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July, 1957

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University of California, Berkeley, California

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ABSTRACT

A series of bombardments using alpha particles from the Berkeley 60-inch cyclotron on rare earth oxides has resulted in the discovery and mass assignment of two new alpha-emitting isotopes, Dy^{153} (5 hr) and Dy^{154} (13 hr). Two other alpha emitters have been mass assigned, Dy^{152} (2.3 hr) and Tb^{151} (19 hr). A new 10 hr electron-capture isotope has been identified as Dy^{155}. Evidence is also presented for the discovery of another isotope, Dy^{149} (8 min), which was produced by a N^{14} ion bombardment on praseodymium.

*This work was done under the auspices of the U.S. Atomic Energy Commission.

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INTRODUCTION

In 1955 Rasmussen et al. reported on a detailed study of neutron-deficient isotopes in the rare earth region.[1] These isotopes exhibited alpha radioactivity. A number of such nuclides were discovered and studied individually.

Among the alpha-emitting nuclides reported were three dysprosium isotopes, whose alpha energies and half-lives were as follows:

(a) 4.2 ± .06 Mev, 7 ± 2 min.

(b) 4.06 ± .04 Mev, 19 ± 4 min.

(c) 3.61 ± .08 Mev, 2.3 ± .2 hr.

A limit was set on the mass numbers of the dysprosium activities, $153 \geq A \geq 149$.

The study presented here was begun with the intent of mass-assigning the 2.3-hr activity, using alpha particles from the Berkeley 60-inch cyclotron. When the experiment was performed, new information was uncovered, which stimulated further work in this region. This paper is concerned with the study of hitherto unreported dysprosium activities and additional information that has been found in connection with previously known rare earth nuclides.

Table I summarizes the information available on the new isotopes. Fig. 1 is a section of the isotope chart, which shows the nuclides that have been studied and used in the investigation.

Table I

Information on New Isotopes

Isotope	Half-life	Mode of decay seen	Alpha-particle energy (Mev)
Dy^{149}	8 min ± 2	E.C. and/or β^+	
Dy^{153}	5 hr ± 0.5	α	3.48 ± .05
Dy^{154}	13 hr ± 2	α	3.35 ± .05
Dy^{155}	10 hr	E.C.	

EXPERIMENTAL METHOD

In the work reported here, elements of atomic number Z were bombarded with alpha particles in the Berkeley 60-inch cyclotron to produce isotopes of elements Z + 2 by means of (α,xn) reactions. One experiment was carried out using a heavy ion (N^{14}) as the bombarding particle in the same cyclotron. The energy of the alpha particles was varied between 48 Mev and 20 Mev to give different values of x.

Materials used in this investigation were

(a) gadolinium enriched in Gd^{152},
(b) " " " Gd^{154},
(c) natural gadolinium,
(d) " europium,
(e) " praseodymium.

Table II gives the isotopic percentages of the three gadolinium oxides. Europium has two stable isotopes, Eu^{151} and Eu^{153}, in almost equal abundance. Praseodymium has only one stable isotope, Pr^{141}. The elements were all bombarded as the powdered oxides. The two enriched gadolinium oxides will be referred to as Gd^{152} and Gd^{154} in further discussions.

Table II

Isotopic Abundances of Target Gadolinium

	152	154	155	156	157	158	160
Enriched Gd^{152}	14.96	9.75	27.26	19.32	10.08	11.67	6.97
Enriched Gd^{154}	.32	33.17	38.57	15.92	5.49	4.50	2.05
Natural Gd	.20	2.15	14.73	20.47	15.68	24.87	21.90

The rare earths were separated from each other by an ion-exchange method described elsewhere.[2] The eluting agent was 0.4 M alpha-hydroxy-isobutyric acid, buffered to a pH of 3.90 with ammonium hydroxide. Separations were very satisfactory. The activity, after being eluted, was evaporated to dryness on platinum plates. The latter were then flamed to destroy the organic eluant and to make the samples essentially weightless for alpha-particle counting.

