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Abnormal Charge Increase in Nuclear Reaction

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April 24, 1950

Berkeley, California

ABNORMAL CHARGE INCREASE IN NUCLEAR REACTION

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This thesis has two parts, the main part is abnormal charge increase in nuclear reaction, but in this phenomenon there were light iodine isotopes involved, lighter than I^{124}, which were not known, and we had to make them separately.[1]

NEUTRON DEFICIENT ISOTOPES OF IODINE

Four new isotopes of iodine have been identified with mass assignments and decay characteristics as indicated in Table I. The method of making mass assignments for some of these was to compare shapes of excitation curves

Table I

Mass No.	Half-life	Radiation	Energy
124	4.5 days	K, β^+	$\beta^+ = 2.1 \pm 0.1$ Mev
123	13 hr.	K, γ, e^-	$e^- = 150 \pm 15$ Kev
122	4 min.	$\beta^+ (K, \gamma ?)$	$\beta^+ = 2.9 \pm 0.1$ Mev
121	1.8 hr.	K, β^+, γ, e^-	$e^- = 185 \pm 10$ Kev
			$\beta^+ = 1.2 \pm 0.1$ Mev
120?	~ 30 min.	$\beta^+, \gamma (K?)$	$\beta^+ = 4.0 \pm 0.2$ Mev

of (α, xn) reactions on antimony in which known iodine activities were produced from the Sb^{123} and the new ones to be assigned from the Sb^{121}. The excitation functions as shown in Figure 1 are fairly crude as the only objective was to compare shapes in order to assign mass numbers. The experimental procedure was to irradiate antimony (57% Sb^{121}, 43% Sb^{123}) with helium ions of different energies, to isolate the iodine by a method in which the chemical yield could be measured, and then to resolve the iodine decay curves into the several components making use of distinctive radioactive properties.

13-hr. I^{123}.--A 13-hr. component was assigned to I^{123} since its excitation function paralleled that for 56-day I^{125}, both curves being of the shape that would be expected for $(\alpha,2n)$ reactions; in this case, on Sb^{121} and Sb^{123} respectively. The yields as shown include corrections for natural abundances of Sb^{121} and Sb^{123}, and are based on the assumption that both the 13-hr. I^{123} and 56-day I^{125} have one K x-ray per disintegration, and assuming one percent counting efficiency in the argon-filled Geiger tubes. The 13-hr. I^{123} has conversion electrons of 150 $\pm$ 15 Kev energy as determined with a low-resolution beta-ray spectrometer and by absorption in beryllium. Gamma-rays corresponding to this energy were also observed. This activity with similar half-life and radiation characteristics has been reported recently by Mitchell, Mei, Maienschein, and Peacock.[2]

4.5-day I^{124} and 13-day I^{126}.--The curves for both of these isotopes using normal antimony as a target are shown as appropriately labeled solid curves in Fig. 1. The curve for I^{126} is characteristic of an (α,n) reaction as is the low-energy part of that for I^{124}. The upswing beyond 30 Mev in the I^{124} curve is due to the $(\alpha,3n)$ reaction on Sb^{123}. The points shown for the broken line projection of the I^{124} curve were obtained from the irradiation of separated* Sb^{121} from which I^{124} can be formed only by the (α,n) reaction.

The discrepancy in yields at 20 Mev between I^{126} and I^{124} is not explained; but it may be remarked thatin other cases such as the fission of

*The sample of separated antimony consisting of 99.3% Sb^{121}, 0.7% Sb^{122} was obtained from the Isotopes Division of the U. S. Atomic Energy Commission.

bismuth[3] and the (γ,n) reaction on iodine,[4] the measured yield of I^{126} has been much lower than expected, making it appear that not all of the decay events take place through the 1-Mev beta-particle upon which the yields are based.

The half-life of I^{124} was measured as 4.5 days and the β^+-energy as 2.1 ± 0.1 Mev. It is estimated from the yield of K x-rays that this isotope decays only about 30% by positron-emission and 70% by electron-capture.

4-min. I^{122}.--In experiments in which an hour was consumed in chemical separation, no activity appeared which followed the excitation function of that part of the 4.5-day I^{124} curve produced by the (α,3n) reaction. It was therefore assumed that I^{122}, which would be formed by the $Sb^{121}(\alpha,3n)\ I^{122}$ reaction, is short-lived. More rapid chemistry showed a 4-min. iodine activity, and a single yield determination at 45 Mev showed it to be in the expected range as indicated in Figure 1. The positron has an energy of 2.9 ± 0.1 Mev, and it is estimated that there is some electron-capture branching.

