part very similar to those reported in this thesis. A comparison of results will be given later.

TABLE III

Neutron Induced Activities in Gd_2O_3 .

Half-life	Element	*	β	γ
218 ± 5 sec.	--	0.18	--	--
18.0 ± 0.2 hr.	Gd	1.1	0.95 Mev	0.055, 0.38 Mev
6.75 ± 0.1 d.	Tb	0.16	0.52 Mev	0.05 (no harder γ)
* barns (10^{-24} cm ²) in natural gadolinium				

The production of an activity in deutron bombarded europium which was chemically different from europium was first observed by Fajans and Voigt (12). The investigation was concerned with short half-lives produced in the deutron bombarded europium. In the course of chemical purification an activity present after irradiation could be separated from europium as it was not reduced or precipitated as a sulfate. The activity did precipitate with $Eu_2(C_2O_4)_3$ in acid solution which is a characteristic reaction of rare earths. The intensity was such that it could not be an impurity. It was further determined that the activity decayed with a half-life of 155-170 days and the emission of negative beta rays and x-rays or gamma rays.

Krisberg, Pool and Hibdon (6) bombarded Eu_2O_3 but made no chemical separation of the activities. Three components were present in the decay curve of the active

sample with half-lives listed as 9.2 hours, a minimum of 75 days, and a minimum of 20 years. The 75 day activity was tentatively assigned to Gd^{153} because the 75 days was longer than would be expected for Gd^{151} . The same investigators (8) in another paper assign a half-life of 62 days to the activity.

The assignment of this long-lived gadolinium to Gd^{153} seemed to be corroborated by mass spectrographic work (9). A neutron induced gadolinium activity with a half-life greater than 72 days was assigned to Gd^{153} (see Table II). Because of the similarity in half-lives it was assumed that Gd^{153} was the long-lived gadolinium activity (13) produced by a (d, 2n) reaction on Eu^{153} . The production of Gd^{151} by a similar process on Eu^{151} would be possible.

Since this study is not concerned with the well known 9.2 hour Eu^{152} , the history of that activity will not be reviewed. The first reference to a long-lived europium activity (1-2 years) produced by slow neutron irradiation was by Scheichenberger (14). The early history of the long-lived europium activity is reviewed by Fajans and Voigt (12) who estimated its half-life as 5 - 8 years, checked its production by n, γ reaction and produced it by (d, p) reaction as well. Other workers measured the decay of deuteron bombarded europium over a period of 4 years and assigned a minimum half-life of 20 years (6).

Mass spectrographic analysis on pile irradiated Eu_2O_3 indicated long-lived activities formed at masses 152 and 154 with approximately equal half-lives (15). Magnetic investigation showed no detectable positron emission. One or both of the activities decay to an unknown extent by K-electron capture (16). In a later paper (9), the half-lives were estimated at approximately 5 years by comparison of line intensities on photographic plates. Table II gives some relevant information in regard to this measurement. Later the same investigators (17) with a refined method involving a mass spectrometer and an isotopic dilution technique reported half-lives of 5.3 and 5.4 years for Eu^{152} and Eu^{154} respectively. These results were believed to be good within 15 percent.

A number of measurements of the gamma ray energies have been made (18, 19, 20, 21). One of these groups of investigators (21) states that about 33 electron lines are detected with a photographic magnetic spectrometer in the decay of the europium activities. A part of a decay scheme was presented but the authors state a satisfactory scheme will have to wait until enriched isotopes of masses 151 and 153 are available.

EXPERIMENTAL INVESTIGATIONS

Statement of Problem

The purposes of this study, as given in the introduction, are restated at this point to give added meaning to the following experimental work. The first objective was to determine the radiation characteristics of gadolinium activities in order to increase our knowledge about nuclei in general and so that these activities could be used in rare earth separations as an identification of the gadolinium fraction. To accomplish this objective, the half-lives, energies of emitted radiations, mode of decay and decay schemes were studied. The second aim was to determine whether a comparison of deuteron and neutron bombarded europium activities would aid in the assignment of half-lives and radiation to Eu^{152} and Eu^{154} .

Instruments Employed

Beta and Gamma Counters

A detailed description will not be given of instruments in common use. However, circuits specially designed will be described in some detail. Models 161 and 165 sealing units manufactured by Nuclear Instrument and Chemical Corporation were used for most of the counting work.

These scale of 64 counters have the Higinbotham trigger circuits which are quite reliable. The resolving time of the circuit is less than five microseconds. A stabilized high voltage supply capable of generating from 600 to 1500 volts is part of the unit. A constant voltage generator in conjunction with the built-in power supply gave a well stabilized voltage.

Geiger-Mueller counter tubes made by Victoreen Instrument Co., Radiation Counter Laboratories and Tracer Lab were used to count beta and gamma radiations. The tubes were all of the self-quenching type. Counter tubes were selected that had less than 5 percent increase in slope per 100 volts and with a threshold of 1000 - 1200 volts so that a suitable plateau could be reached within the limits of the power supply. The counter tubes were mounted in lead housings to minimize background counts and care was taken to exclude light when the tubes were at operating potential. The absorption curves, short half-life measurements and quantitative work were done with the G-M tubes and scalers. As usual background and standard sample readings were taken each time an experimental sample was counted.

Electroscope

All of the long half-life measurements were taken on a Lauritsen electroscope because of its reliability over long periods of time. A standard sample read with this instrument over a period of 7 years has given identical readings. The instrument employed was purchased from Fred C. Henson Co. The ionization chamber was changed to one of cast aluminum 8 cm. on an edge. It was equipped with two glass receptacles which were filled with a dehydrating agent to keep the air inside the chamber dry. All of the instruments were kept in a constant temperature room. The case was designed so that measurements could be made from either the top or bottom of the ionization chamber. A detailed description of such a chamber has been published (22). The same part of the scale (20 to 40 divisions) was used for all measurements.

Survey Meters

A variety of survey meters were used during the investigation. These instruments were of value in checking the progress and separation of rare earth activities on ion exchange columns as well as checking glassware, clothing, etc. Low energy beta particles were generally not detected because of the window thickness of the probe so this factor was taken into account when testing activities.

Coincidence Circuit

A coincidence circuit was built by the electronics shop of this laboratory. The circuit was similar to one described by Moak (23) although some modifications were incorporated. Diagrams of the electronic circuit are given in Figures 1 and 2. The circuit was designed with two high voltage power supplies so that matched counter tubes were not necessary. Conventional scalers were employed to count the pulses from either channel. Pulses through both channels as well as the coincidence count were recorded simultaneously. The coincidence pulses were unscaled and recorded directly by a Mercury recorder. With a high coincidence rate the loss in this unscaled circuit would be considerable; however, the highest coincidence counting rate in any of this work was 10 c/m at which rate the loss is less than 1%.

The circuit was designed with four resolving times of the order of 0.5, 1.0, 1.5 and 2.0 microseconds. These resolving times were then experimentally determined by the method of independent samples and found to be 0.543, 1.05, 1.42 and 2.06 microseconds. In this method of determining resolving times, the two counter tubes were widely separated so as to minimize the chance of one particle passing through both counters. Samples were then counted on the two channels and the number of accidental coinci-

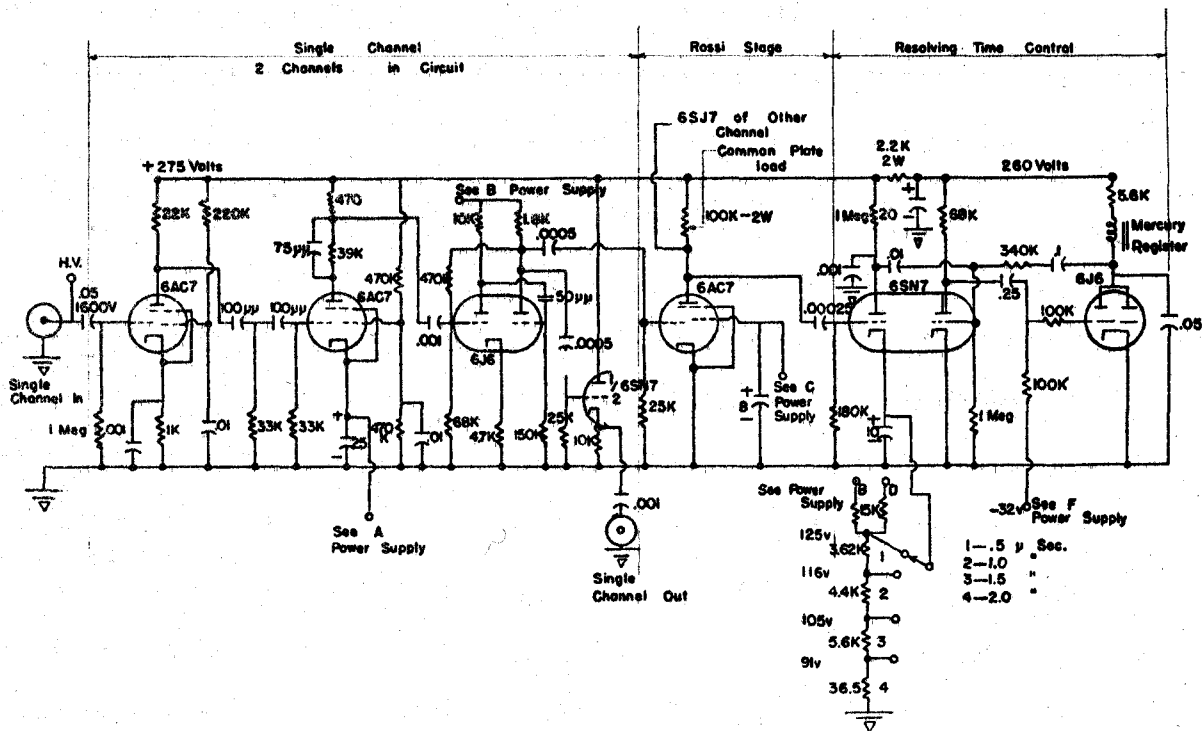


Fig. 1. Coincidence Circuit with One Channel, Rossi Stage and Resolving Time Control.



dences determined. The resolving time T is related to the accidental coincidences by the following equation (24):

$$G_{acc/min} = KT N_1 N_2$$

where N_1 and N_2 are the counts per minute recorded in channels 1 and 2. K is a constant very often taken as two, but since KT is measured experimentally the value of K is not important. It is evident that any coincidence experiment must be corrected for these accidental coincidences. The resolving times were determined over a six months period and no variation greater than was reasonable from statistics was observed. In the coincidence studies on gadolinium activities, the resolving time setting was kept at 2.06 microseconds.

The coincidence mount consists of an aluminum box with the counter tubes, facing one another, inset in the aluminum sheet with a distance of 2.7 cm separating the counter windows. The aluminum box is 3.1 cm wide, 8.4 cm high and 8.9 cm long. A slot for insertion of sample plates with the active sample is centered between the counter tubes. Foils can be placed between either counter and the sample. The aluminum box and counter assembly is mounted on a larger aluminum base and the whole assembly is covered to exclude light.

A sample of Co^{60} was used to determine the γ -ray efficiency of the counters. This activity decays by the

emission of a 0.31 Mev β followed by γ -rays of 1.1 and 1.3 Mev in cascade (25). The γ -ray efficiency is determined in the following fashion (26):

$$\begin{aligned}C_{\beta} &= C_0 F(x) W_{\beta} \\C_{\gamma} &= C_0 W_{\gamma} (E_1 + E_2) \\C_{\beta\gamma} &= C_0 F(x) W_{\beta} W_{\gamma} (E_1 + E_2) \\C_{\beta\gamma}/C_{\beta} &= W_{\gamma} (E_1 + E_2)\end{aligned}$$

C_0 is the true disintegration rate and the solid angle subtended by the β and γ counters are given by W_{β} and W_{γ} . $F(x)$ represents the fraction of β -particles passing through a thickness x of absorber. E_1 and E_2 represent the efficiency of the γ counters for the 1.1 Mev and 1.3 Mev γ -rays.

Experimentally $\beta\gamma$ + $\gamma\gamma$ coincidences were determined with a Co^{60} source. Both of these values were corrected for accidental and background coincidences. The corrected $\beta\gamma$ value was then obtained from these data. The pertinent information is given in Table IV. The value represents the total efficiency including solid angle for both γ -rays.

TABLE IV

Determination of γ -ray Efficiency

$(\beta\gamma + \gamma\gamma)/\text{min}$	$\gamma\gamma/\text{min}$	$\beta\gamma$ corrected	C_{β}/min	$C_{\beta\gamma}/C_{\beta}$
10.65	0.397	10.14	4125	2.45×10^{-3}

Rate Meter Circuit

The combination of a small ion exchange column, eluate counting cell, counter tube and recording rate meter was used in the purification of activities*. The small column 11 cm long and 0.1 cm I.D. packed with Dowex-50 resin was so designed that eluting agents could be forced through the column. Free flow through the finely divided resin was much slower than the optimum flow rate of 0.04 ml/min desired. The eluate then entered a lead housing containing the counting cell and counter. The counting cell was designed to present a maximum volume of active solution to the counter with a minimum of self absorption. Drawings of the assembly are on file in laboratory reports of D. Christian. Similar experimental arrangements have been reported (27).

The electronic circuit has six different scales of 200, 500, 1000, 2000, 5000 and 10,000 c/m so that a wide intensity of samples could be observed. An Esterline-Angus automatic recorder graphed activity per unit volume versus time as the elution of an activity progressed. This graph was of value in making volume cuts of the eluate at the proper time.

* Constructed by D. S. Martin and Darleane Christian of this laboratory.

Materials

The gadolinium oxide bombarded in this investigation was loaned by the rare earth group of this laboratory. Purification of the gadolinium had been effected by large scale ion exchange fractionations (28). After recycling of the gadolinium fraction a sample of high purity was obtained. Early spectrographic analysis indicated 0.28% samarium and a trace of terbium. Later analyses also listed dysprosium, holmium, yttrium and ytterbium as trace contaminants. The purity of the sample will be discussed again in connection with the activities produced in the neutron irradiated gadolinium.

Europium oxide, originally purified by H. N. McCoy and loaned for this investigation by F. H. Spedding, on spectrographic analysis indicated traces of samarium and gadolinium were present. The percentage of either of these contaminants could not be estimated. Before the Eu_2O_3 was bombarded with neutrons an extensive repurification of the europium was undertaken which involved precipitations of europous sulfate, europic fluoride and oxalate. However, spectrographic analysis still indicated a trace of samarium and gadolinium following this procedure. After bombardment of the Eu_2O_3 with neutrons and deuterons, separations designed to detect any active contaminants failed to do so.

Nalcite high capacity resin and Dowex-50 were used as cation exchangers in this investigation. These resins are the same chemically as both are a sulfonated polystyrene-divinyl-benzene co-polymer whose nuclear sulfonic acid groups are the sole ion-active groups at any pH value. Generally the resins were treated with saturated solutions of ammonium chloride so that the exchange reactions took place between the ammonium form of the resin and the rare earth cation.

Gadolinium Produced by
Deuteron Bombardment
of Europium

Survey of Previous Work

A brief recapitulation of previous investigation is given before the experimental results are discussed. Fajans and Voigt (12) chemically separated the gadolinium and europium activities produced in deuteron bombarded europium and assigned a half-life of 155 - 170 days to the gadolinium activity. Another group of investigators (6) reported an activity with a half-life of about 75 days in deuteron bombarded europium but no chemical separations were effected. Mass spectrographic work (9) showed that a half-life greater than 72 days could be produced by neutron bombardment of gadolinium, the mass number of the

active isotope being ^{153}Gd . Based on the similarity of half-lives, it was assumed that the 155 - 170 day activity was ^{153}Gd (13) being produced both by (d, 2n) reaction on ^{153}Eu and (n, γ) on ^{153}Gd .

Separation of Europium and Gadolinium Activities

A sample of Eu_2O_3 (130 mg) was given 870 microampere hours of bombardment with 20 Mev deuterons by the Berkeley 60-inch cyclotron. The cyclotron target was prepared by filling a small platinum dish with the powdered Eu_2O_3 . The assembly was raised to 800°C and a pressure of four tons applied. The target was then allowed to cool under pressure. A thin tantalum foil was placed over the material to protect it during bombardment. During the bombardment, the assembly was cooled by water circulating through a copper coil attached to the target.

Since these other materials had been bombarded with the Eu_2O_3 , a chemical separation of rare earth activities from other target activities was performed. The target and the foil were leached several times with hydrochloric acid to effect solution of the Eu_2O_3 . The acid solutions were concentrated and copper present in the solutions was deposited electrolytically. The copper was inactive but ^{65}Zn was present as a result of the (d, 2n) reaction. The europium was precipitated as the oxalate and then

ignited to the oxide. The oxide was redissolved and EuF_3 precipitated to purify further from tantalum and other fluoride soluble contaminants. The rare earth fluoride was then converted back to the oxide via the oxalate. The recovery on this portion was 97.3 mg.

The resulting pure rare earth fraction was separated into europium and gadolinium by precipitation of europous sulfate. The McCoy method of reduction with minor modifications as given in Inorganic Synthesis Volume II (29) was followed. After preparation of the Jones reductor, the active rare earth fraction 0.1 M with respect to Eu^{+3} and HCl was poured through the reductor at a slow rate. Air was carefully excluded by sweeping nitrogen through the filter flask.