Two of the experiments were carried out using a stacked-foil technique. The oxide was glued onto aluminum foils with a solution of "Duco" cement in amyl acetate. Four such plates were prepared in each case, and were stacked with other aluminum absorber foils to give required energies on the target plates. After irradiation no chemical separations were made. The aluminum plates were alpha-counted directly because it was assumed that the aluminum would not form any alpha emitters. The alpha activity seen would belong to the rare earths produced in the course of the bombardment.

Alpha activity was detected in these experiments by counting samples in an argon flow-type ion chamber. Alpha-particle energies were determined by use of a 48-channel differential pulse-height analyzer.[3]

RESULTS

Dy^{152} and Dy^{153}

Gadolinium-152 was bombarded with 48-Mev alpha particles. After irradiation the material was chemically separated by the procedure described in the previous section. The dysprosium fraction was evaporated on a platinum plate and counted in the 48-channel alpha-pulse analyzer. Two peaks appeared.

The peaks were integrated and their half-lives were determined. The one with the higher energy decayed with a 3-hr half-life, while the other peak decayed with a 6-hr half-life. Gadolinium-148 (3.16 Mev), Np^{237} (4.79 Mev) and Pu^{239} (5.15 Mev) were used as alpha energy standards. The two dysprosium peaks were in this manner measured to be at 3.66 Mev and 3.48 Mev.

The nuclide with an alpha-particle energy of 3.66 Mev and a 3-hr half-life certainly was the same isotope as reported by Rasmussen et al.[1] The other alpha emitter was recognized as a new activity. The alpha spectrum with the two activities is shown in Fig. 2.

To mass-assign both of the activities an experiment was carried out using the stacked-foil technique described in the section on experimental methods. The energies of the alpha particles striking each of the four target foils are listed below, together with the most probable reactions expected at these energies.

Target 1 48 Mev $(\alpha,4n)$, $(\alpha,3n)$
" 2 41.5 Mev $(\alpha,3n)$, some $(\alpha,4n)$, some $(\alpha,2n)$
" 3 33.5 Mev $(\alpha,2n)$, some $(\alpha,3n)$
" 4 23.3 Mev $(\alpha,2n)$, (α,n)

The target foils were counted directly in the alpha pulse analyzer and in an alpha counter. The alpha-counter results were as follows:

Target 1, 2.5-hr half-life, with a 17-hr "tail"
" 2, 5-hr half-life, with a 15-hr "tail"
" 3, scattered counts
" 4, no noticeable alpha activity.

The targets were thick because no chemical separations were made, and no distinct peaks were observed in the pulse analyzer. The counts on various channels were summed and plotted against time on semilog paper. Results were essentially the same as given above. Since the targets were not counted for as long a time on the pulse analyzer as on the alpha counter, the tail, mentioned above, was not seen on either Target 1 or Target 2. Counts on Target 3 in this case were observed to decay with a 5-hr half-life.

The 2.5-hr isotope was seen in the first target foil at 48 Mev but was absent at 41.5 Mev. At least, it was not present in a sufficient amount to be noticed. The $(\alpha,4n)$ reactions, in this region, have thresholds at about 39 Mev.

It seems quite reasonable to assume then that the activity was produced by an $(\alpha,4n)$ reaction on Gd^{152} and must be Dy^{152}. The 5-hr isotope was present only in a small amount at 33.5 Mev and was absent at 23.3 Mev. Since $(\alpha,3n)$ thresholds are approximately at 28 Mev, one is forced to the conclusion that the 5-hr alpha emitter must have been made by an $(\alpha,3n)$ reaction on Gd^{152}. It must be Dy^{153}.

By analogy, the $(\alpha,5n)$ threshold must lie in the neighborhood of 48 to 50 Mev. It would be quite improbable for an $(\alpha,5n)$ to occur at 48 Mev and account for a large amount of activity. It follows that the 2.5-hr activity could not be Dy^{151}. The two isotopes discussed here could have been produced by $(\alpha,4n)$ and $(\alpha,3n)$ reactions on Gd^{154}. However, bombardments of that nature have been carried out and have resulted in new activities. The bombardments are discussed in the next sections.