1.8-hr. I^{121}.--An activity with 1.8-hr. half-life with a 1.2-Mev positron and conversion electrons of 185 Kev appeared in irradiation of antimony with 60-, 100-, and 360-Mev helium ions. Its decay is followed by the appearance of 17-day Te^{121} in approximately the proper yield for a parent-daughter relationship.

30-min. I^{120}(?).--In the higher energy irradiations (100 and 360 Mev) a 30-min. activity appeared, having a hard positron of 4.0 Mev. The decay of the iodine containing this activity was followed by the appearance of a tellurium activity which could not be resolved uniquely but which does have a half-life of several days. It is possible that this activity is 4.5-day Te^{119}. This would indicate that I^{119} was present. However, the observed positron energy of the 30-min. period is in better agreement with the prediction for I^{120} which may mean that I^{119} was also present but unobserved.

ABNORMAL CHARGE INCREASE IN NUCLEAR REACTION

We have made several experiments in which we bombarded ordinary Sn with high energy protons and alpha particles (~ 350 Mev) and observe the formation of several iodine activities. The activities observed were I^{120}, I^{121}, I^{123}, I^{124} and I^{126}. If this effect were real, it could not be explained by a one-step reaction, as could be seen by observing the following fragment of the chart of isotopes in this region.

	112	113	114	115	116	117	118	119	120	121
I									β^+, (K) 30 m	β^+, K 1.8 hr
Te									S 0.091%	K 17 d
Sb										S 57.25%
Sn	S 0.90%		S 0.61%	S 0.35%	S 14.07%	S 7.54%	S 23.98%	S 8.62%	S 33.03%	

	122	123	124	125	126	127	128	129	130
I	β^+, (K) 4m	K 13 hr	K, β^+ 4.5 d	K 56 d	β^- 13 d	S 100%	β^- 25 m	β^- long	β^- 12.6 hr
Te	S 2.49%	S 0.89%	S 4.63%	S 7.01%	S 18.72%	β^- 9.3 hr	S 31.72%		S 34.46%
Sb		S 42.75%							
Sn	S 4.78%		S 6.11%						

The heaviest stable Sn isotope is Sn^{124}, so the reactions you would expect with protons and alpha particles are:

$$Sn^{124}(p,xn)Sb^{124.123.122--}$$

$$Sn^{124}(\alpha,xn)Te^{127,126,125--}$$

By the possible decay of these isotopes they could not go to the iodine activities observed, since they are stable or K-capture, or β^+-emitter isotopes, with the exception of Sb^{124} that goes to stable Te^{124} and Te^{127} that goes to stable I^{127}.

We have investigated the phenomenon more thoroughly to try to find out how it happens.

For the bombardment we used Sn targets of about 1 gm/cm^2 thickness and 1 1/2" x 1/4" section. They were bombarded in the circulating beam of the 184" cyclotron.

Since the yields are extremely small, of the order of 10^{-5} barn, we did the following chemistry to make sure that the activity observed was iodine. The target and 10 mg of I^- carrier were put in a distillation flask with about 30 cc of H_2SO_4, the target dissolved by heating, and the iodine, probably as HI, was collected in SO_2 or NaOH solution. The solution was acidified if necessary, and the I^- oxidized with NO_2^-, put again in a distillation flask and distilled. The iodine was collected in SO_2 or NaOH solution, acidified with H_2SO_4, oxidized with NO_2^- and extracted into CCl_4. From the CCl_4 it was extracted back again into SO_2 or NaOH solution. The CCl_4 cycle repeated three times and finally the iodine was precipitated as AgI.

The first thing we did was to make sure that the activity was iodine. In order to do that, we observed the decay curves of several bombardments, and we were able to resolve the curves into half lives in close agreement with known iodine half lives, and also in the long bombardment with protons we were able to follow the e^-, β^- and β^+ activities in the β spectrograph.

Figure 2 shows the e^- from I^{123}, the β^- spectrum from I^{126}, and the β^+ spectrum from I^{124}, all of them made from alpha bombardments on Sb. Figure 3 shows the combined spectrum of I^{123} and I^{126} in a bombardment of Sn with 350 Mev protons, the shape of the spectra are very much alike the previous one, and the decay of the I^{123} is roughly what you would expect.