A feathery white precipitate of EuSO_4 was formed first in the 8 M H_2SO_4 in the receiving flask. After heating the solution and precipitate under nitrogen to 80°C , the α form of EuSO_4 changed to a more stable β form which is dense and crystalline settling to a compact mass. The β form unlike the α is only slightly soluble in H_2SO_4 . The EuSO_4 was then filtered and air dried at 75°C . As will be discussed later, this EuSO_4 fraction was further purified for radiation and half-life studies of the europium activities.

After the bulk of the active europium had been separated in the above manner, the filtrates containing the active gadolinium were combined and reduced in volume. The remainder of the europium (also any samarium or ytterbium present as impurities) was extracted from this gadolinium rich fraction with sodium amalgam. Inactive europium carrier was added in later extractions to ensure the removal of traces of active europium. The extent of the separation of activities was determined by means of aluminum absorption curves of the activity in both fractions. As the radiation energies of the gadolinium and europium fraction were quite different, aluminum absorption curves give a sensitive method of analysis as indicated in Figure 3. When similar absorption curves were obtained for both fractions, the separation was considered complete. This method of analysis was reported earlier in separation of antimony and titanium in this laboratory (30). Some gadolinium was occluded in the mercury phase in each extraction, so the analysis method mentioned above could be applied. As gadolinium was present in tracer quantities, cerium carrier was added. This carrier was chosen because it can be readily separated from trivalent rare earths such as gadolinium by utilizing the Ce^{+4} state.

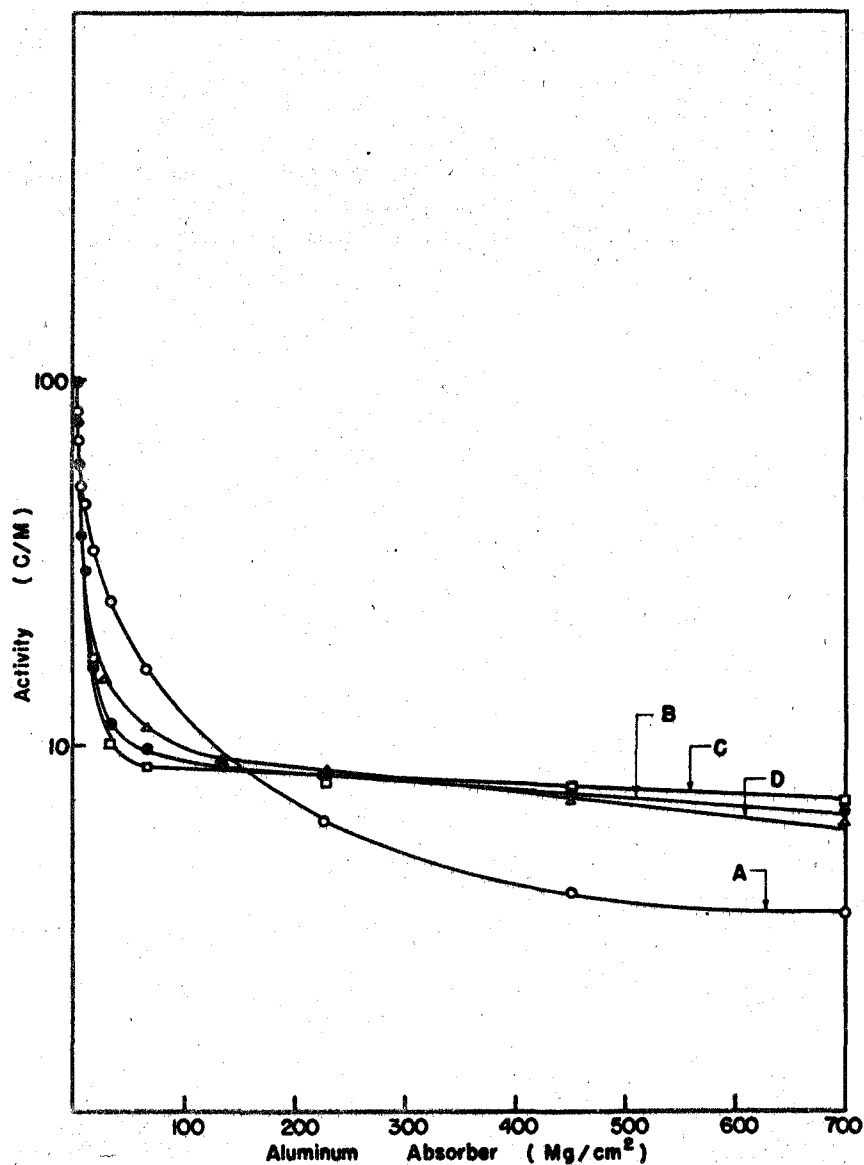


Fig. 3. Absorption Curves on Europium and Gadolinium Fractions.

- A. First EuSO₄ fraction.
- B. Filtrate from A containing gadolinium activity.
- C. Gadolinium further purified from europium (filtrate from D).
- D. Second EuSO₄ fraction after inactive Eu⁺³ added to B.

Purification of Gadolinium by Ion Exchange Methods

The purification of the gadolinium activities with respect to europium (also samarium and ytterbium) is discussed in the preceding portion. Any other rare earth contaminant could not be separated in this way. Although the spectrographic analysis of the original europium showed the presence of only samarium and gadolinium (in trace amounts) it was thought necessary to test the active gadolinium fraction for rare earth contaminants. Ion exchange techniques have been discussed in many recent papers and a review of this method will not be given here. Suffice it to say that a number of sulfonated aromatic resins will adsorb the rare earths. These rare earths may then be eluted from the resin with a variety of agents the most common one being solutions of citrate ion. The rare earths are thus separated on the resin bed according to their aqueous ionic size.

The first experiments with the active gadolinium utilized a small ion exchange column, eluate counting cell, and recording rate meter. A sample of the active gadolinium was adsorbed on Dowex-50 resin and eluted with 5% ammonium citrate solution at a pH of 3.4. Only one sharp peak was observed on the graph recorded by the rate meter circuit. Pre and post peak fractions gave identical

aluminum absorption curves indicating either that contaminants were absent or that no separation was effected. To check the possibility that a trace of active europium could still be present and also to test the efficiency of the experimental setup, a known mixture of active europium and gadolinium was adsorbed and eluted from the column. The eluting agent and resin were the same as used in the earlier experiment. Only one broad peak was observed. However, aluminum absorption curves on the pre and post peak fractions indicated that a separation of approximately 20% was achieved. It was suspected that two peaks were not observed because the volume of the counting cell (0.5 ml) was too large with respect to the total volume of liquid in the column (0.9 ml) allowing mixing of the peaks at this point. Accordingly a much smaller cell (0.01 ml) was prepared. Another sample of the active gadolinium was adsorbed on the resin column and eluted with a 5% ammonium citrate solution at a pH of 2.5. Once again only one peak was observed and no differences were detectable in pre and post peak fractions. These experiments indicate that contaminants other than the immediate neighbors of gadolinium were not present. Further, if either terbium or europium were present, they could only be present in trace amounts. Further separation of known

mixtures of active europium and gadolinium were not attempted.

Two more attempts were made later to establish the purity of the gadolinium with large ion exchange resin columns. A column 1 cm. I.D. was filled with Nalcite high capacity resin (60 - 80 mesh) to a height of 20 cm. The resin had previously been stirred with a saturated ammonium chloride solution so that it would be in the ammonium cycle. All the excess chloride ion was removed by backwashing the column with distilled water. In the first and second column runs the activity was eluted with 0.1% and 0.3% ammonium citrate solution respectively. In the second run the specific activity (c/min/ml) was determined with a dipping counter tube and only one sharp peak of activity was observed. In both the first and second runs pre and post peak fractions gave identical aluminum absorption curves. As gadolinium and terbium activities produced in neutron bombarded gadolinium were separated effectively by similar ion exchange methods the inference is that the gadolinium from the deuteron bombarded europium is quite free from any contamination.

Half-life Studies

After evaporation of a small sample of the active gadolinium in an aluminum cup, a drop of Zapon solution

was evaporated over the sample thus holding the activity in place. The aluminum cup was set on a brass plate and the whole assembly was thus inserted below the electro-scope chamber.

The decay of the activity has been followed over a period of 600 days (see Figure 4). As will be noted in the figure the half-life for nearly 300 days can be given by a 155 day line; however, the half-life is definitely lengthening into a longer period, subtraction of which shortens the 155 day line to 150 days. The longer period is assigned to Gd^{153} and the 150 day activity to Gd^{151} . A further discussion of the activities and mass assignments is given in another part of this thesis.

Energy Measurements

The energy of γ -rays associated with the gadolinium from deuteron bombarded europium has been determined by absorption in copper, tin, lead and tantalum. In all cases the composite curves consisted of three components, a 265 Kev and 101 Kev γ -rays and a 42 Kev x-ray. In copper the half-thicknesses were 5.8, 1.6 and 0.17 g/cm² corresponding to the above energies. The experimental curves are given in Figure 5. The experimental half-thicknesses for lead were 1.3, 0.28 and 0.062 g/cm² giving 268, 85 and 47 Kev as the energies (Figures 6 and 7). The result of three absorption curves in tin place the energy of

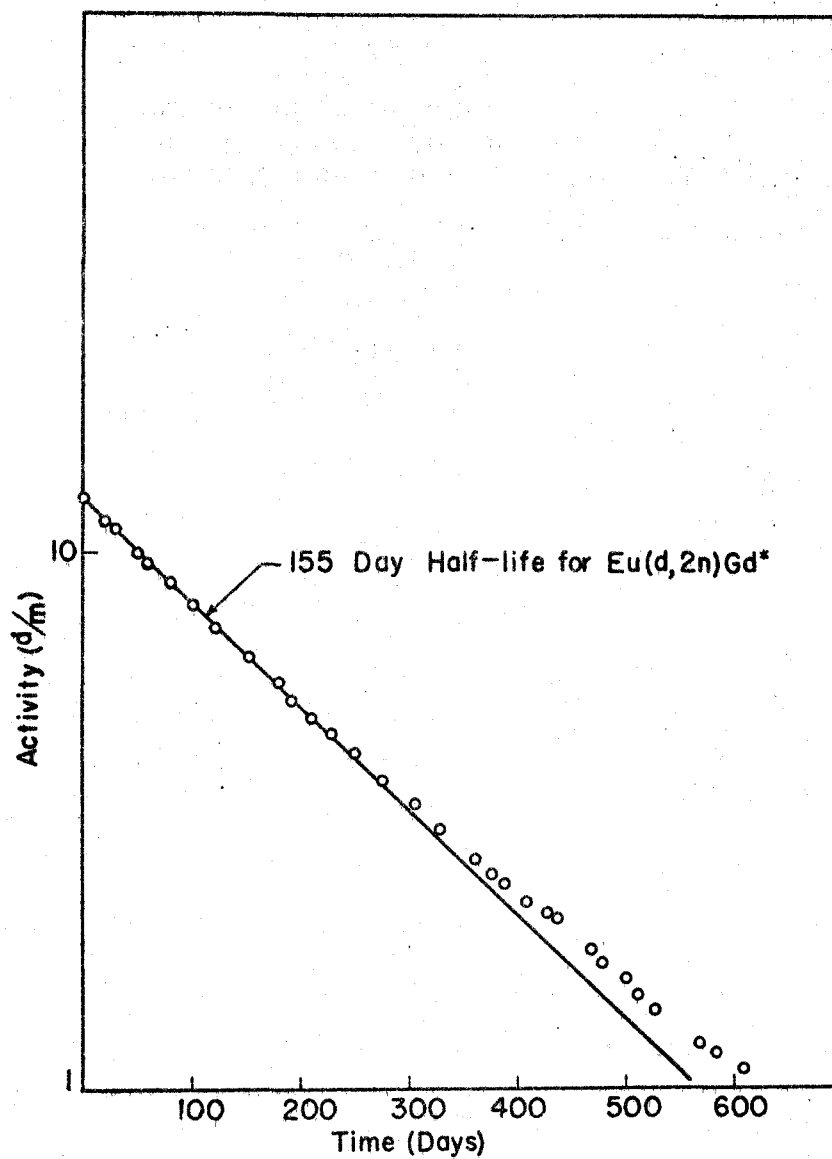


Fig. 4. Decay Curve of Gadolinium Produced in Deuteron Bombarded Europium.

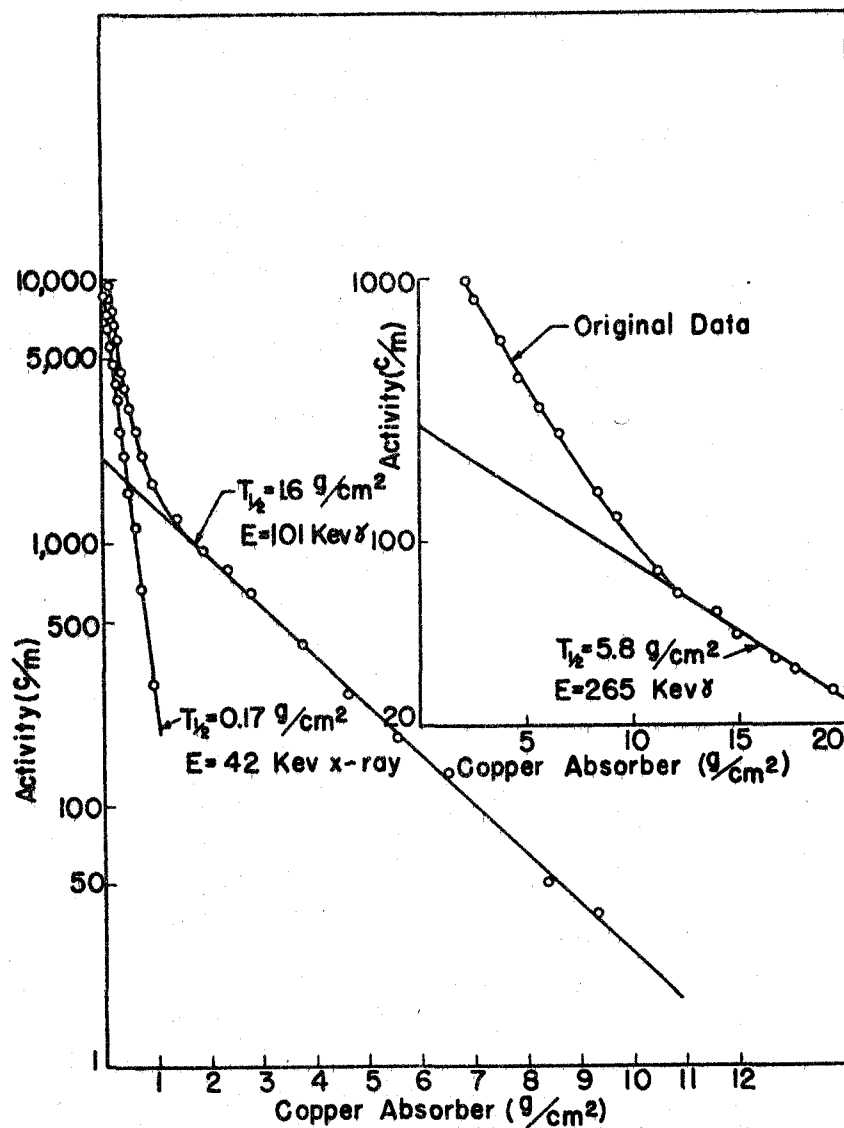


Fig. 5. Copper Absorption Curve on Gadolinium Produced in Deuteron Bombarded Europium.

The X-ray and 101 Kev γ -ray are Subtractions from the Original Data in the Inset.