Dy^{154}

It was thought that the 16-hr activity observed in the stacked-foil experiment described above could have been Dy^{154}. Therefore, a series of experiments was carried out in which Gd^{154} was irradiated with alpha particles. The discussion in the present subsection is limited to the alpha radioactivity that resulted.

After a full-energy bombardment the material was chemically separated and the dysprosium fraction studied in the alpha-pulse analyzer. Alpha activity was low, but three peaks of comparable heights were recognizable. The peak with the highest alpha energy decayed with a 2.5-hr half-life. Presumably the activity must have been Dy^{152}, produced by an $(\alpha,4n)$ reaction on Gd^{152}, an isotope which constituted 0.3% of the bombarded material. The peak with the next-to-highest alpha energy had a 4.5-hr half-life. This was thought to be Dy^{153}, made by an $(\alpha,3n)$ reaction on Gd^{152}. The third peak decayed with a 13-hr half-life. Since no such third peak was in evidence in the Gd^{152} bombardments, the new alpha emitter could not have been an isomer of either Dy^{152} or Dy^{153}. Rather, it had to be Dy^{154} made by an $(\alpha,4n)$ reaction on Gd^{154} (33% of the bombarded material). Of course, Dy^{155} could not be ruled out as a possibility. However, if the new activity were Dy^{155}, this would place an alpha emitter, (with partial half-life surely less than 10^5 yrs by yield considerations) just one mass number away from naturally-occurring Dy^{156}.

Placing the new activity as Dy^{154} relieves the situation somewhat, although there is evidently a very unusual discontinuity in the trend of partial alpha half-lives in going from Dy^{154} to Dy^{156}. Porschen and Riezler[4] searched for and failed to find alpha activity in natural dysprosium. From their study one can say that the alpha half life of Dy^{156} is greater than 5×10^{16} yrs. The alpha decay rate discontinuity between Dy^{154} and Dy^{156} may be the result of a discontinuity in decay energy, in the specific rate dependence on energy, or on both. Perhaps the discontinuity is related to the abrupt nuclear structural changes between 88 and 90 neutrons in neighboring lower rare earths, as reflected in first excited state energies and spectroscopic isotope shifts of even-even nuclei and of quadrupole and magnetic moments in Eu^{151} and Eu^{153}.

The experiment was repeated. The dysprosium fraction, in this case, was counted for the first time in the pulse analyzer a day after bombardment time. Only two peaks were observed. Their half-lives were 5 hr and 13 hr. The peak with the longer half-life dominated the spectrum. The pulse analysis curve is shown in Fig. 3. With the 5-hr activity (3.48 Mev) as one standard and Gd^{148} (3.16 Mev) as the other standard, the 13-hr peak was calibrated and found to be at 3.37 ± .04 Mev.

The same material was irradiated at 37 Mev. As the energy was below the $(\alpha,4n)$ threshold, only the 5-hr peak was visible. Dysprosium-152 and Dy^{154} would not be produced by $(\alpha,4n)$ reactions on Gd^{152} and Gd^{154} respectively. Dysprosium-154 could have been made by an $(\alpha,2n)$ reaction on Gd^{152}. One would not expect to produce a sufficient amount of alpha activity by this means, because the nuclide in question must have an exceedingly low alpha-branching ratio. The reason that one expects a low branching ratio is that Dy^{152} and Dy^{153}, though produced from 0.3% of the bombarded material, accounted for more alpha activity than did Dy^{154}, this in spite of the fact that the latter isotope was made from 33% of the bombarded oxide. The new alpha emitter could not have been Dy^{155}. This isotope could have been made in a large quantity by an $(\alpha,3n)$ reaction on Gd^{154} at the energy used, but in contrast, the 13-hr alpha emitter was definitely missing.

In a bombardment carried out at 27 Mev, below the $(\alpha,3n)$ threshold, no alpha activity was observed. The result can be explained on the basis of the same kind of arguments as in the previous paragraph. The new 13-hr alpha emitter was in this manner mass-assigned as Dy^{154}.