Figure 4 shows the β^+ spectrum of I^{124}, and its decay through two days, its shape is like the very well established I^{124} in Figure 1.

In Figure 2, 3, and 4 the abscissa is $H\rho$ in kilogauss - cm, and the ordinate is activity in an arbitrary scale.

Figure 5 shows the gross activity of the iodine in a bombardment of Sn with 350 Mev protons and its resolution in half-lives.

Figure 6, 7, and 8 show the gross activity, the x and γ-rays, and γ-rays activities respectively, from a long bombardment of Sn with 350 Mev protons. The resolution in half-lives is indicated too.

Figure 9 shows the iodine activities observed in a bombardment of Sn with 360 Mev alpha particles; the curve shows the gross counting rate, and its resolution in half-lives is indicated.

The half-lives of the resolved curves, the energy and decay of the e^-, β^- and β^+ activities in the β spectrograph, and the chemistry done, place almost unmistakable the activity as iodine, and we felt confident to continue work on this problem.

We wanted to make sure that the activity was not coming from some impurity. The spectroscopic analysis of the Sn sample is as follows:

Pb	.0005%	Pb + Cu + Fe	.0025%
Cu	.0005%	As + Sb	.0001%
Fe	.00015%	Bi + Ag + In	.0001%
As + Sb + Bi + Ag	.0001%	In	None found

Since the cross section is of the order of 10^{-5} barn for each of the iodine activities observed, it is almost impossible that the activity comes from an impurity.

Sb impurity would not give iodine activities with protons, since $Sb^{121,123}(p,xn)Te^{123,122--}$; you would get iodine activities with alpha particles $Sb^{121,123}(\alpha,xn)I^{126,125--}$, but the fact that you get the iodine activities with both protons and alpha particles, rules out Sb as impurities.

Te impurity would give with both protons and alpha particles iodine activities, but the most abundant isotopes of Te are Te^{126}, Te^{128} and Te^{130}, you would expect to get in this case heavier iodine isotopes too, such as I^{130} and I^{131} which have not been observed. It could not explain either the upward trend of the excitation functions that will be shown later.

Iodine impurity could show roughly the behavior observed, but we made a determination of iodine in the Sn sample, and we were not able to detect it to the limit of our experiment (one part in 100,000). The cross-section for Sb activities on Sb irradiated with 360 Mev alphas is about 4×10^{-1} barn.[5] Assuming the same cross-section for the formation of iodine activities with alpha particles on iodine, it would require 1 part in 10,000 of iodine impurity to account for the activity observed, and this is 10 times more than the upper limit we set.

Besides this we can make the following arguments; if there were U impurities, most of the activity would be I^{131}.[6] If there were Bi, the fission cross-section of Bi with 200 Mev yields about 4×10^{-3} barn of I;[7] assuming that the cross section for 350 Mev protons and alphas is of the same order, it would require about 1% of Bi impurity, which seems impossible. If it were coming from the spallation of some heavy element, such as Cs or heavier, you would expect to get almost as much Ba activity, and probably much more as iodine. We made a search for Ba activity and we coulnd not reduce it to zero by doing chemistry of Ba, but we could find an upper limit for the Ba activity making safe assumptions for the nature of the activity and mode of decay of possible Ba, and we found its yield to be at most 10^{-6} barn, probably 10^{-8} barn, which is much too small to allow any impurity such as Cs or heavier to account for the I activity. We also looked for alpha emitters in the bombarded target, and we did not see any which would again rule out the fission of U or Th as a source of iodine.

We ran bombardments at lower energies than 350 Mev with both protons and alpha particles, and the result of the excitation functions are shown in Figure 10 and 11.

The points below 100 Mev were uncertain, but we made bombardments under exactly the same condition at 25 Mev and 50 Mev protons. We observed that at 25 Mev there was no iodine activity whereas at 50 Mev it was barely detectable, but there seemed to be some. That puts the threshold for the reaction at around 50 Mev. We made a similar experiment with alpha particle at 30 Mev and 60 Mev, and the results were the same. There was no I activity at 30 Mev, and a little bit at 60 Mev, giving a threshold for alpha close to the proton threshold.

The best values for the cross sections were determined at full energy alpha and protons and are shown in the following table.