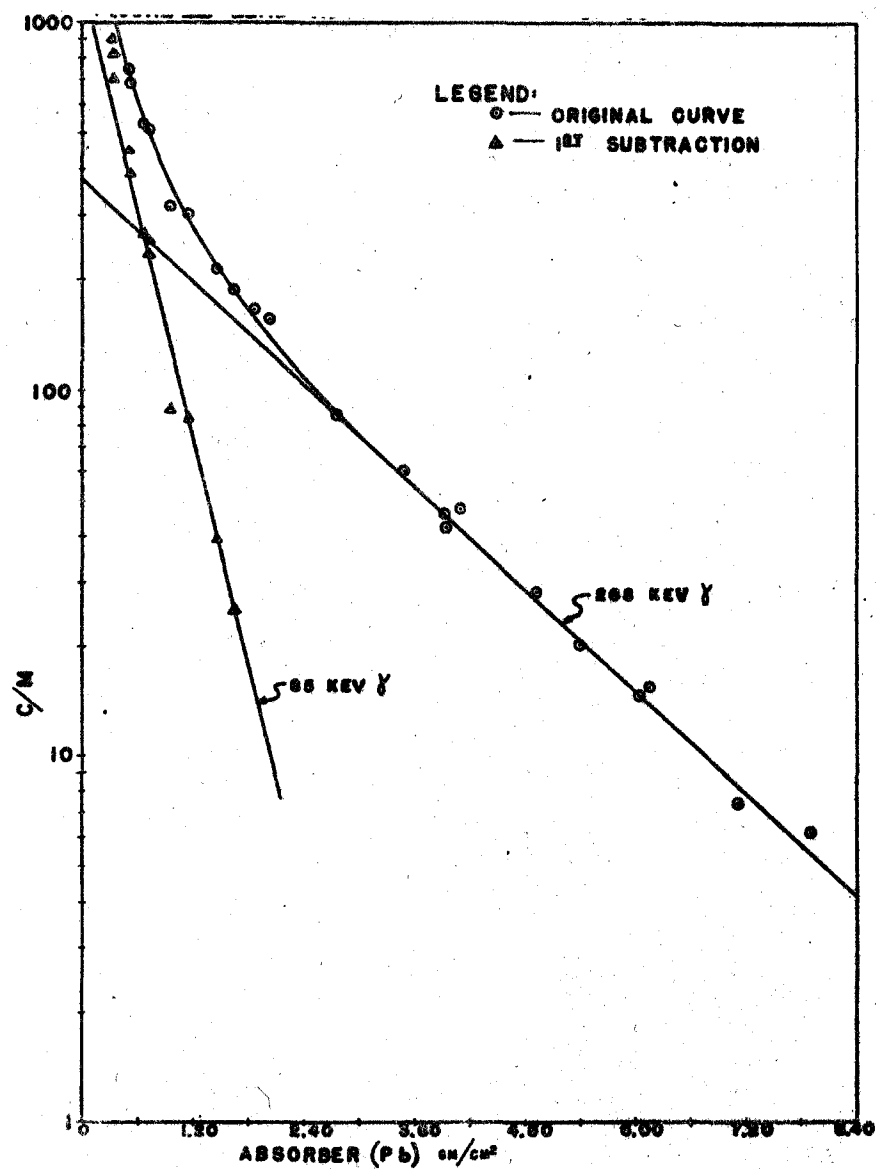


Fig. 6. Lead Absorption Curve on Gadolinium Produced in Deuteron Bombarded Europium.

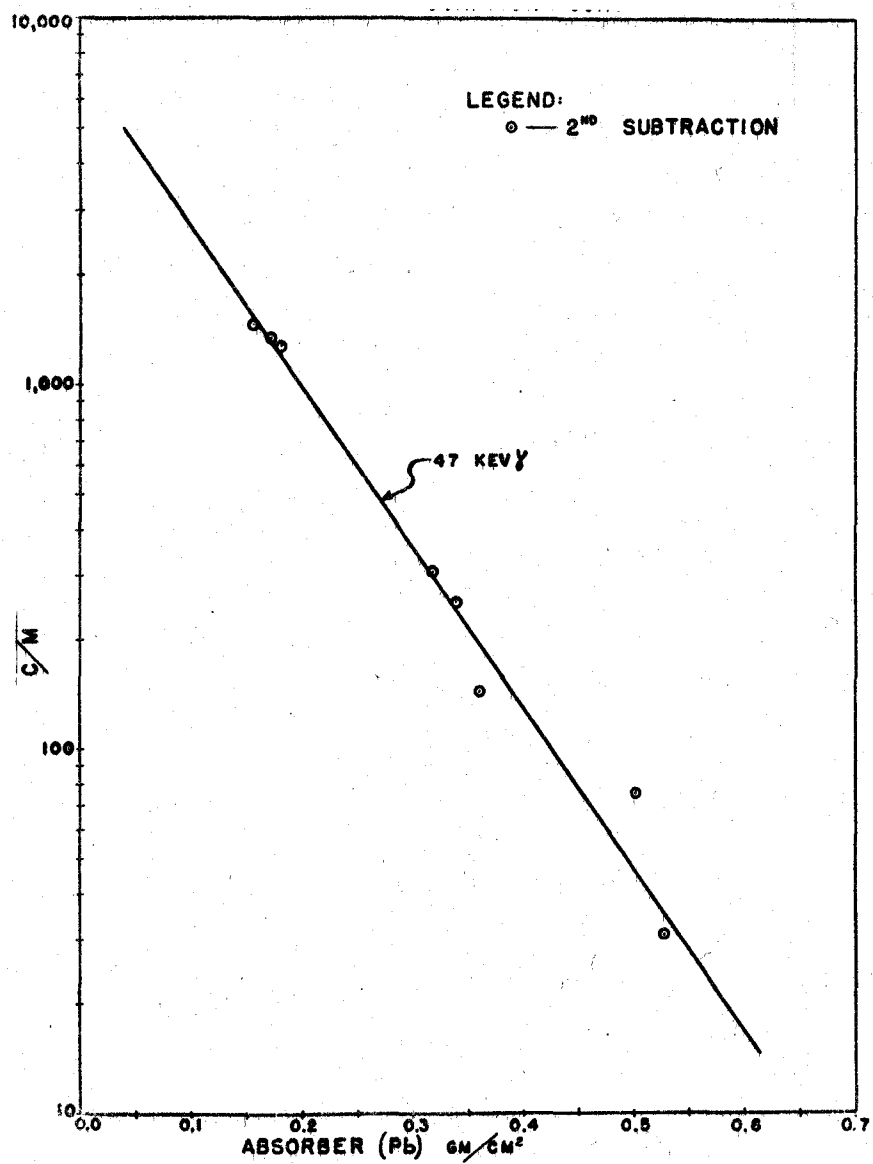


Fig. 7. X-ray Present in Gadolinium Decay after Subtraction of Higher Energy γ -rays (see Fig. 6).

the intermediate γ -ray between 105 and 110 Kev. Tantalum has also been tried as an absorber but its use led to values considerably higher than those from other elements. A part of the difficulty may lie in the absorption coefficient versus energy data for tantalum since several authors are in considerable disagreement in this regard. The proximity of the 101 Kev γ -ray to the K absorption edge (87.5 Kev) in lead may make that element a rather poor choice in this study. The 101 Kev γ -ray is most probably identical with a 102 Kev γ -ray assigned to Gd^{153} by Cork, Shreffler and Fowler (10).

Since the gadolinium activity must be either an isomeric transition or a decay to europium, the decay by positron emission is a good possibility. Results from attempts to detect positrons by bending them into a counter with a permanent magnet show that a small percentage of positron decays is statistically possible but not at all probable. The experimental data for two measurements with different geometry of sample, magnet and counter are given in Table V.

There are several groups of conversion electrons one of high intensity at 220 Kev which is probably K conversion electrons of the 265 Kev γ -ray (see Figure 8). There may also be some L conversion electrons present from the 265 Kev γ . A similar aluminum absorption curve

taken 500 days later shows a higher preponderance of low energy electrons (see Figure 9). Conversion of the 102 Kev γ -ray would lead to such low energy electrons.

TABLE V
Magnetic Deflection of Positrons

Measure- ment	No magnet	Positrons	Negatrons
1	89 ± 5 c/m	47 ± 3 c/m	12,160 c/m
2	94 ± 2 c/m	117 ± 2 c/m	8,267 c/m

Critical Absorption Measurements

As mentioned previously the possibility exists that the observed transition is isomeric in gadolinium (e.g. $Gd^{154*} \rightarrow Gd^{154}$ stable) instead of K-electron capture. Therefore the 42 Kev x-ray was studied in detail, as its energy would be indicative of the element to which the activity decays. To determine whether the x-ray originated from a gadolinium or europium nucleus, critical absorption measurements were performed using lighter rare earths as absorbers. Europium K_{α} radiation has an energy of 41.5 Kev while that of gadolinium is 43.5. Since the K absorption edges of lanthanum, cerium, praseodymium, and neodymium occur at 39.0, 40.5, 42.5 and 44.5 Kev respectively, these elements were selected as absorbers. An europium K_{α} x-ray

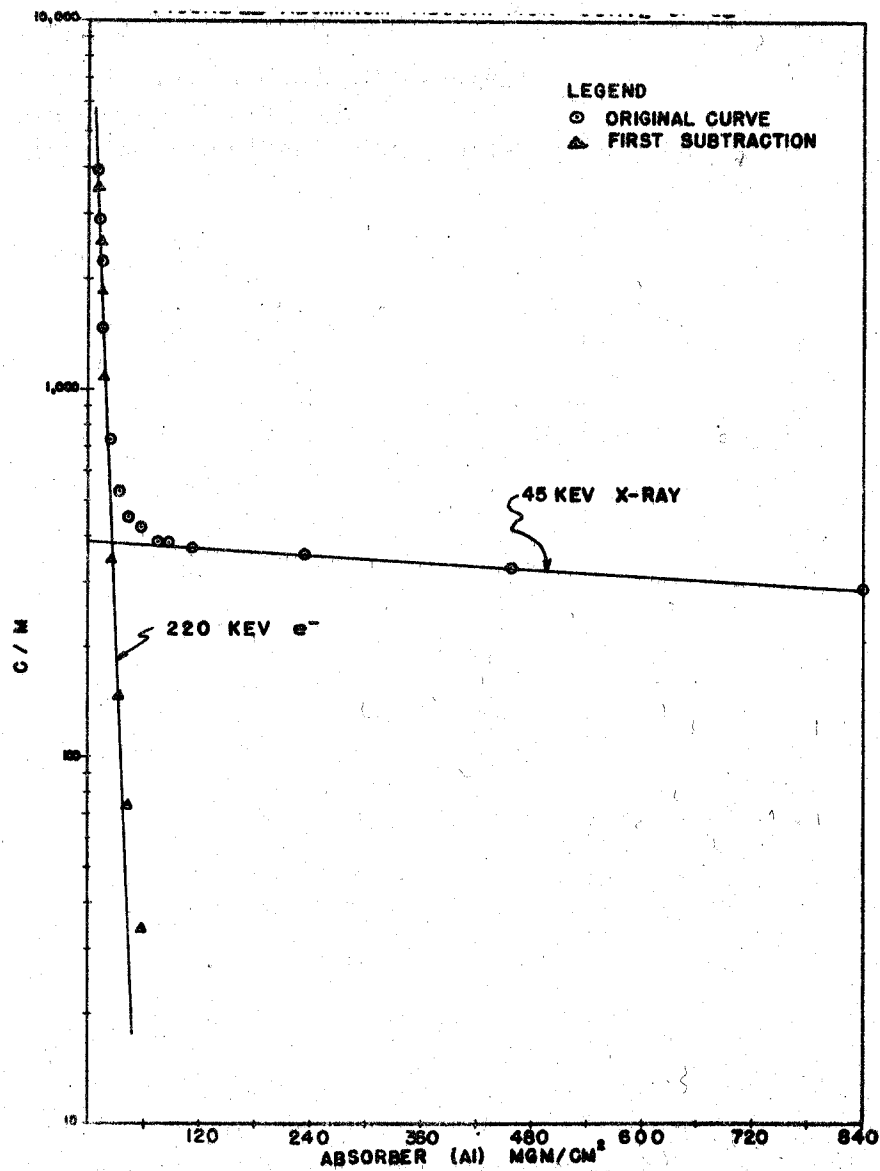


Fig. 8. Aluminum Absorption Curve on
Eu(d, 2n)Gd.

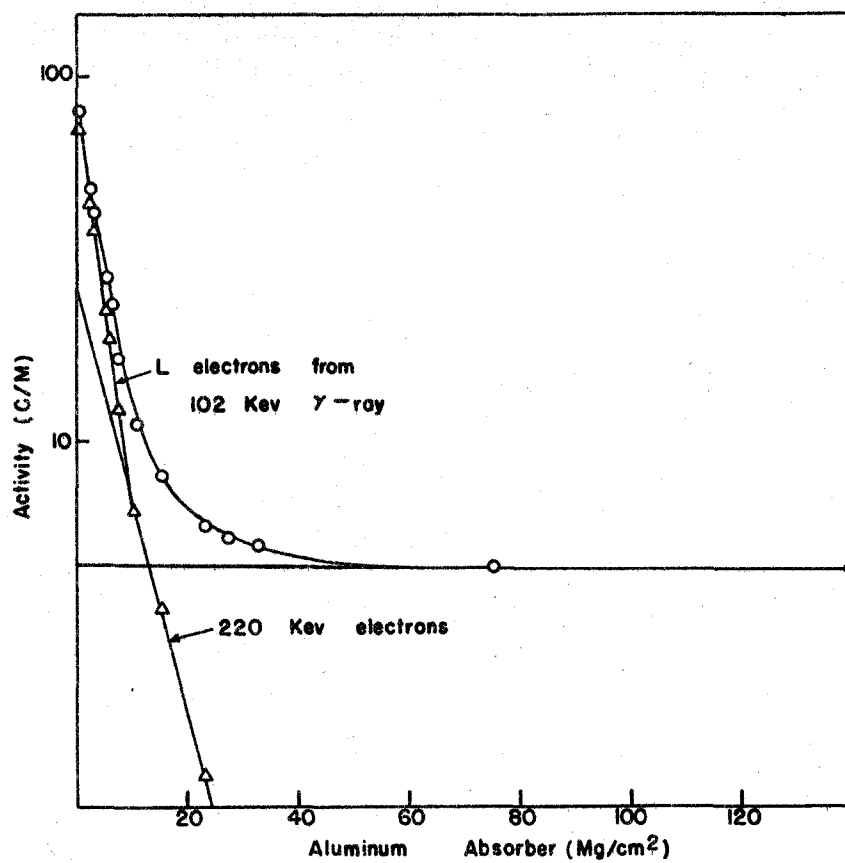


Fig. 9. Absorption Curve on Eu(d, 2n)Gd
500 Days after End of Bombardment.

would be strongly absorbed by lanthanum and cerium and transmitted by the other two elements. A gadolinium K_{α} x-ray would be transmitted only by neodymium and absorbed by the other three rare earths. Although absorption versus energy data for these elements are far from complete, an expected set of half-thicknesses for the four elements can be estimated (31).

TABLE VI
Expected Absorption Data
for γ -rays in Rare Earths

γ -ray Energy	Half-thickness in g/cm ²			
	La	Ce	Pr	Nd
41.5 Kev	.027	.027	.127	.127
43.5 Kev	.027	.027	.027	.127
102 Kev	.270	.260	.250	.240
265 Kev	1.93	1.88	1.83	1.78

The absorbers of the rare earth oxide were prepared from pure oxides loaned through the courtesy of F.H.Spedding and the rare earth separation group. By slurring a weighed amount of the particular oxide in an ether solution of paraffin, a uniform layer of the oxide could be formed in an aluminum dish. The concentration of paraffin was such as to hold the oxides in place after the volatile solvent

was removed. In the first series of measurements, only one absorber was prepared from each element with less than 100 mg/cm^2 . Only 0.3 g/cm^2 of absorber could be placed in each dish because of size limitation. To obtain greater absorber thickness one dish was piled on top of another.

The first attempt to establish the energy of the x-ray was inconclusive in that no significant difference could be detected in the absorption curves of the x-ray in these elements. As the other two γ -rays are of higher energy than the x-ray, the curves were continued to large absorber thickness in order to correct for these components. This procedure proved impractical because of the amount of rare earth oxide needed to reach a flat portion in the curve (see Table VI). Accordingly the x-ray absorption in the region from 0- 180 mg/cm^2 was examined. Absorbers of 180, 150, 120, 90, 60, 40 and 20 mg/cm^2 of the four rare earths were made up in aluminum cups. The lighter weight absorbers were difficult to prepare evenly and the experimental points were somewhat erratic. The same sample of Gd^{153} was used with the same geometry for all of the curves. Each curve was rerun twice and the results were very similar. The experimental data are plotted in Figure 10. From the experimental half-thicknesses of 75 mg/cm^2 for La_2O_3 , 72 mg/cm^2 for CeO_2 , 160 mg/cm^2 for Pr_6O_{11} and 156 mg/cm^2 for Nd_2O_3 , the

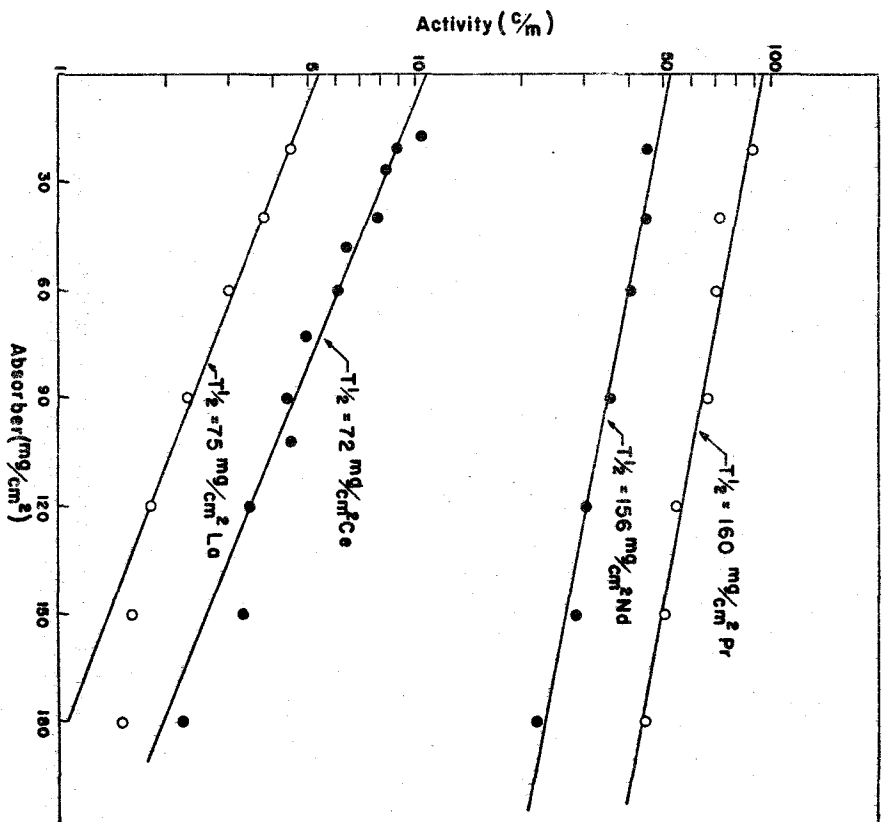


Fig. 10. Critical Absorption of X-ray Present in Eu(d, 2n)Gd with La, Ce, Pr, and Nd Absorbers.

observed ratio for half-thicknesses for cerium/praseodymium is 1:2.2. The calculated ratio (see Table VI) for the absorption of an europium x-ray is 1:4.7 for these absorbers. As was stated earlier, the fact that other gamma rays are present and that the subtraction of these more energetic components cannot be made tend to make the experimental ratio approach 1 rather than 1:4.7. From these data it appears quite conclusive that the x-ray energy is between the absorption edges of cerium and praseodymium and is therefore the K_{α} x-ray of europium.