Dy^{155}

A new 10 hr electron-capture dysprosium isotope has been identified as Dy^{155}. The nuclide was produced by $(\alpha,4n)$ and $(\alpha,3n)$ reactions on Gd^{155} and Gd^{154} respectively. Dysprosium-155 was discovered independently at Oak Ridge by Mihelich and coworkers.[5] They made the isotope by a (p,pn) reaction on Dy^{156}, and they report a half life of 20 hrs. They list a 230 kev gamma transition belonging to Dy^{155} decay.

Our mass assignment of Dy^{155} was accomplished in the following manner: This new isotope has a prominent gamma transition of 225 kev which was found to decay with a 10 hr half-life. The photon was seen in greatest abundance in a 48 Mev alpha bombardment on Gd^{154}. The isotope could be made by an $(\alpha,4n)$ reaction on Gd^{155} (38% of bombarded material) and by an $(\alpha,3n)$ reaction on Gd^{154} (33% of bombarded material). The photon was present in lesser abundance in a 37 Mev bombardment. The energy was below the $(\alpha,4n)$ threshold and the isotope could have been produced only by an $(\alpha,3n)$ reaction on Gd^{154}. This particular information shows that the isotope could not be Dy^{154} since the latter nuclide could not be made below the $(\alpha,4n)$ threshold. The photon was also seen in much less abundance in 48 Mev bombardments on natural gadolinium. Here the isotope could be made by an $(\alpha,4n)$ reaction on 15% of the material bombarded and by an $(\alpha,3n)$ reaction on 2%. The photon was absent in bombardments below the $(\alpha,3n)$ threshold on both Gd^{154} and natural gadolinium. This information shows that the photon could not belong to Dy^{157} (8 hr).

Dy^{149}, Dy^{150} and Dy^{151}

Stable praseodymium was bombarded in the Berkeley 60-inch cyclotron with N^{14} ions to produce dysprosium isotopes by means of (N,xn) reactions. No chemical separation was made because short-lived activities were sought. When the sample was counted on the alpha pulse-height analyzer, only a broad, unresolved peak was observed on account of degradation in the thick sample. The counts on the various channels were summed in groups of five. Decay curves were then obtained by following the change with time in activity of each group of five channels.

The shorter half-lives indicated the presence of the 19 min and 7 min dysprosium alpha emitters,[1] but the two outstanding results were

(a) a 4-hr activity that appeared in all channels, except the higher ones (above channel 35);

(b) a period of time wherein the alpha activity increased rather than decreased. This was again seen on all channels but the higher ones.

The 4-hr activity was quite probably Tb^{149}, an alpha emitter with a decay energy of 3.96 Mev.[1] The activity could have been made originally in the bombardment by an (N,pxn) reaction. It also might have grown in from its dysprosium parent, Dy^{149}. The increase in alpha activity with time indicated that one of the alpha emitters seen in the experiment grew in from a parent. None of the dysprosium isotopes could have grown in because it would have been impossible for any holmium isotopes to be produced by nitrogen ions on praseodymium. Therefore the increase in alpha activity was due to the growth of a terbium isotope. The only terbium nuclide seen was Tb^{149}. Furthermore, some of the decay curves showed the growing-in period followed directly by the 4-hr activity. It would seem reasonable to assume that Dy^{149} was produced originally in the bombardment. The isotope, having 83 neutrons, would be expected to decay primarily by positron emission, electron capture, or both, rather than by alpha decay because the maximum in the rare earth alpha-decay energies occurs at 84 neutrons as a consequence of the decreased neutron binding energies just beyond the closed shell of 82 neutrons. This is in analogy to the maximum at 128 neutrons resulting from the low neutron binding energies just beyond the closed shell of 126 neutrons. The Tb^{149} growth curve indicates the presence of Dy^{149}, which decays to Tb^{149}. The growing-in period and the 4-hr tail are shown in Fig. 4, and from this curve we determine a half life of 8 ± 2 min for Dy^{149}.