Isotope	350 Mev Protons	350 Mev Alphas
40m I^{120}	$.5 \times 10^{-5}$ barn	$.9 \times 10^{-5}$
1.8 hr I^{121}	$.1 \times 10^{-5}$	1.1×10^{-5}
13 hr I^{123}	$.7 \times 10^{-5}$	1.3×10^{-5}
4.5 days I^{124}	$.4 \times 10^{-5}$	$.4 \times 10^{-5}$
13 d I^{126}	$.6 \times 10^{-5}$	$.1 \times 10^{-5}$
Total	2.3×10^{-5}	3.8×10^{-5}

We estimate that these values are correct within a factor of two.

We believe that 4 min. I^{122} and 56 d I^{125} were formed, but they were not detectable, actually any iodine very short lived would not be seen, because the chemistry took at least 30 minutes, and any iodine very long lived would not give enough counts per minute to be observed.

DISCUSSION

The result of the experiment seemed to indicate that in the first approximation things happen very much the same, regardless of the nature of the bombarding particle, alpha or protons, and only depend on the energy, also the trend of the excitation function, the threshold of the reaction and the values for the cross section at full energy proton and alpha seem to indicate the same thing. The following mechanism is suggested.

$$_{50}Sn + \begin{matrix} p \\ \alpha \end{matrix} \longrightarrow \text{fragments} + {}_{3}Li$$

$$_{50}Sn + {}_{3}Li \longrightarrow {}_{53}I$$

This is not surprising since the hammer tracks of the Li^8 are very well known in reactions induced by cosmic radiation, also Li^8 has been observed by Wright, bombarding several elements up to Xe, using 350 Mev protons and 190 Mev deuterons.[8] In another experiment we have been able to make Be^7 with 350 Mev protons on Cu.

We have calculated the threshold for the reactions

$$_{50}Sn^{122,123,124} + p \longrightarrow {}_{3}Li^{6,7,8} + {}_{48}Cd^{117}$$

they are all endothermic, having thresholds of 8 Mev, 3 Mev and 16 Mev respectively. Actually the Li nucleus has to come out with at least 30 Mev, because this is the height of the barrier in Sn for Li, and since it has to penetrate another barrier of 30 Mev, you would expect that you would not get any iodine unless the energy of the bombarded particle is about 50 Mev.

The increase of activity with increasing energy is obvious, there is more energy available to knock out Li, and also more kinetic energy available to the Li nucleus, so you would observe an increase in the iodine activity.

We have tried to estimate the cross section of the formation of Li in the primary reaction, (this includes several possible Li, such as $_3Li^6$, $_3Li^7$, $_3Li^8$, etc. and also probably excited states.)

In order to do that we have made the assumption that the cross section for an Li nucleus to penetrate a $_{50}Sn$ nucleus is

$$\sigma = \pi R^2(1 - B/E) \text{ for } E > B$$
$$\sigma = 0 \text{ for } E < B$$

E is the energy of the Li nucleus, and B = 30 Mev is the height of the potential barrier.

We also assumed that practically all Li entering Sn nucleus yields finally I, the total cross sections for iodine would be taken as 4×10^{-5} barn.

From this assumption and range energy relations for Li we can calculate the cross section for Li if its energy distribution were known. The only thing definite about it is that there will not be Li with less than 30 Mev (the height of the barrier), nor with more than 350 Mev (the maximum energy available).

If the Li emitted are monoenergetic with energy ϵ, it can be shown that the cross sections are related by the formula

$$\sigma_{Li} = \frac{\sigma_I}{\pi R^2 \int_{E=B}^{E=\epsilon} n(1 - B/E)\, dx}$$

$\sigma_I \cong 4 \times 10^{-29}$ barn

$R = 1.48\ A^{1/3} \times 10^{-13}$ cm $= 7.37 \times 10^{-13}$ cm

$\pi R^2 = 1.67 \times 10^{-24}\ \text{cm}^2$

n = number of nuclei/cm^3 in Sn

dx = distance in cm

Putting this value we get:

$$\sigma_{21} = \frac{.24 \times 10^{20}}{\int_{E=B}^{E=\epsilon} n(1 - B/E)\,dx} \text{ barn}$$

The relation between E and x is obtained from range energy curves. We performed the integration numerically and will show some typical results in the following table in which σ_{Li} indicates the cross-section for the production of Li ions of indicated energy necessary to give the observed overall yield of iodine

ϵ (Mev)	36	40	50	80	120	200
$10^{-20} \int_{E=B}^{E=\epsilon} n(1-B/E)\,dx$	.0745	.2051	.8039	4.2954	13.146	45.18
σ_{Li} (barn)	3.2	1.2	3.0×10^{-1}	5.6×10^{-2}	1.8×10^{-2}	5.3×10^{-3}

Notice that as the energy of the Li increases they become much more effective to make I since its range and cross section increase.