Decay Schemes

Although the coincidence circuit (see Figures 1 and 2) and counter assembly were described earlier, a few statements regarding the operation of the instruments are given here. The end-window (2.2 mg/cm^2) counters inset in an aluminum box were protected from light by a cardboard box covering the whole assembly. Brass or copper plates 1.2 mm in thickness with a small hole 4 - 6 mm in diameter were inserted in a slot midway between the counters. If a sample plate which would absorb scattered and secondary radiations were not present between the counters, other investigators from this laboratory found that a number of spurious coincidences occurred. The number of spurious coincidences with a brass plate

(1.1 g/cm²) and small hole for sample mounting was assumed to be negligible. With measurements on high intensity samples a lead sheet was used as a sample holder instead of brass. A straight line connecting the anodes of the end-window counters would pass through the center of the hole in the sample plate.

The activity to be studied was evaporated on thin mica sheets or formvar films (0.1 mg/cm²). The mica or formvar was then glued to the sample plate with the activity viewing both counters through the central orifice. With the sample located midway between the counters, approximately equal counting rates were observed on scalars connected to channels 1 and 2. A dilute solution of zapon evaporated on the activity held the sample in place.

Before an actual measurement began, the coincidence rate due to cosmic and stray γ radiation was determined. After insertion of a sample plate in the slot between the counters, the background counts on both channels and the coincidence background were determined over a period of 24 hours. A list of coincidence backgrounds with different thickness of aluminum foils between the counters is given in Table VII.

The coincidence rate was slightly lower (.060) and constant for a range of lead absorbers. It should be noted that the active areas of the counters were in a

TABLE VII

Background Coincidences

Mg/cm ² of Al	0	32.4	64.8	150	272
Coinci- dence Rate	.155±.008	.143±.010	.120±.010	.102±.004	.092±.004

horizontal plane in the experimental assembly. With the counters in a vertical plane the coincidence background is increased by about a factor of three. This increase is due to the fact that a single cosmic ray coming through a vertical assembly will give a coincidence count while it takes a cosmic ray shower to give a count in the horizontal assembly.

The variable gate was maintained at the resolving time of 2.06 microseconds for all of the experimental determinations of coincidence rates. The resolving time remained constant over the course of the investigation.

Two long-lived gadolinium activities are produced in deuteron bombarded europium as previously stated. The shorter-lived Gd¹⁵¹ is produced by about a factor of 20 over Gd¹⁵³, so this coincidence study will deal mostly with Gd¹⁵¹. The decay scheme of Gd¹⁵³ will be discussed in detail later in connection with activities produced in

neutron bombarded gadolinium.

As both Gd^{151} and Gd^{153} decay by K-electron capture the europium K x-ray (41.5 Kev) is present. The γ -ray energies were determined (see Figure 5) as 102 and 265 Kev. On the basis of work to be discussed later, the 265 Kev γ -ray is associated with the decay of Gd^{151} . Both γ -rays are internally converted.

A tentative decay scheme is assumed in which Gd^{151} decays to an unstable Eu^{151} by K electron capture and then to the ground state by emission of a 265 Kev γ -ray. Coincidences may then be determined in the decay of Gd^{151} between the 220 Kev K conversion electrons and the K x-rays from either the conversion process or the electron capture step. Obviously these K x-rays are also in coincidence in such a decay scheme as are also the K x-ray from the electron capture step and the 265 Kev γ -ray. These measurements may be complicated by the presence of Gd^{153} , but the determinations are assumed to be valid because of the low intensity of Gd^{153} as compared to Gd^{151} .

If the tentative decay scheme for Gd^{151} is correct only electrons from the conversion of the 265 Kev γ -ray are present. To check this assumption a sample of Gd^{151} - Gd^{153} activity was mounted on mica sheet (2 mg/cm²) and inserted in the counter assembly. Sufficient aluminum was placed between counter 1 and the sample to absorb

all electrons and absorbers were added for each measurement to channel 2 until all the electrons were also absorbed. After correcting the experimental values for background and accidental coincidences the ratio of $e\gamma$ coincidences to electron count at the same absorber thickness was a constant. This result indicates that only one group of electrons is emitted. The experimental data are plotted in Figure 11.

The same experiment was repeated four months later with slightly different geometry conditions. The ratio of $e\gamma$ coincidences to electron count is the upper curve in Figure 11. The experimental results are given in Table VIII. The same aluminum absorber (64.8 mg/cm^2) was kept on the number 1 side for all measurements.

The $N_{e\gamma}/N_e$ value is probably affected in this case by Gd^{153} coincidences. The Gd^{151} to Gd^{153} ratio decreases with time because of the longer half-life of Gd^{153} .

In another series of measurements, absorbers were added to channel 2 and no absorbers were present between the sample and counter 1. After the coincidences were corrected for background, accidental and $x\gamma$ coincidences, the resulting points on a semi-log plot gave a straight line with a half-thickness of 5 mg/cm^2 . This half-thickness corresponds to the 220 Kev conversion electrons. This result is in agreement with the tentative decay

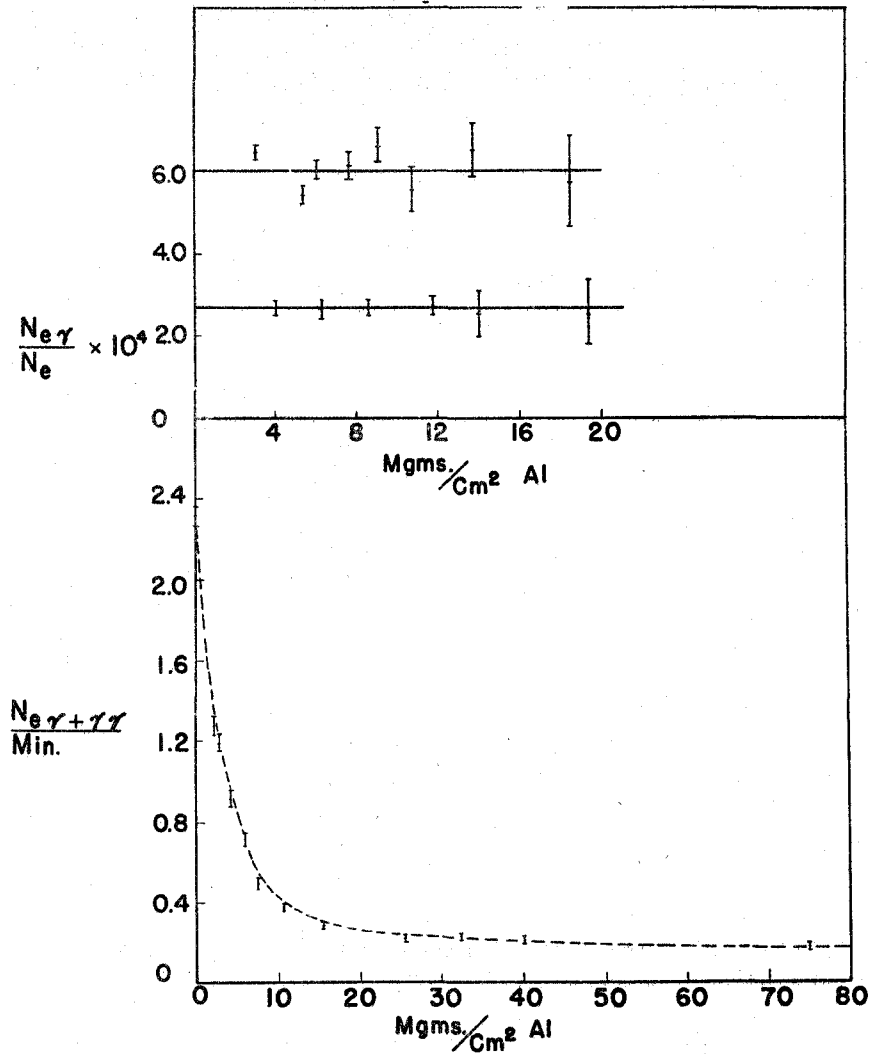


Fig. 11. Coincidence Measurements on Eu(d, 2n)Gd .

scheme of Gd^{153} as these coincidences between the 220 Kev electrons and K x-rays were predicted. The first two points on the above mentioned plot were above the straight line and are assumed due to a contribution from Gd^{153} electron x-ray coincidences.

TABLE VIII
Coincidence Measurements on Gd^{151} - Gd^{153}

I c/m	II mg/cm ²	II c/m	$N_{e\gamma+x\gamma}/\text{min}$ uncorr.	$N_{e\gamma}$ corr.	$(N_{e\gamma}/N_e)10^4$
313	0	3430	$2.305 \pm .053$	$2.054 \pm .054$	$6.44 \pm .17$
312	2.3	2177	$1.275 \pm .042$	$1.051 \pm .044$	$5.43 \pm .23$
311	3.0	1842	$1.188 \pm .0305$	$0.972 \pm .032$	$6.06 \pm .20$
312	4.6	1395	$0.916 \pm .036$	$0.709 \pm .038$	$6.13 \pm .33$
316	6.0	1020	$0.714 \pm .031$	$0.516 \pm .032$	$6.62 \pm .41$
314	7.7	773	$0.489 \pm .025$	$0.296 \pm .028$	$5.56 \pm .53$
324	10.7	524	$0.372 \pm .015$	$0.185 \pm .019$	$6.51 \pm .67$
318	15.4	407	$0.280 \pm .015$	$0.096 \pm .019$	5.75 ± 1.14
323	23.1	302	$0.212 \pm .012$		
315	32.4	250	$0.221 \pm .012$		
317	40.1	244	$0.206 \pm .011$		
324	75	239	$0.167 \pm .011$		

With lead absorbers on both channels, $\alpha\alpha$ and $\alpha\gamma$ coincidences were observed although few in number. No coincidences were observed when the x-ray was absorbed on both channels. This would support the assumption that only one gamma ray is present in the decay of Gd^{151} .

Neutron Bombarded Gadolinium

Survey of Previous Work

A detailed review of literature on gadolinium activities was given earlier. The status of gadolinium activities was reviewed by G. T. Seaborg and I. Perlman (13) at about the time the study of neutron induced gadolinium activities began in this laboratory. A 155-day activity decaying by K electron capture was assigned to Gd^{153} (32). This activity formed in deuteron bombardment of europium was assigned to Gd^{153} because a long-lived neutron induced activity had been assigned to Gd^{153} on the basis of mass spectrographic methods. It was presumed that the two long-lived activities were the same. The table of isotopes also lists a 4.5 minute Gd^{161} (33) and tentatively assigns an 18-hour activity to the same nuclide (8). An 8.6 day activity was also tentatively assigned to gadolinium although the investigators state the activity may be due to an impurity (34).

An extensive investigation of short-lived gadolinium activities reported recently (7) assigned the 18-hour activity to Gd^{159} . A 218 sec. Gd^{161} - 6.75 day Tb^{161} chain was also reported.

Na-Hg Extractions of Possible Contaminants

Samples of gadolinium oxide were irradiated at two different times at the Argonne Laboratory. The first bombardment of 400 hours did not give a sufficiently intense sample of the long-lived gadolinium, so a second bombardment lasting 854 hours was obtained.

As samarium and possibly ytterbium were spectrographically detected in the gadolinium and as (n,p) and (n, α) reactions on gadolinium would give europium and samarium activities, the irradiated sample was extracted several times with sodium amalgam. The small amount of activity extracted into the amalgam had the same energy characteristics as the non-extracted portions. The extractions were performed several days after removal of the sample from the pile and short-lived activities such as the 47-hour Sm^{153} would have decayed. No attempt was made to extract contaminants in the longer irradiated gadolinium sample.

Another sample of Gd_2O_3 bombarded in the Oak Ridge pile for one week was obtained to study the short-lived activities. A Na-Hg extraction performed on this irradi-

ated sample 30 hours after removal from the pile did reveal the presence of Sm^{153} . The extracted activity decayed with a 48 hour half-life. The energy of this activity based on absorption in aluminum was 730 Kev. These experimental results are in good agreement with published values of a 47 hour half-life (35) and 780 Kev β (36) assigned to Sm^{153} .

Separation of Activities by Ion Exchange Methods

The possibility that two or more long-lived activities were present in the neutron irradiated gadolinium seemed likely because of the consistent difference in half-life curves measured with and without absorbers. Without added absorber, the observed half-life was shorter. Since the x-ray emitted in the gadolinium decay is not appreciably absorbed in 225 mg/cm² of aluminum, the contaminant must have the shorter half-life and little or no γ -ray. As the possibility of long-lived samarium, europium and ytterbium contaminants was investigated repeatedly with negative results, the extraneous activity could not be due to them. To separate other rare earths it was necessary to set up ion exchange columns.

Separation of impurities in the long-lived gadolinium was not attempted with the small column, counting cell

and recording rate meter. The combination of a low load capacity of the small column (1.5 - 2.0 mg) and low specific activity of the gadolinium prohibited investigation with this instrument.

Several cation exchange column runs were carried out on the irradiated gadolinium in separating and identifying impurities. These experiments will be discussed in chronological order. In the early runs the activity in a known fraction of the eluate was determined by evaporation on a watchglass and subsequent counting of the sample. Later dipping counters were used to determine the activity in the eluate.

Nalcite high capacity resin was ground at dry ice temperatures both to aid in the grinding and also to ensure that no chemical change would occur in the resin. The mesh size of the resin varied in different experiments. In all runs, the resin was stirred twice with saturated ammonium chloride solutions and then backwashed in a column until a chloride ion test was negative. The same columns (1 cm I.D.) were used in all experiments with the height of the resin bed varying from 12 - 20 cm. Usually the activity was eluted with 0.1% ammonium citrate solution at a pH of 5.5. In all cases the eluting solutions were 0.1% in phenol to inhibit mold growth.

In the first elution, an activity other than gadolinium was detected in the eluate when the bulk of the activity was still in the upper fifth of the column. A complete separation of this activity from the gadolinium was effected as evidenced by aluminum absorption curves on the two fractions (see Figure 12). The activity decayed by emitting a β of about 0.9 Mev and little or no γ -ray. Chemically this activity appeared to be a rare earth, since it was carried by YF_3 when iron was present as a hold-back carrier.