The 19-min and 7-min dysprosium alpha emitters, mentioned at the beginning of this paper, have not been definitely mass-assigned. However, because of the evidence for the existence of a new isotope, Dy^{149}, and because the 2.5-hr dysprosium alpha emitter has been shown to be Dy^{152}, it would be logical to assume that the 7-min and 19-min nuclides are Dy^{150} and Dy^{151}. Also, if alpha energy systematics holds in the rare earth region, then the 7-min isotope with an alpha energy of 4.2 Mev must be Dy^{150}, and the 19-min isotope with a 4.06-Mev alpha energy must be Dy^{151}.

Tb^{151}

In 1953 Rasmussen et al. reported a terbium alpha emitter with a half-life of 19 hrs and an alpha energy of 3.4 Mev.[1] Later, Rasmussen and Rollier

tentatively mass-assigned this isotope as either Tb^{150} or Tb^{151}.[6] Handley and Lyon[7] reported that no alpha activity was seen in 14.5- and 22-Mev proton bombardments on Gd^{152}. Going on this evidence, the latest General Electric Chart of Isotopes has the activity listed as Tb^{150}.

In order to mass-assign this alpha emitter a stacked-foil experiment was performed similar to the one described previously. Natural europium was glued to the target foils. The energies of the impinging alpha particles on each target foil, together with the reactions expected to take place at those energies were:

Target 1 48 Mev $(\alpha,4n)$, $(\alpha,3n)$
" 2 40.6 Mev $(\alpha,3n)$, some $(\alpha,4n)$, some $(\alpha,2n)$
" 3 31.2 Mev $(\alpha,2n)$, some $(\alpha,3n)$
" 4 20.2 Mev $(\alpha,2n)$, (α,n)

Alpha radiation was present in only the first two targets. The total amount of alpha activity on the second target was down by a factor of 20 below the amount on the first target. In each case the only half-life seen was 19 hrs

If the 19-hr isotope were Tb^{150}, it would have had to be made by an $(\alpha,5n)$ reaction on Eu^{151}. That this reaction would take place at a bombarding energy of 48 Mev is quite improbable. As mentioned before, one would estimate the $(\alpha,5n)$ threshold to be about 50 Mev. In the event that such a reaction did take place at 48 Mev, it would certainly not account for the appreciable amount of activity that was observed on Target 1. The nuclide was also seen on Target 2 at an energy of 40.6 Mev. An $(\alpha,5n)$ reaction at this energy would seem to be even more improbable. The alpha emitter was not observed at 31.2 Mev.

The results indicate that the 19-hr isotope was produced by a characteristic $(\alpha,4n)$ reaction and, hence must be Tb^{151}. Terbium-151 was also mass assigned independently by Mihelich and coworkers at Oak Ridge,[5] who studied the electron capture decay of the isotope.

Alpha activity due to Tb^{152} was looked for in Targets 2 and 3. Other than the 19-hr Tb^{151} no alpha activity could be detected. According to a recent communication, the half-life of Tb^{152} is 1 hr.[5] Since the samples were counted directly after bombardment, the alpha activity of Tb^{152} should have been seen if its alpha branching ratio were of a reasonable magnitude. The conclusion is that the alpha branching must be extremely small.

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LEGENDS FOR FIGURES

Fig. 1. The section of the chart of isotopes with which the study reported here has dealt.

Fig. 2. A pulse-analysis curve which shows two peaks, representing Dy^{152} and Dy^{153}. One of the standards used in the calibration, Gd^{148}, is also shown.

Fig. 3. A pulse-analysis curve which shows two peaks, Dy^{153} and Dy^{154}. The standard, Gd^{148}, is also shown.

Fig. 4. Growth and decay curve obtained in a nitrogen-ion bombardment on praseodymium. The growth period exhibits an 8-min half-life. The line representing the 8-min Dy^{149} is drawn on an expanded time scale.