It is known that at high bombarding energies the liquid drop model does not hold,[9] and probably there are localized collision putting a lot of energy in a small spot, and it could happen that a few Li are knocked out with high energy, and give the iodine activity observed. However, we have tried to see if we still could fit the result with the liquid drop model. Bethe has shown that the energy distribution is roughly Maxwellean, and if we take into account the barrier effect, we can write for the energy distribution of the Li

$$W(\epsilon) \approx (\epsilon - B)e^{-\frac{\epsilon - B}{T}}$$

where T is the nuclear temperature.

The cross section for a Li nucleus with energy ϵ then would be

$$\frac{d\sigma_{Li}}{dE} = A(\epsilon - B)e^{-\frac{\epsilon - B}{T}}$$

where A is a constant. That is of course for $\epsilon > B$, and for $\epsilon < B$

$$\frac{d\sigma_{Li}}{d\epsilon} = 0$$

It can be shown that if the energy distribution for the Li is $W(\epsilon)$ then;

$$\frac{\sigma_{Li}}{\sigma_I} = \frac{\int_{\epsilon = B}^{\epsilon = \epsilon_m} W(\epsilon)\, d\epsilon}{\pi R^2 \int_{\epsilon = B}^{\epsilon = \epsilon_m} (W(\epsilon) \int_{E = B}^{E = \epsilon} n(1 - B/E)\, dx)\, d\epsilon}$$

ϵ_m = maximum energy of Li

or calling I_1 and I_2 the integrals:

$$\frac{\sigma_{Li}}{\sigma_I} = \frac{I_1}{\pi R^2 I_2}$$

The integrations have been performed numerically, we used T = 10 Mev, as suggested by Professor Wick, and we get

$I_1' = 100.17$

$I_2 = 117.32 \times 10^{20}$

so $\sigma_{Li} = \dfrac{4 \times 10^{-5} \times 10^{2}}{1.67 \times 10^{-24} \times 1.17 \times 10^{22}} = .21$ barn

This value of T = 10 Mev is close to what you would get using the formula $E^* = 1/2\ \gamma_A T^2$ with $\gamma_A^{-1} = 0.1$ Mev which gives T = 8.5 Mev.[10]

We have tried to fit the data with the Weisskopf evaporation formula also, in this case the probability for emission of a particle with energy ϵ is:

$$W(E_A, \epsilon) = \frac{\sigma_o\, gm}{\pi^2 h^3}(\epsilon - V)\, e^{S_B (E_A - E_O - \epsilon) - S_A(E_A)}$$

Where $W(E_A, \epsilon)$ = probability per second of emission of a particle of energy between ϵ and $\epsilon + d\epsilon$.

A = excited nucleus

B = residual nucleus

E_A = excitation of A, above ground state

σ_o = geometrical cross-section of nucleus A

g = statistical weight of the particle

m = mass of the particle

E_o = binding energy of the particle to nucleus A

V = potential barrier of the particle with respect to B

and $S_A(X) = \left(\frac{AX}{2.2}\right)^{1/2}$ where X is in Mev.

We calculated the integrals numerically, and we got

$\sigma_{Li} = .81$ barn

which is even worse; also we can calculate from this formula the ratio of the probabilities of emission of L_1 nucleus to the emission of a nucleon, and it comes out to be

$$\frac{\Gamma_{L_1}}{\Gamma_n + \Gamma_p} = 1.6 \times 10^{-4}$$

From M. Lindner's experiment[5] we estimate that the cross section of emission of a nucleon is about 6 barn, this gives:

$$\sigma_{L_1} = 6 \times 1.6 \times 10^{-4} \cong 10^{-3} \text{ barn}$$

This result is different from the above one by a factor greater than 100, and we don't see any way to reconcile them within the liquid drop model or any other statistical model. Furthermore, we used 350 Mev as excitation energy for the nucleus, which on the average is less than that, and also not all the nucleons will be emitted with the nucleus having the maximum excitation energy.