The intensity of this extraneous activity in the first bombardment of Gd_2O_3 was such that aluminum absorption curves of the gadolinium fraction before and after the ion exchange separation seemed to be identical. The sample of active gadolinium measured through 225 mg/cm^2 of aluminum would give a correct value for the half-life as the contribution of the extraneous activity through this absorber would be negligible. This statement is corroborated as the purified gadolinium from this column run has been measured over 280 days decaying with a 240-day half-life. This value agrees well with the 236 ± 3 day half-life measured with the additional absorber. Enough of the extraneous activity from this elution and others which followed was collected to mount a low intensity sample

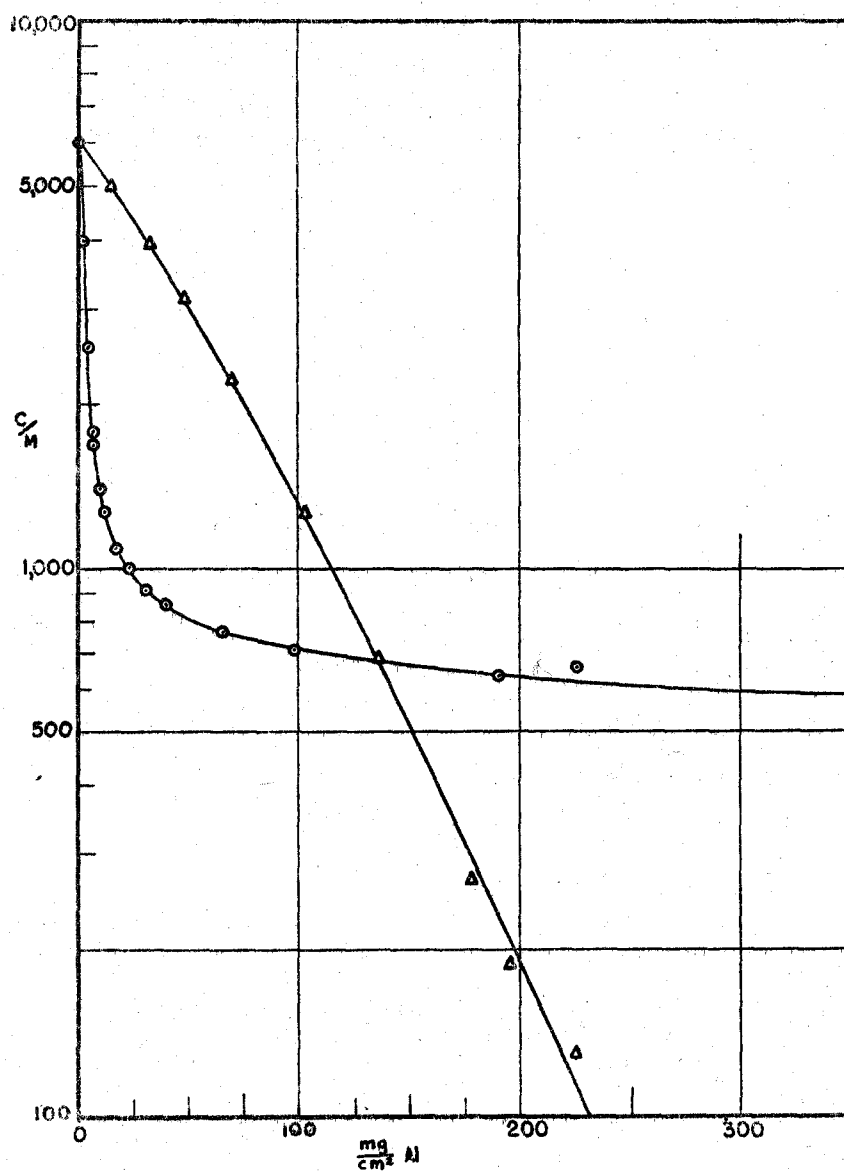


Fig. 12. Absorption Curves on Contaminant (Δ) and Gadolinium (○) before Column Separation.

for half-life measurements. This will be discussed later in the identification of this activity.

The second sample of Gd_2O_3 which had received twice as much irradiation as the first sample also was purified by ion exchange methods. The first elution started 7 days after the Gd_2O_3 was removed from the pile. Again the first fraction off the column was the extraneous activity with the 0.90 Mev beta ray. This fraction was quite active and a sample was mounted for half-life measurement. As the elution progressed other fractions were also mounted for half-life studies from this run. A sample of the eluate preceding the gadolinium peak contained an activity which was neither gadolinium nor the extraneous activity mentioned earlier. The identification of these activities are discussed in the following section.

Identification of Contaminants

Two samples (22.3 and 156 mg) of the Gd_2O_3 which was bombarded for 854 hours were adsorbed and eluted from the cation exchange columns. In both cases the activity was determined in the eluate with a dipping counter tube. The graduate in which the counter was immersed and the counter were both clamped so that the same geometry was reproduced for all samples. A plot of the c/min/ml versus liters of eluate for the 156 mg sample is given in Figure 13.

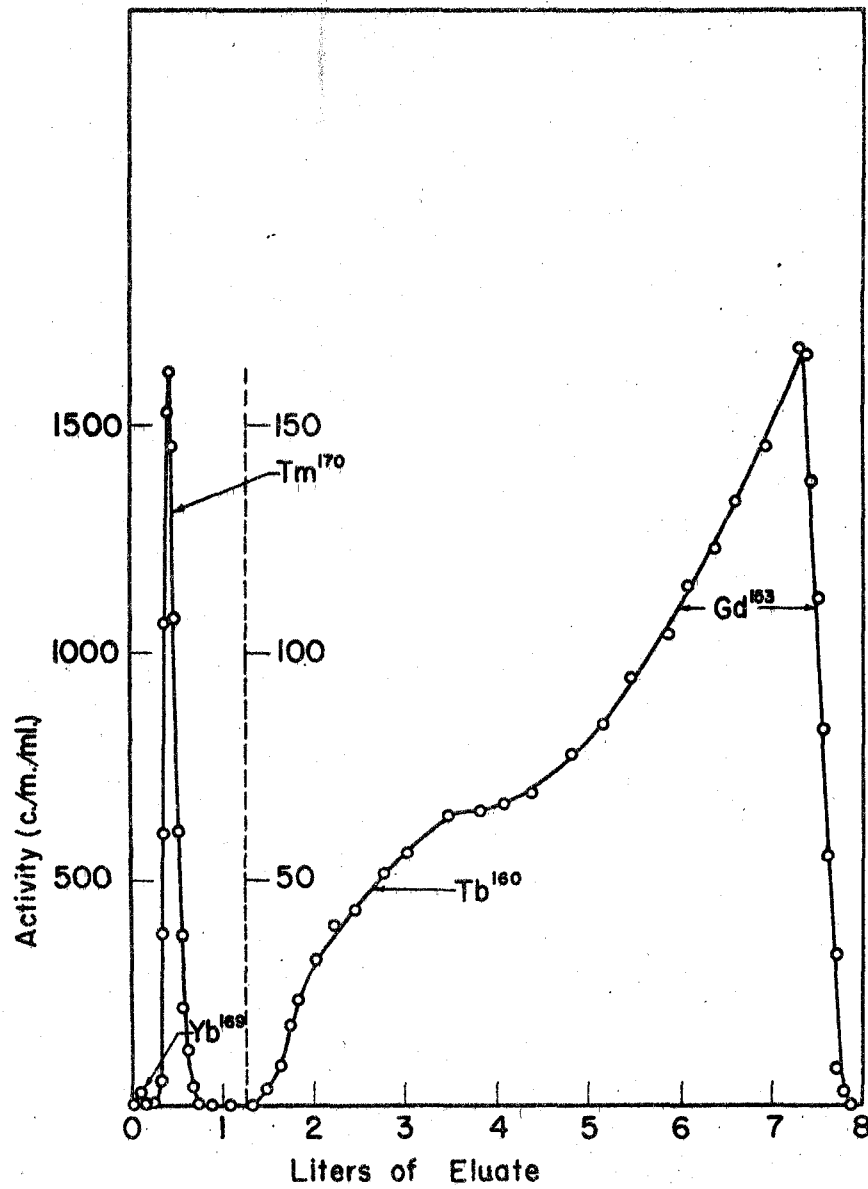


Fig. 13. Elution Curve of Neutron Induced Activities in Gd_2O_3 .

The extraneous activity mentioned earlier is the same as the activity in the sharp peak preceding the gadolinium off the column (see Figure 13). This activity was identified as Tm^{170} in a number of ways. As thulium had not been detected spectrographically and as Tm^{170} was the bulk of the long-lived contaminating activity, considerable time was spent in verifying the identification. Tm^{170} decays with a half-life of 127 days emitting a β of 1.1 Mev (37) as determined by absorption in aluminum. The decay of a sample of Tm^{170} separated from the first bombardment of Gd_2O_3 was measured over 8 months (see Figure 14). The experimental value of 127 days is the same as that previously reported for Tm^{170} . A β -ray energy of 0.95 Mev was determined by aluminum absorption on a purified Tm^{170} fraction agreeing well with the literature value. A high intensity source of Tm^{170} gave a γ to β ratio of 1×10^{-4} . This ratio agrees well with a value of $.5 \times 10^{-4}$ reported by von Bothe (35).

As a final check on the identification of the activity as Tm^{170} , it was decided to bombard Tm_2O_3 and compare the radiations. The rare earth group loaned a sample of Tm_2O_3 which contained both erbium and ytterbium as impurities. After a one week bombardment at the Argonne pile, the activities present in the sample were separated on an ion exchange column. Two peaks of activity were

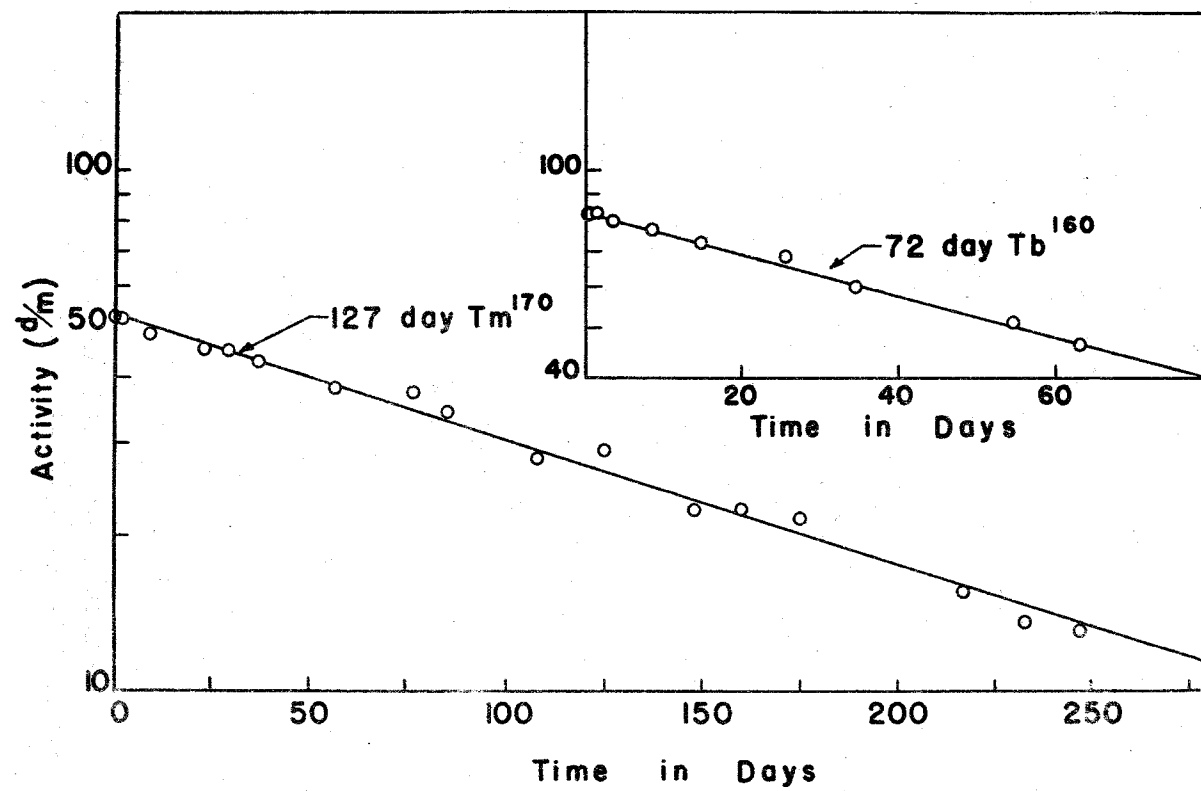


Fig. 14. Decay Curves for Tm^{170} and Tb^{160} .

separated with the thulium fraction about 120 times as intense as the second peak later identified as Ho^{166} . The radiations from the Tm^{170} of this sample were the same as the activity thought to be Tm^{170} present as a contaminant in the Gd_2O_3 .

In the elution curve of neutron bombarded gadolinium, the first small peak off the column was identified as Yb^{169} . The identification was based on energy and coincidence measurements in addition to the fact that this activity precedes Tm^{170} from the column which would point to a ytterbium isotope. Aluminum absorption curves of known Yb^{169} and this activity were similar but not identical. If a few per cent of Tm^{170} were assumed to be present, this difference in absorption curves could be explained. Finally, comparison of electron-electron coincidence rates for known Yb^{169} and the activity in question were very similar, both being exceptionally high, a striking characteristic useful for identification.

The same elution curve (see Figure 13) of activities in the neutron bombarded Gd_2O_3 indicates the presence of still another long-lived contaminant. Half-life and energy measurements in addition to the position of activity in the elution curve are the basis for assignment of the activity to Tb^{160} . An aluminum absorption curve on

this pre-gadolinium fraction gave a value of 760 Kev for the β whereas the reported value for Tb^{160} is 750 Kev (35). Half-life measurements over a period of 70 days are plotted in Figure 14, giving a value of 76 days as compared to the reported value of 72 days (35).

Estimation of Impurities in the Gadolinium

Spectrographic analysis did not list thulium as a contaminant and the amount of other trace contaminants could not be estimated by this method. It is possible to calculate the per cent thulium in the Gd_2O_3 from the experimentally observed disintegration rate and known neutron capture cross section value. Knowing the per cent thulium in the Gd_2O_3 , the percentages of other contaminants can be estimated as will be discussed in the following paragraphs.

The disintegration rate for Tm^{170} was calculated from two different elution curves where the starting weight of gadolinium was 156 mg in one case (see Figure 13) and 22.3 mg in the other, both as Gd_2O_3 . The thermal neutron capture cross section for the process $Tm^{169} (n, \gamma) Tm^{170}$ is reported as 100 barns (37) and 118 barns (38). The amount of thulium in 156 mg of gadolinium is calculated as follows:

$$\text{gms of Tm} = \frac{(\text{dis/sec}) (\text{atomic wt of Tm})}{(\sigma) (Pf) (6.03 \times 10^{23}) (P) (1 - e^{-\lambda t})}$$

$$\begin{aligned} \text{gms of Tm} &= \frac{(3.44 \times 10^5) (169)}{(10^{-22}) (3.6 \times 10^9) (6.03 \times 10^{23}) (250) (.177)} \\ &= 61 \times 10^{-7} \text{ gm.} \end{aligned}$$

σ = capture cross section for thermal neutrons

Pf = flux factor for the Argonne pile

P = average power of pile in kilowatts

$1 - e^{-\lambda t}$ = growth factor where λ is decay constant and
t is time of bombardment

The per cent thulium was calculated as 4.5×10^{-3} and 4.8×10^{-3} for the two runs of 156 and 22.3 mg of gadolinium. This amount of thulium is at the limit of spectrographic detection. This illustrates the potential usefulness of the activation method of analysis in certain cases.

As a check on the above method, the disintegration rate for the Tm^{170} produced in the neutron irradiated Tm_2O_3 was estimated. Again the same calculation was performed, i.e. knowing the disintegration rate the weight of thulium in the sample could be calculated. The known weight of thulium adsorbed in the column was 25 mg and the calculated value was 50 mg. On this basis the per cent thulium in the sample is probably good within a factor of two.

Since thulium was in the Gd_2O_3 , erbium, holmium, and dysprosium must also be present in equal or greater amounts

as their natural abundance in ores is greater and these elements are more difficult to separate from gadolinium by ion exchange methods. On the basis of abundance data of the rare earths (39) and separation effected by the ion exchange columns, the following ratios based on thulium as unity are estimated in the Gd_2O_3 .

TABLE IX
Percentages of Contaminants in Gd_2O_3 .

Element	Tm	Er	Ho	Dy	Tb
Ratio	1	40	2.5	60	3
%	4.5×10^{-3}	0.16	1.2×10^{-2}	0.27	1.4×10^{-2}

The neutron capture cross section for the process $Tb^{159} (n, \gamma) Tb^{160}$ was also calculated. The disintegration rate of Tb^{160} was estimated from the elution curve of activities in neutron bombarded gadolinium (see Figure 13). The weight of terbium in the 156 mg of gadolinium was based on the $1.4 \times 10^{-2}\%$ value in Table IX. The capture cross section was calculated as 9.6 barns. Values of <22 barns (38) and 10 barns (40) are reported in the literature. As the calculated and reported cross sections are similar, credence is lent to the percentages of contaminating rare earths in the gadolinium shown in Table IX.

Half-life Studies

Half-life measurements on two gadolinium samples from the first bombardment now extend over 500 days. The decay of one of the samples was measured through 225 mg/cm² of aluminum while readings on the second sample were taken with no absorber. Neither sample was purified by ion exchange methods.

As stated earlier the half-life of 236 ± 3 days determined with the added absorber is considered correct since the contribution of Tm^{170} through the absorber is negligible (see Figure 15). As determined visually, the best fit of a straight line to the experimental points gave the value of 236 ± 3 days. Application of the least square method (41) to the points gave a half-life of 233 days. Another sample of gadolinium with the Tm^{170} separated by ion exchange methods has been measured over 280 days giving a half-life of 240 ± 5 days.

Two half-life determinations on the short-lived components in the first neutron irradiated gadolinium sample gave values of 24 ± 3 hours and 7.2 ± 0.2 days (see Figure 16). The short-lived values were obtained from a composite curve after the subtraction of the 236 day line. Neither sample was purified by ion exchange methods or sodium amalgam extractions. Measurements with and without added absorber gave the same short half-life values.