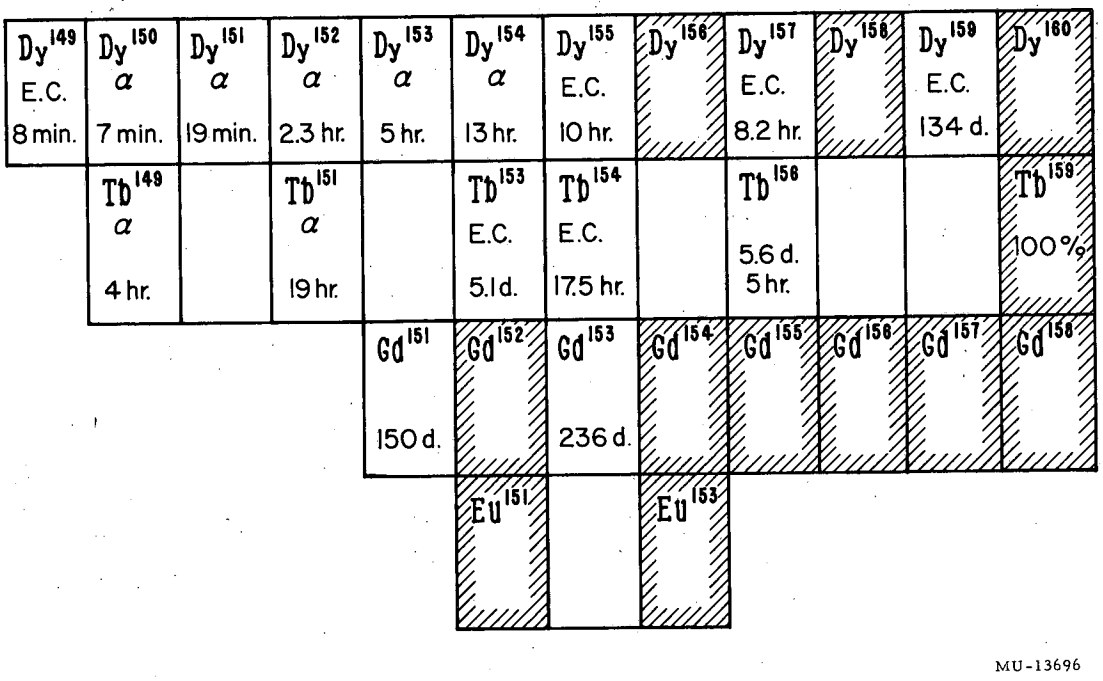
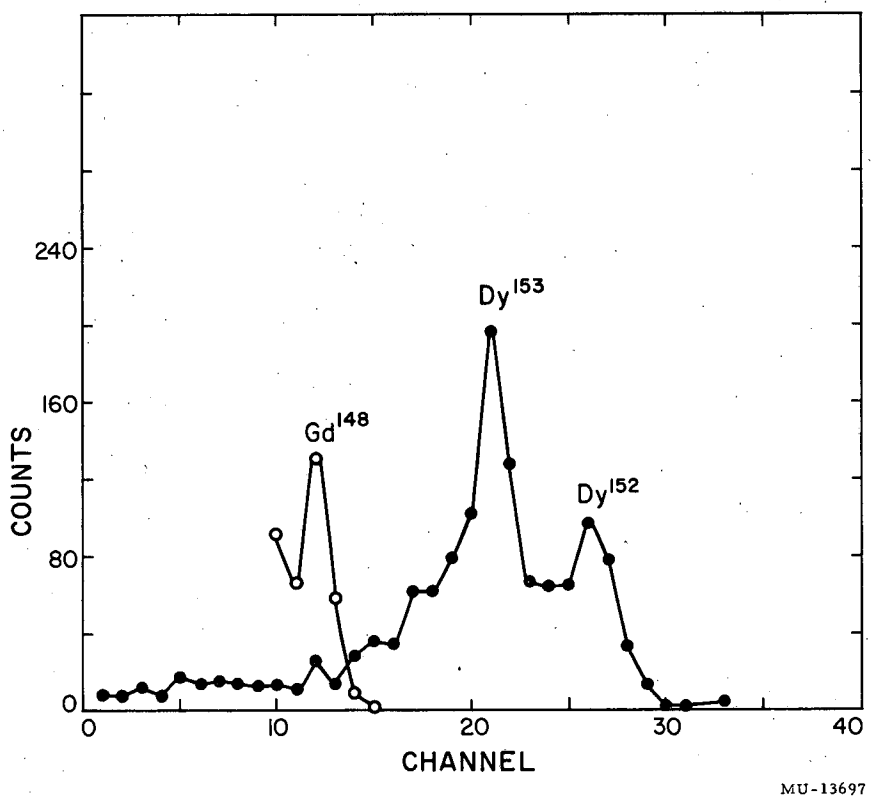

Dy149 E.C. 8 min.
Dy150 α 7 min.
Dy151 α 19 min.
Dy152 α 2.3 hr.
Dy153 α 5 hr.
Dy154 α 13 hr.
Dy155 E.C. 10 hr.
Dy156
Dy157 E.C. 8.2 hr.
Dy158
Dy159 E.C. 134 d.
Dy160
Tb149 α 4 hr.
Tb151 α 19 hr.
Tb153 E.C. 5.1 d.
Tb154 E.C. 17.5 hr.
Tb158 5.6 d. 5 hr.
Tb159 100%
Gd151 150 d.
Gd152
Gd153 236 d.
Gd154
Gd155
Gd156
Gd157
Gd158
Eu151
Eu153
MU-13696

Fig. 1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0
COUNTS
Gd148
Dy153
Dy152
0
10
20
30
40
CHANNEL
MU-13697

Fig. 2

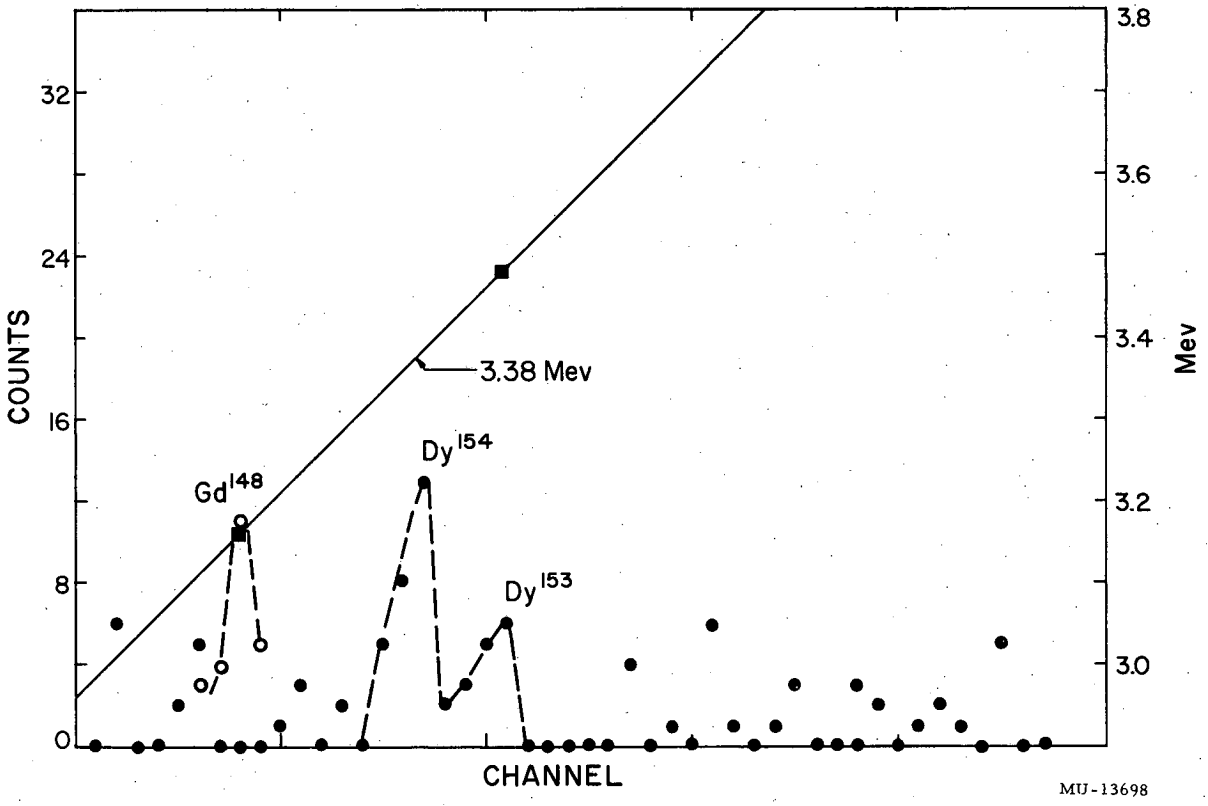
COUNTS
32
24
16
8
0
3.8
3.6
3.4
3.2
3.0
Mev
3.38 Mev
Gd148
Dy154
Dy153
CHANNEL
MU-13698