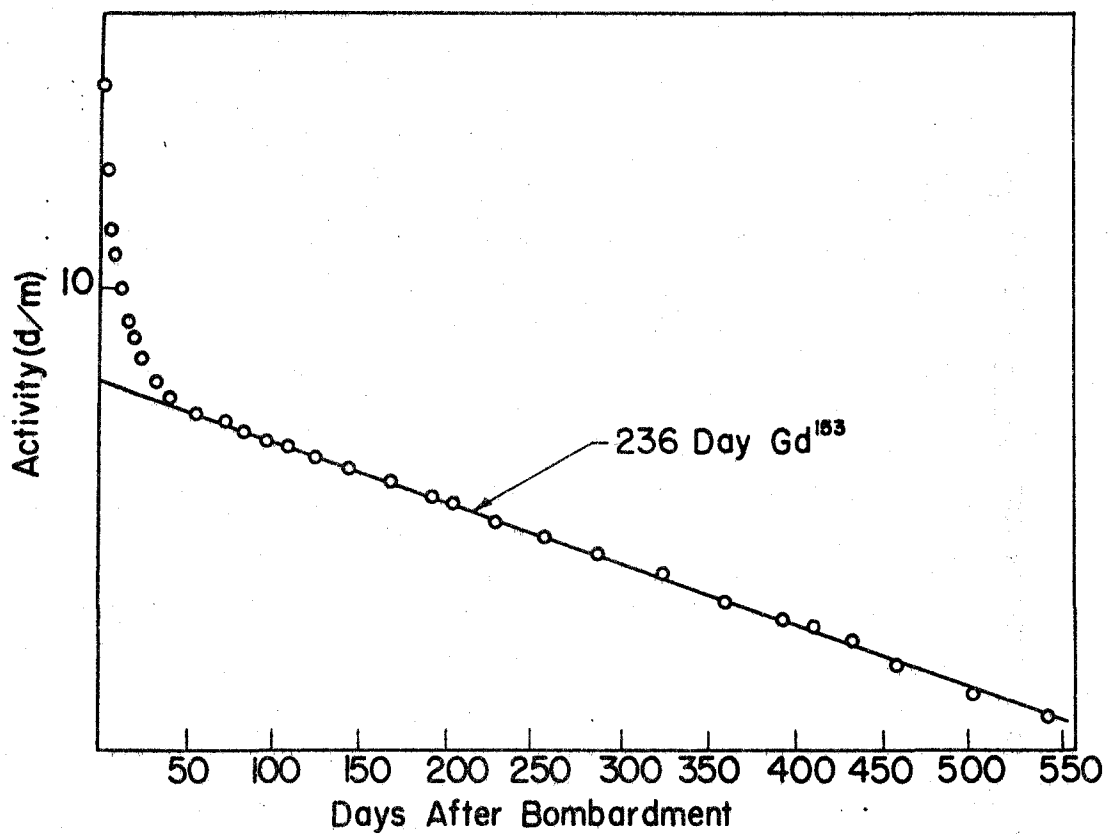


Fig. 15. Decay Curve for Gd^{153} from Neutron Irradiated Gd_2O_3 . Measurements Taken with 225 mg/cm^2 of Aluminum Present.

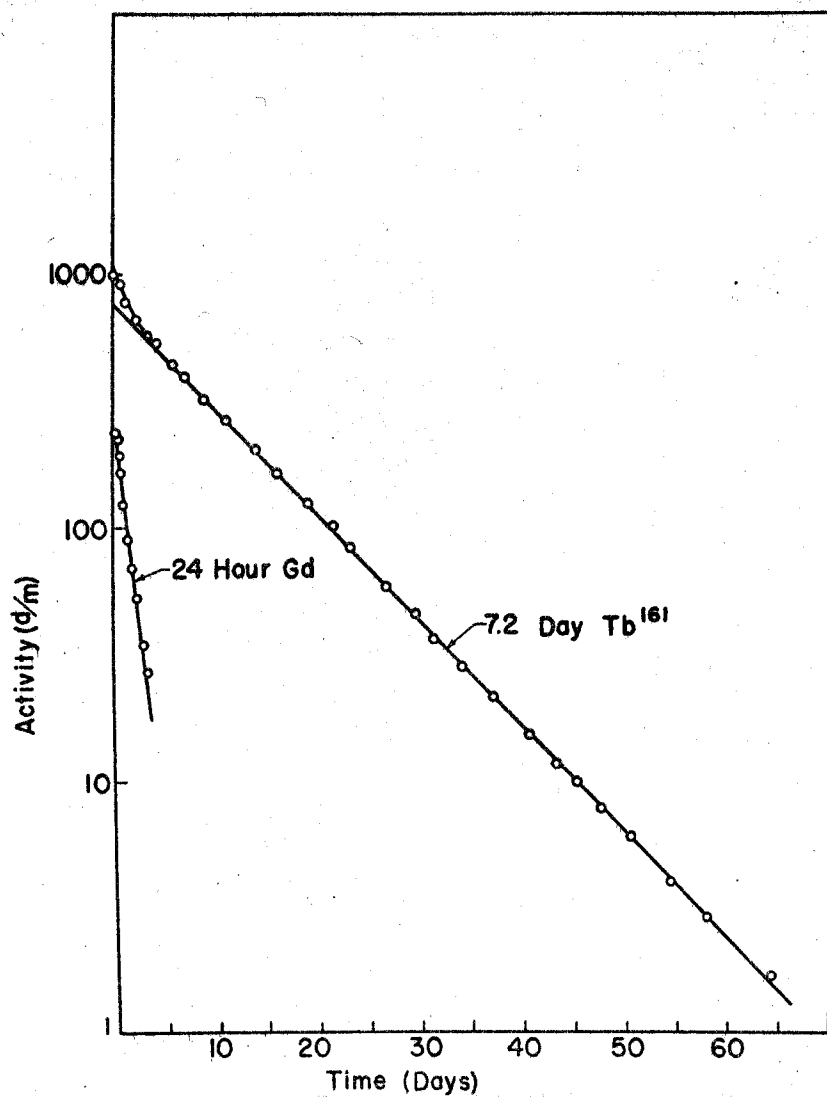


Fig. 16. Decay Curve of Short-lived Components in Neutron Irradiated Gd_2O_3 .

The 24 hour activity and the 7.2 day activity present in the Gd_2O_3 irradiated for 854 hours were separated on an ion exchange column (see Figure 17). As the separation was completed eight days after the sample was removed from the pile, the short-lived component had decayed to a low intensity. These activities cannot be assigned to the same element as the 7.2 day activity preceded the other off the column. Furthermore as the 7.2 day activity did not grow back into the short-lived activity a parent-daughter relationship does not exist with this pair. The assignment of these activities will be discussed later. Suffice it to say that the experiment corroborates the assignment of the 24 ± 3 hour and 7.2 day activities to isotopes of gadolinium and terbium respectively.

Another sample of Gd_2O_3 was bombarded at the Oak Ridge pile for one week. This bombardment was procured for two reasons: first, to check the separation and half-lives of the short-lived activities and second to test for the presence of Sm^{153} . The latter nuclide with a 47 hour half-life was suspected since samarium was a known impurity in the Gd_2O_3 and the slow neutron capture cross section for the process $Sm^{152} (n, \gamma) Sm^{153}$ is large, reported as 280 barns (38). The 24 ± 3 hour activity was thought to be a combination of 47 hour Sm^{153} and 18 hour Gd^{159} .

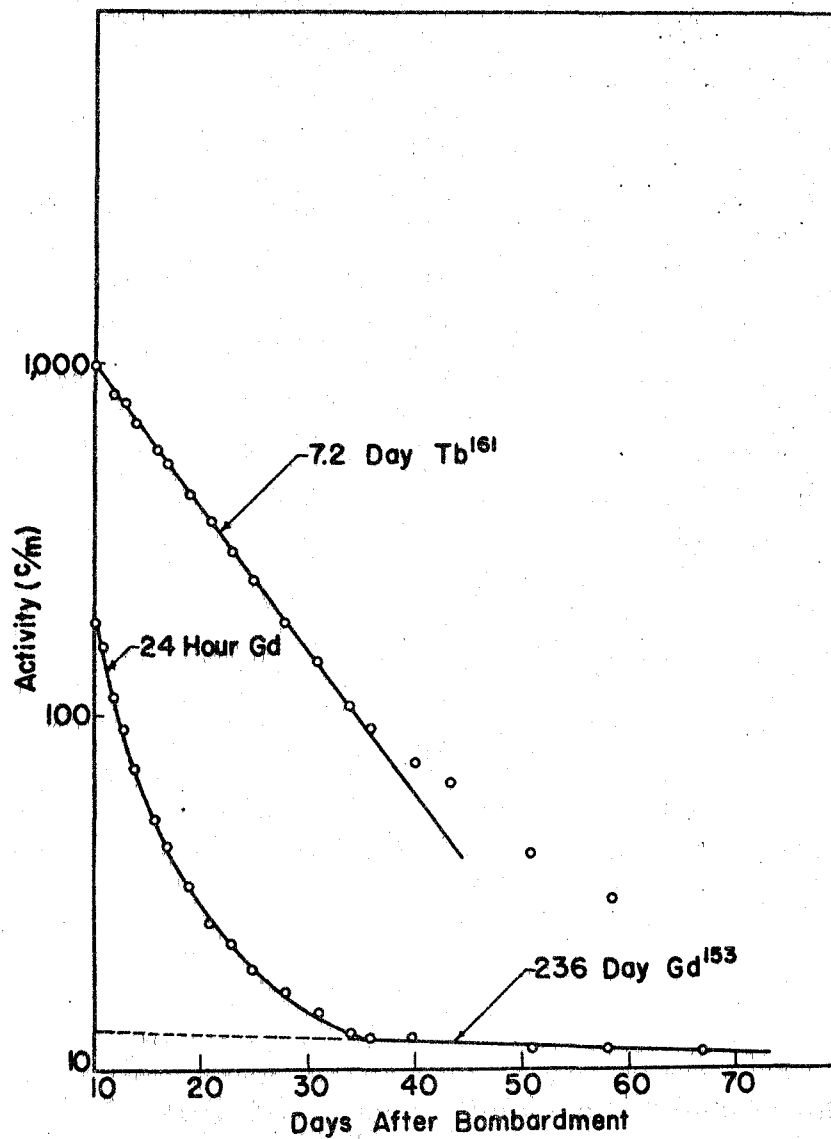


Fig. 17. Decay Curves for 24 Hour and 7.2 Day Activities Separated on Cation Exchange Column.

As was mentioned earlier, sodium amalgam extraction of solutions of irradiated gadolinium did show the presence of Sm^{153} both by half-life and energy measurements. After separation of the terbium and gadolinium fractions on an ion exchange column, samples were mounted for half-life measurements. The terbium decayed with a half-life of 7.2 days. The gadolinium activity after the separation of Sm^{153} showed a half-life of 20 hours. Again no growth of the 7.2 day activity into the gadolinium activity was observed.

Critical Absorption Measurements

Critical adsorption measurements were described in detail in the identification of the Eu K x-ray present in the gadolinium produced by deuteron bombardment of europium. If the x-ray present in the decay of the 236 day neutron induced gadolinium activity is an Eu x-ray, the activity must be Gd^{153} . The possibility of an $(n, 2n)$ reaction producing Gd^{151} is improbable with pile neutrons at least to the extent that the 236 day activity was produced.

Absorbers of cerium and praseodymium oxides in an ether-paraffin solution were dried in aluminum cups giving a thin adhering layer. Absorbers of 20, 40, 60, 90, 120, 150 and 180 mg/cm^2 of the rare earth oxide were placed in reproducible positions between the sample and counter.

The ratio of the Eu K x-ray half-thickness in cerium and praseodymium is $1/4.7$ (see Table VI). Experimentally the ratio was $1/2.1$. As given earlier, the same ratio for gadolinium from deuteron bombarded europium was $1/2.2$. As was stated earlier the subtraction of the more energetic γ -ray cannot be made so the experimental ratio approaches 1 rather than $1/4.7$. Since this experimental evidence identifies the x-ray as originating in an europium nucleus, the 236 day activity is assigned to Gd^{153} .

Energy Measurements

Copper, tin and lead absorption curves on Gd^{153} indicated that a 106 Kev γ -ray was present in addition to the europium K x-ray. The copper curve and subtraction are plotted in Figure 18. This measurement was made on the peak fraction of gadolinium purified by ion exchange methods (see Figure 13). Tin and lead curves gave a similar value for the energy of the gamma ray.

As the K conversion electrons from the 106 Kev γ have an energy of about 58 Kev, these electrons are not detected under ordinary counting conditions. Europium L x-rays and L conversion electrons are present in the decay of Gd^{153} . An absorption curve with the electrons magnetically deflected from the counter gave a component after subtraction of the K x-rays with a half-thickness of 6.3 mg/cm^2

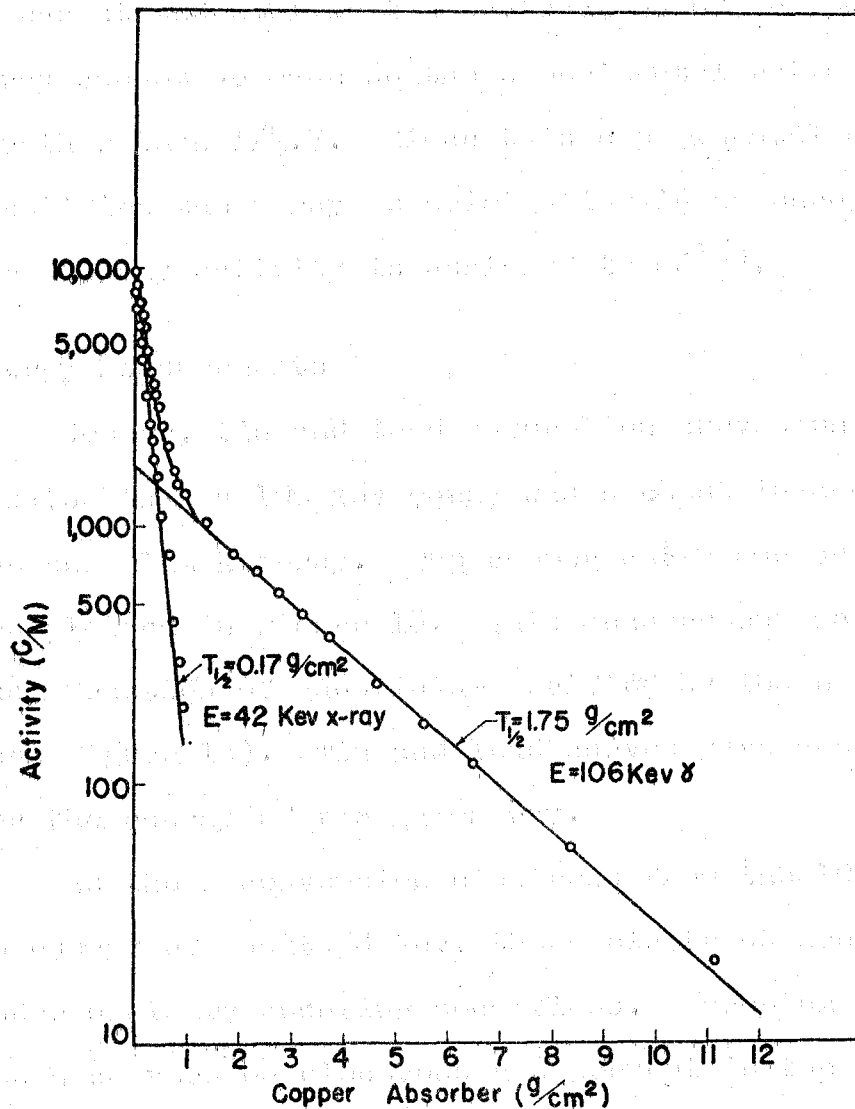


Fig. 18. Copper Absorption Curve of Gd^{153} Radiations.

in aluminum. This half-thickness corresponds to a 6.5 Kev x-ray which agrees well with an energy of an Eu L x-ray apparently present in high intensity. The magnet was then removed without changing the geometric relation between the counter and sample. The points for this absorption curve were identical with the previous curve except for the first few absorbers. The difference between the readings for low absorber thicknesses obtained by subtracting those with the magnet from those without when plotted linearly versus absorber thickness give the range of the L conversion electrons. The results from this series of measurements are plotted in Figures 19 and 20.

Aluminum absorption curves on a purified fraction of the 7.2 day terbium indicate the activity decays by emission of a 0.50 Mev β (see Figure 21). As determined by copper absorption curves a γ -ray of 0.05 Mev is also present.