Fig. 3

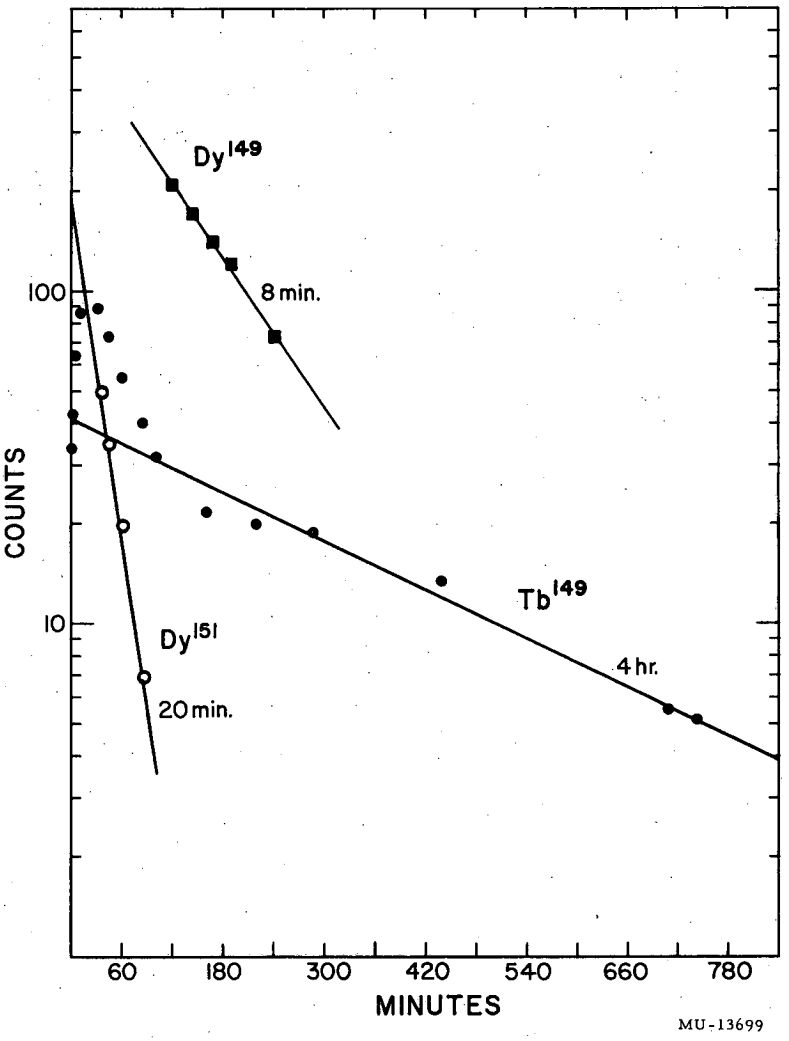
Dy149
8 min.
100
COUNTS
10
Dy151
20 min.
Tb149
4 hr.
60
180
300
420
540
660
780
MINUTES
MU-13699

Fig. 4