A β of 0.82 Mev and γ -rays of 0.05 and 0.42 Mev are associated with the decay of the short-lived gadolinium. As the intensity of the 47 hour Sm^{153} contaminant was greater than the short-lived gadolinium and as the possibility exists that all of the Sm^{153} was not removed by two extractions with sodium amalgam, these energies may not be exact. The energies are in general agreement with those reported by Butement (7) for an 18 hour gadolinium

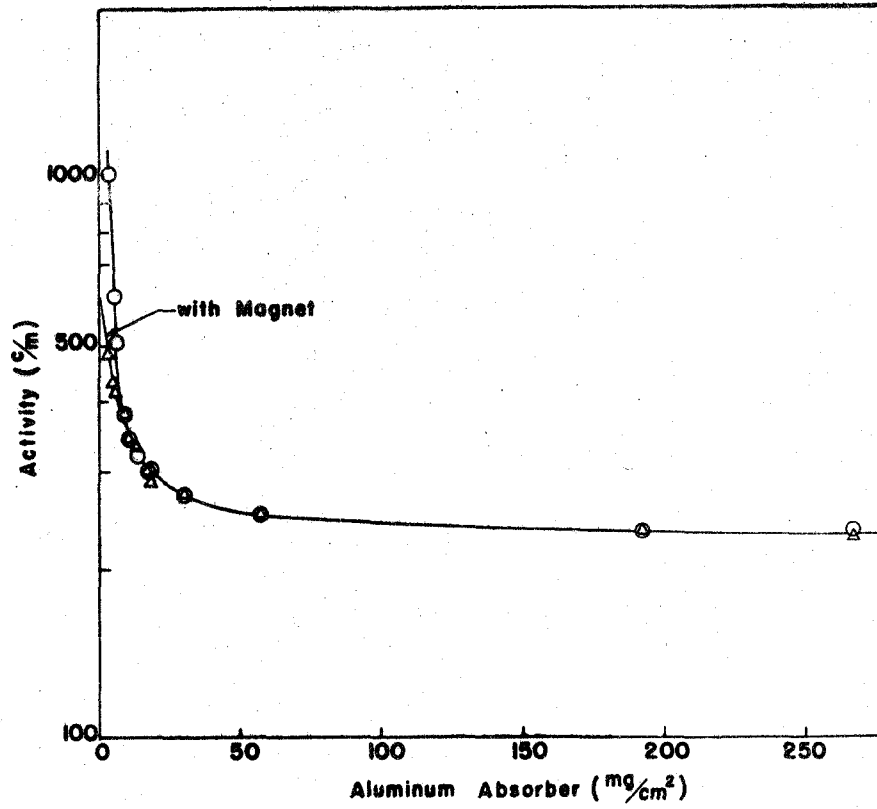


Fig. 19. Gd^{153} Absorption Curves With and Without Magnetic Deflection of the Electrons.

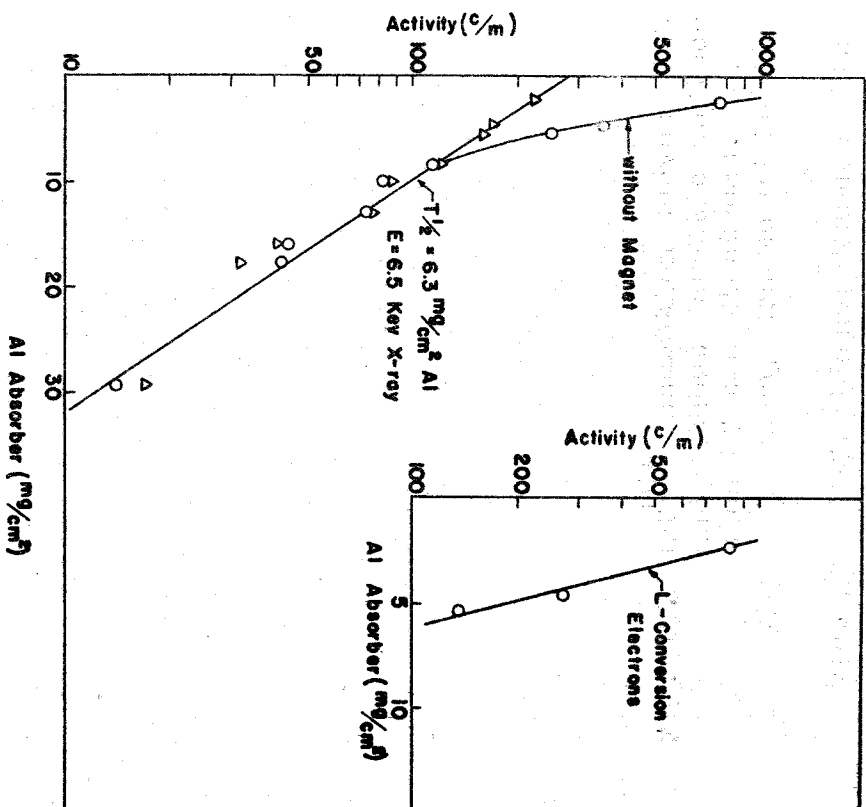


FIG. 20. Absorption Curves of L X-ray and L Conversion Electrons after Subtraction of K X-ray Background (see Fig. 19).

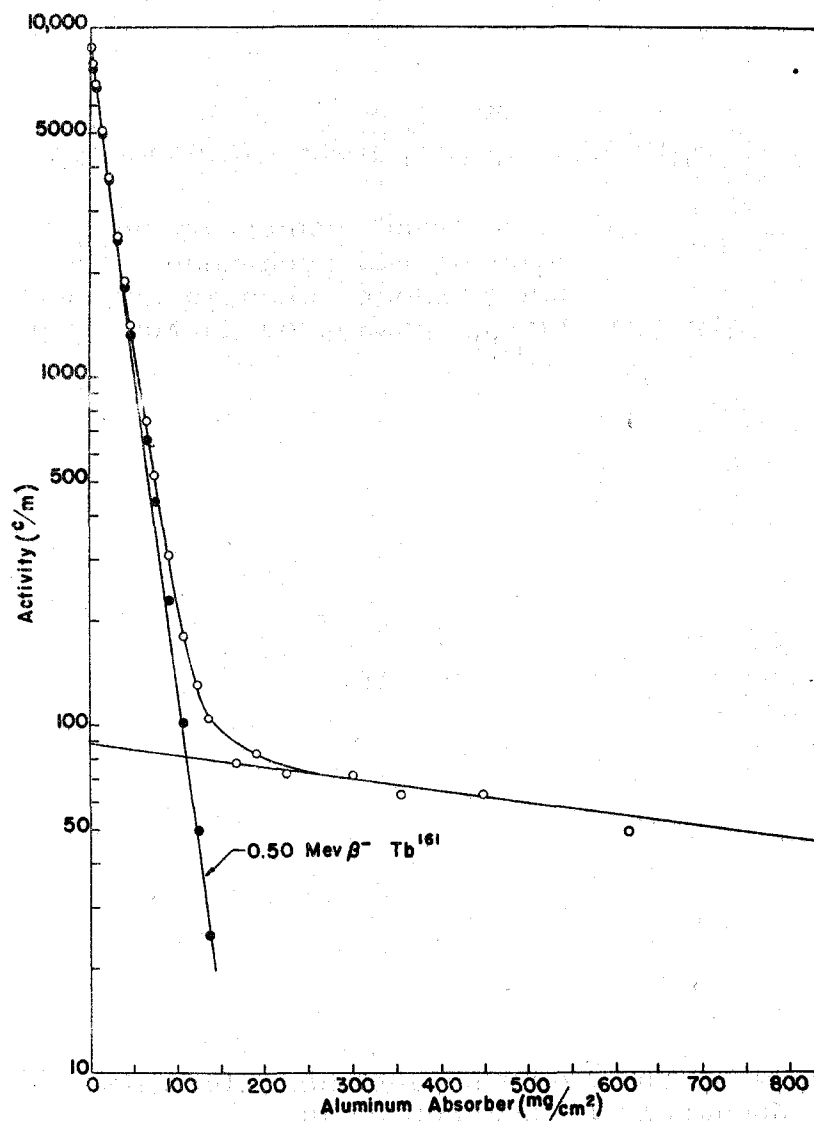


Fig. 21. Aluminum Absorption Curve of 7.2 Day Tb¹⁶¹.

activity decaying by emission of 0.95 Mev β and 0.055 and 0.38 Mev γ -rays.

Decay Scheme of Gadolinium-153.

As given in Figure 22, Gd^{153} decays to an excited state of Eu^{153} by K electron capture and then to the ground state by γ -ray emission. Experimentally the γ -ray energy was determined as 106 Kev in Gd^{153} from neutron bombarded gadolinium and 101 Kev in Gd^{153} from deuteron bombarded europium. It is assumed from these values that this γ -ray is the same as the 102 Kev γ -ray measured spectrometrically (10) and assigned to Gd^{153} .

The highly converted γ -ray gives rise to both K and L electron groups. However, the K conversion electrons were not detected because the low electron energy of 54 Kev does not allow them to go through a thin window. The L x-rays and L electrons comprise the low energy radiation (see Figures 19 and 20). The range of the electrons and half-thicknesses of the γ -rays in aluminum are given in Table X.

The same assembly, variable gate setting, geometry conditions etc. were used in these coincidence determinations as in those on the gadolinium activities produced in deuteron bombarded europium. The Gd^{153} was evaporated on a formvar film (0.10 mg/cm²) and the sample held in place

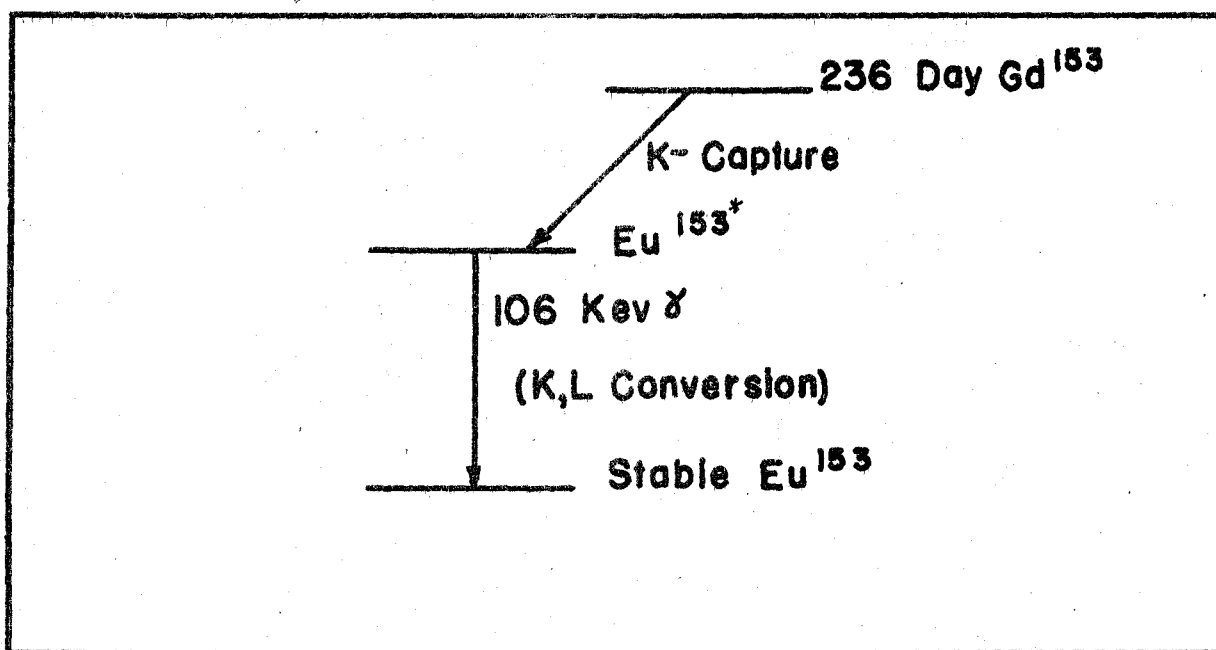


Fig. 22. Decay Scheme of Gd^{153} .

with a zapon film of about the same thickness.

TABLE X
 $T_{\frac{1}{2}}$ and Range of Gd^{153} Radiations in Al

Radiation	Energy	Range or Half-thickness
L conversion electrons	94 Kev	12 mg/cm ²
L x-rays	6.5 Kev	6.3 mg/cm ²
K x-rays	41.5 Kev	1.3 g/cm ²
γ -ray	102 Kev	4.3 g/cm ²

A series of measurements on Gd^{153} were taken with sufficient aluminum in channel 1 to absorb the L x-ray and L electrons. Readings were then taken as aluminum foils were placed between the sample and counter 2 until sufficient aluminum had been added to absorb all L x-rays and L electrons. The experimental results are given in Table XI. A fixed absorber of 52.4 mg/cm² of aluminum was left in channel 1 for all determinations.

The corrected coincidence rate from the last column of Table XI is plotted in Figure 23. Before discussion of the components present in the plot the nomenclature employed is presented. The K x-ray arising from K electron capture is designated as X_K while the K x-ray from the conversion of the 102 Kev γ is designated as K_{γ} . The

TABLE XI
Coincidence Measurements on Gd^{153}

mg/cm ² on 2	(c/m) ₂	(c/m) ₁	Coin.Rate uncorr.	N Acc/min	N B/min	Coin.Rate corr.
0	4500	476	2.219 \pm .037	0.143	0.139	1.937 \pm .038
2.3	2500	476	1.233 \pm .022	0.082	0.138	1.012 \pm .023
3.0	2164	473	1.043 \pm .028	0.068	0.138	0.837 \pm .028
6.0	1098	471	0.573 \pm .019	0.034	0.137	0.402 \pm .019
7.7	845	472	0.455 \pm .017	0.027	0.137	0.291 \pm .017
10.7	685	467	0.396 \pm .016	0.021	0.136	0.239 \pm .016
15.4	606	473	0.333 \pm .013	0.019	0.135	0.179 \pm .013
26.2	535	466	0.268 \pm .011	0.017	0.133	0.118 \pm .011
52.4	511	463	0.264 \pm .010	0.016	0.128	0.120 \pm .010

coincidences that can occur with absorber thicknesses of 52.4 mg/cm² are $K_X X_K$, $X_K K_X$, $X_K \gamma$ and γX_K as based on the decay scheme given in Figure 21 and ranges of radiations as given in Table X. Writing the coincidences between the γ -ray and K x-ray as γX_K signifies the simultaneous observance of a γ -ray in channel 1 and K x-ray in channel 2. As the radiations are not absorbed by the aluminum present on either channel, $X_K \gamma$ coincidences can also be observed. After subtraction of these coincidences and replotting the data, another straight line group is present with a half-

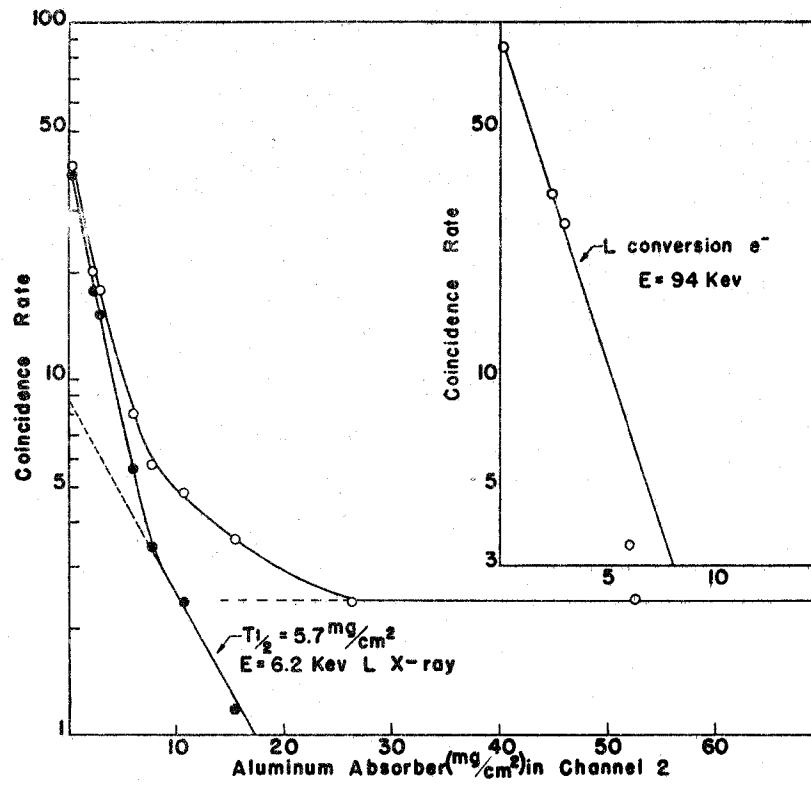


Fig. 23. Coincidence Measurements on Gd^{153} .

thickness of 5.7 mg/cm^2 . This half-thickness corresponds to a 6.2 Kev x-ray and this component is due to L x-ray and X_K coincidences.

After this component is subtracted, a line with a half-thickness of 1.4 mg/cm^2 of aluminum can be drawn through the remaining four points. This half-thickness corresponds to the $9/4$ Kev electron group and these points arise from L electron and X_K in coincidence. Since these components can be explained on the basis of the tentative decay scheme, the decay scheme is substantiated by this experiment.

The decay scheme is further corroborated by another series of measurements. In this experiment, aluminum foils were added to channel 2 with no absorbers between the sample and counter 1. After the corrected coincidence rate was plotted (see Figure 24), a straight line component was present due to $X_K X_K$, $K_X X_K$, $X_K \gamma$, γX_K , $L_X X_K$ and $L_e X_K$ coincidences.

After subtraction of this component (see Figure 24) a straight line with a half-thickness of 6.5 mg/cm^2 can be drawn through another set of coincidence points. This half-thickness corresponds to the L x-ray and the coincidences are between $L_e L_X$ and $X_K L_X$ radiations.

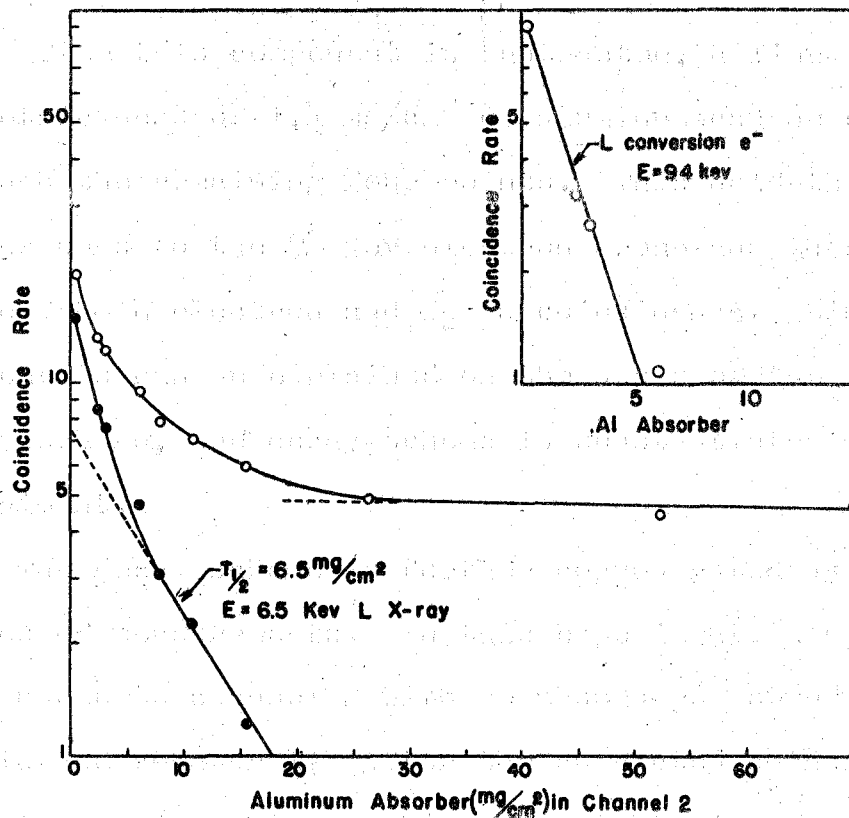


Fig. 24. Coincidence Measurements on Gd^{153} .

After subtraction of this component a straight line can be drawn through a third group of coincidences involving L electrons being counted in channel 2. The coincidences are due to $X_K L_\alpha$ and $L_X L_\alpha$ radiations

Another determination with sufficient copper in both channels to absorb everything except the 102 Kev γ -ray did not give a measurable coincidence rate above background and accidentals. Also the L conversion electrons are the only electron group present as $N_{e\gamma}/N_e$ was a constant.

On the basis of these experimental results, the tentative decay scheme of Gd^{153} as proposed earlier (see Figure 22) is thus verified.

Neutron and Deuteron Bombarded Europium

Survey of Previous Work

As a survey of the pertinent work concerning the long-lived europium activities was given earlier, only a few statements will be made before the experimental work will be discussed. Long-lived neutron induced activities in europium (17) are assigned to Eu^{152} (5.3 yr) and Eu^{154} (5.4 yr). The γ -radiations are quite complex with about 33 conversion electron lines present in a mixture of the two isotopes (21). One or both of the nuclides decay to an unknown extent by K-electron capture (16). No satis-

factory decay scheme has been advanced as yet and will probably have to wait until enriched isotopes are available.

Purification of Activities

At the time the studies were started, much of the information just given was not known. The question of whether Eu^{152} or Eu^{154} might be formed preferentially in a neutron or deuteron bombardment of europium appeared worth investigating. If such were the case, a comparison of activities produced by the two methods could be of value in assigning half-lives and energies to the particular isotopes involved.

Samples of Eu_2O_3 , originally separated and purified by H. N. McCoy, on spectrographic analysis indicated trace amounts of samarium and gadolinium were present. One sample was given 870 microampere hours of bombardment with 20 Mev deuterons by the Berkeley 60-inch cyclotron. The other sample received a pile neutron bombardment at Oak Ridge. Both samples were again purified after bombardment by a succession of EuSO_4 precipitations. The complete chemical process is outlined earlier in connection with the separation of gadolinium and europium.

Half-life Measurements

Half-life determinations with an electroscope now extend over 550 days (see Figure 25). The europium activities produced by deuteron bombardment were measured with and

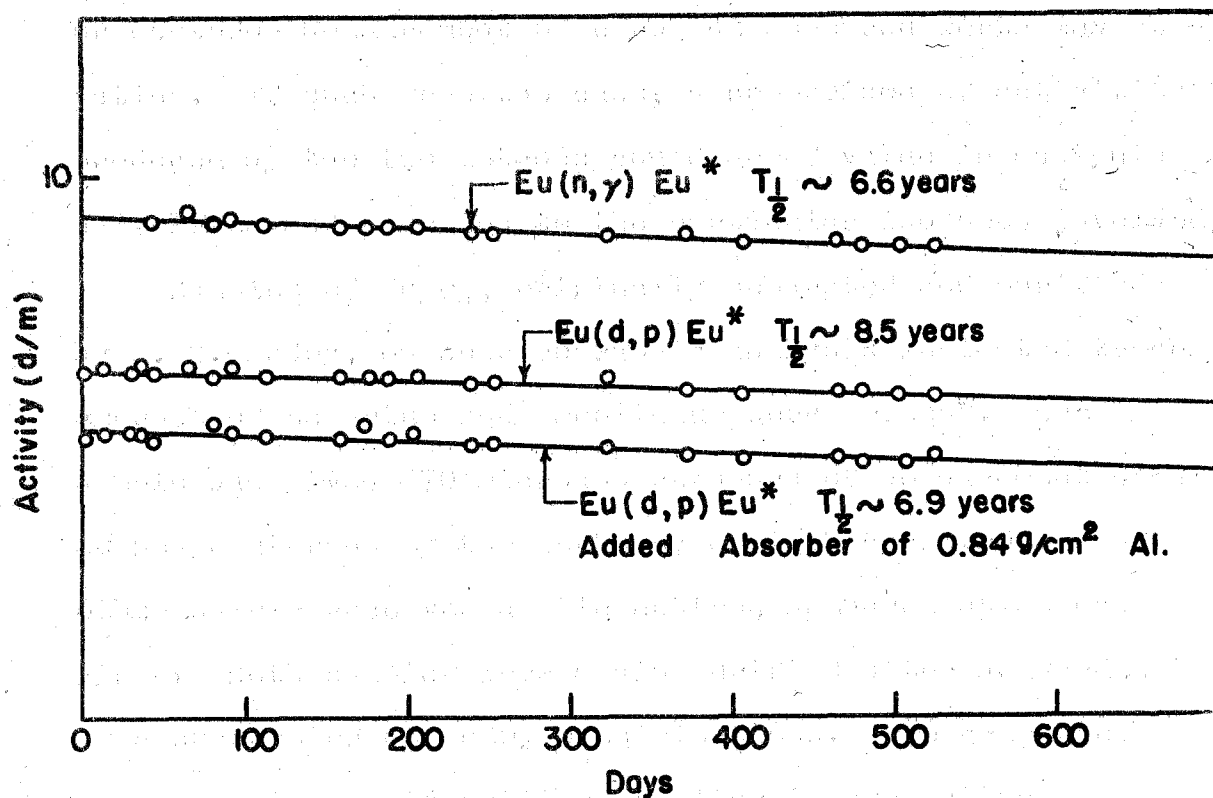


Fig. 25. Decay Curves for Eu^{152} - Eu^{154} Activities from Neutron and Deuteron Irradiated Eu_2O_3 .

without absorbers giving the values of 6.9 and 8.5 years respectively. The radiations from neutron induced europium without absorber present decays with a half-life of 6.6 years. These results are of course a composite of Eu^{152} and Eu^{154} half-lives and the measurements have not continued long enough to give values which are valid. The results are believed to be good within 20%.

Energy Measurements

Absorption curves of the radiation in lead, copper and aluminum do not yield much valuable information for these nuclides since the combined activity is so complex that only a rough average of the energy in each energy region is obtained. The value for the gamma radiations from lead absorption curves was 1.4 Mev and that for the beta radiations by aluminum absorption curves was 0.9 Mev in agreement with published absorption data (42).

As a comparison of the neutron and deuteron produced activities, aluminum absorption curves were run on both under identical geometry. It can be seen from Figure 26 that the two curves are superimposable. Thus there is not enough difference in the relative amounts of Eu^{152} and Eu^{154} as produced by the two different methods to be observed in the aluminum absorption curves. In a similar way the lead absorption curves are essentially identical in the two cases.

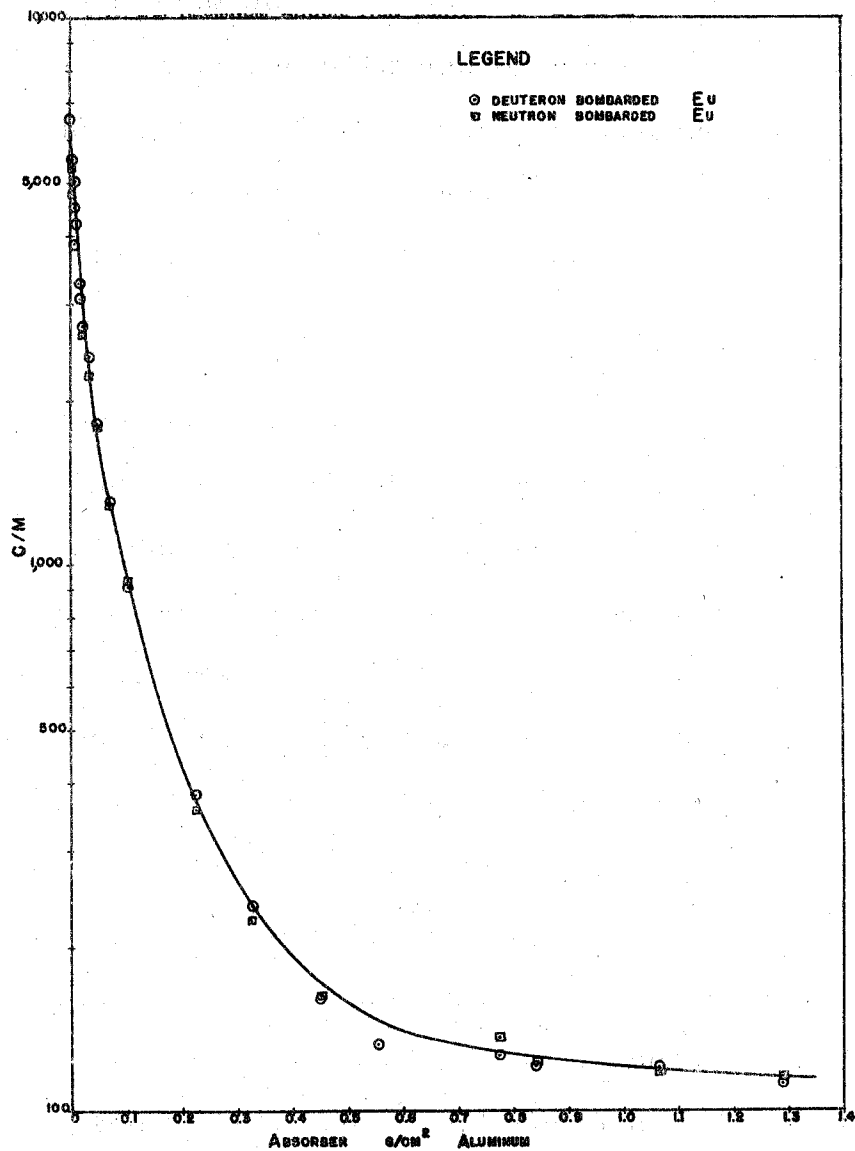


Fig. 26. Absorption Curves on Eu^{152} - Eu^{154} from Neutron and Deuteron Irradiated Eu_2O_3 .

DISCUSSION

Interpretation of Data

Mass Assignment of Gadolinium Activities

A 236 day activity produced in neutron irradiated gadolinium is assigned to Gd^{153} . The activity decays by K electron capture and the resulting x-ray has been identified as the europium K x-ray. As Gd^{151} and Gd^{153} are the only nuclides that can decay to a stable europium the choice is thus limited. The (n, 2n) reaction necessary to produce Gd^{151} is not likely with pile neutrons.

Based on similarity of half-lives and γ -ray energies, one of the long-lived gadolinium activities produced in deuteron bombarded europium appears to be the same as that assigned to Gd^{153} . The assignment of the other activity to Gd^{151} is based on the following premises. First, as an europium x-ray is associated with the decay of the 150 day activity, the possible choices are limited to Gd^{151} and Gd^{153} . Second, the assignment of the 150 day activity to Gd^{153} is improbable since the activity was not observed in neutron irradiated gadolinium. Possibly Gd^{151} could also be produced by high energy γ -radiation of gadolinium as well as fast neutron bombardment thus providing a check on these observations.

The half-life of 150 days for Gd^{151} is a calculated value based on changes in the ratios of γ -ray intensities over a period of time. As has been stated earlier the 102 Kev γ -ray is associated with the decay of Gd^{153} and the 265 Kev γ -ray with Gd^{151} . With a period of 590 days intervening copper absorption curves give a ratio of intensities of the 102 Kev γ -ray to the 265 Kev γ -ray of 2.9 at $t = 0$ and 8.1 at $t = 590$ days. The following equations can then be solved for the half-life of Gd^{151} .

$$\text{at } t = 0 \quad \lambda_{153} N_{153} = 2.9 \lambda_{151} N_{151}$$

$$\text{at } t = 590 \quad \lambda_{153} N'_{153} = 8.1 \lambda_{151} N'_{151}$$

$$\lambda_{153} N'_{153} = \lambda_{153} N_{153} e^{-\lambda_{153} t}$$

$$\lambda_{151} N'_{151} = \lambda_{151} N_{151} e^{-\lambda_{151} t}$$

$$\therefore e^{-\lambda_{151} t} = 2.9 e^{-\lambda_{153} t} / 8.1$$

$$\text{or } T_{1/2} = 150 \text{ days}$$

The calculated half-life agrees well with what would be expected from the overall decay curves of the gadolinium activity from deuteron bombarded europium (see Figure 4). Apparently the process $Eu^{153}(d, 2n)Gd^{153}$ occurs to about 5% of the $Eu^{151}(d, 2n)Gd^{151}$ process.

In cation exchange experiments, the 7.2 day activity is eluted just prior to the gadolinium fraction. The relative position of the activity peaks in the elution curve would indicate the 7.2 day activity is a terbium isotope.

Energy measurements on the 7.2 day activity are in agreement with those reported for a 5.5 to 7 day activity assigned to Tb^{161} by other investigators. The Tb^{161} by virtue of its relationship with stable nuclides must have a gadolinium parent. As the 7.2 day activity tentatively assigned to Tb^{161} did not grow back into the 18 to 24 hour gadolinium fraction from which it was separated, this gadolinium nuclide cannot be the parent. This would indicate the 18 to 24 hour activity is Gd^{159} . Other investigators have assigned a 3.6 minute activity to Gd^{161} .

Limitations of Studies

The energies of radiation associated with gadolinium activities were determined by absorption in various elements. The limitations of this method are well known in that similar energy levels cannot be differentiated. Highly erroneous values may result in γ -ray determinations with elements having a K absorption edge slightly less than the energy of the γ -ray in question. The ranges of the electron groups were estimated visually.

The half-life values for Gd^{151} and Gd^{153} are believed to be accurate as the electroscope was extremely stable over the period of measurement. The 24 ± 3 hour value for Gd^{159} is in doubt because of possible Sm^{153} contamination and the usual difficulty in measuring short-lived activ-

ties at a site removed from the irradiation source. The measurements on the decay of the europium activities have not extended over a long enough time interval to give valid results. The purity of the samples is such as to warrant continued measurements.

SUMMARY

A 236 day neutron induced activity in gadolinium purified by ion exchange methods is assigned to Gd^{153} . A simple decay scheme is proposed in which the Gd^{153} decays to an unstable Eu^{153} by K electron capture and then to the ground state by emission of a 106 Kev γ -ray. The γ -ray is highly internally converted giving rise to K, L, and possibly M electrons.

A 7.2 ± 0.2 day and 18 to 24 hour activity present in the neutron irradiated gadolinium were separated on an ion exchange column. The 7.2 day activity assigned to Tb^{161} decays by emission of 0.50 Mev β and 0.05 Mev γ . The 18 to 24 hour activity tentatively assigned to Gd^{159} decays by emission of 0.82 Mev β and γ -rays of 0.05 and 0.42 Mev.

Half-life measurements on gadolinium produced by deuteron bombardment of europium extend over 600 days. The activity decaying with a half-life of 155 days initially is slowly lengthening into a longer half-life. A 265 Kev γ -ray is detected in addition to the 106 Kev γ -ray previously found in Gd^{153} . The 265 Kev γ -ray is assigned to Gd^{151} which decays with a half-life of 150 days. This half-life is based on ratios of the 106 Kev γ -ray to the 265 Kev γ -ray observed over a 600 day period. The decay scheme of Gd^{151} appears to be very similar to Gd^{153} with only the 265 Kev γ -ray present in the decay.

Energy measurements on Eu^{152} and Eu^{154} produced by neutron and deuteron bombardment of Eu_2O_3 give highly similar results, indicating that the same ratio of isotopes is produced in the two irradiations. Half-life measurements on the combined samples of Eu^{152} and Eu^{154} extend over 550 days giving values of 6.6 to 8.5 years.

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