However, it could be possible to explain this experiment if we assume that when the collision takes place, the heavy particles L_1 are ejected almost at the same time as the collision, that is, the L_1 are ejected before the excitation energy is evenly distributed among all nucleons and the compound nucleus formed; this mechanism would enable heavier particles to be ejected easier and also, they could take away much more energy than what they would get from a statistical model. This argument, although qualitative, will explain the observed experiment, for instance a cross-section of about 2×10^{-2} barn for L_1 and an energy of 120 Mev will explain satisfactorily the results observed in bombardments with 350 Mev.

1. For summary of data see: G. T. Seaborg and I. Perlman, Rev. Mod. Phys. 20 585 (1948).

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3. R. H. Goeckermann and I. Perlman, Phys. Rev. 76 628 (1949).

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5. M. Lindner: Thesis UCRL-143

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7. R. Goeckermann and I. Perlman, Phys. Rev. 76 5, 628 (1949)

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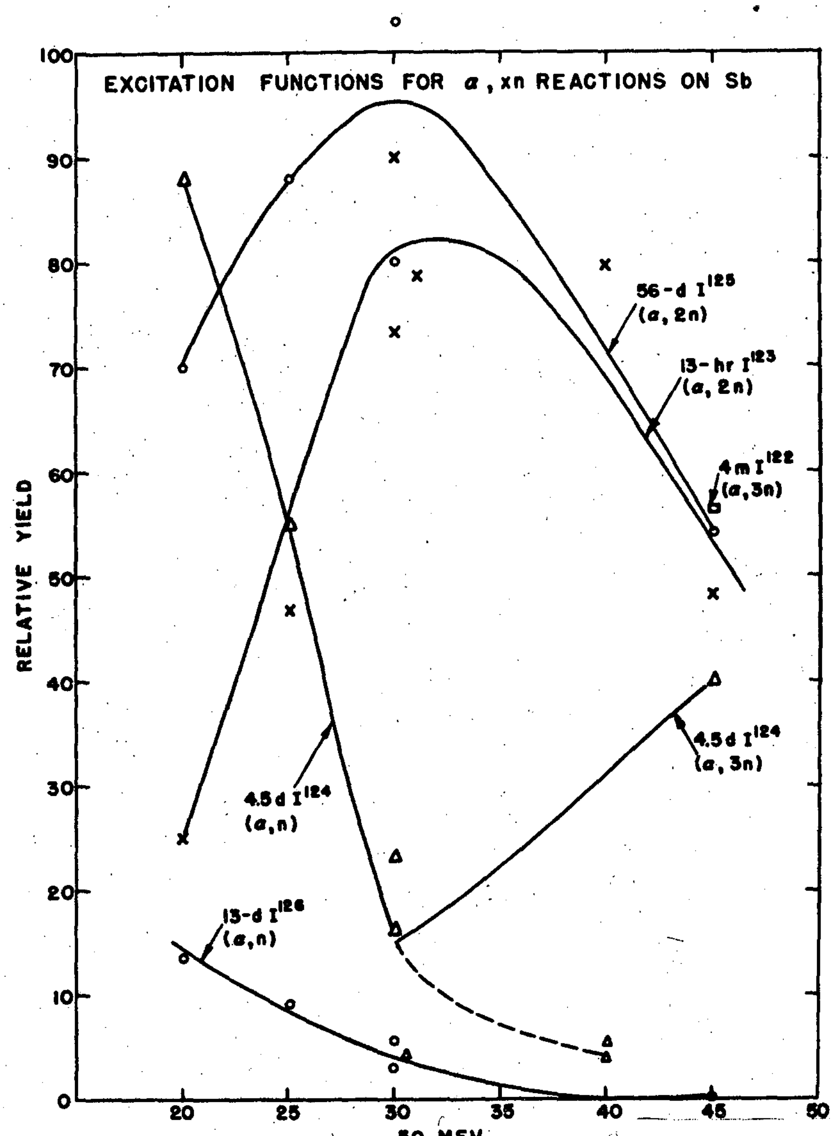


EXCITATION FUNCTIONS FOR α, xn REACTIONS ON Sb
RELATIVE YIELD
0
10
20
30
40
50
60
70
80
90
100
20
25
30
35
40
45
50
50 MEV
56-d I125 (α, 2n)
13-hr I123 (α, 2n)
4 m I122 (α, 3n)
4.5 d I124 (α, 3n)
4.5 d I124 (α, n)
13-d I126 (α, n)

FIG. 1

Mu 145

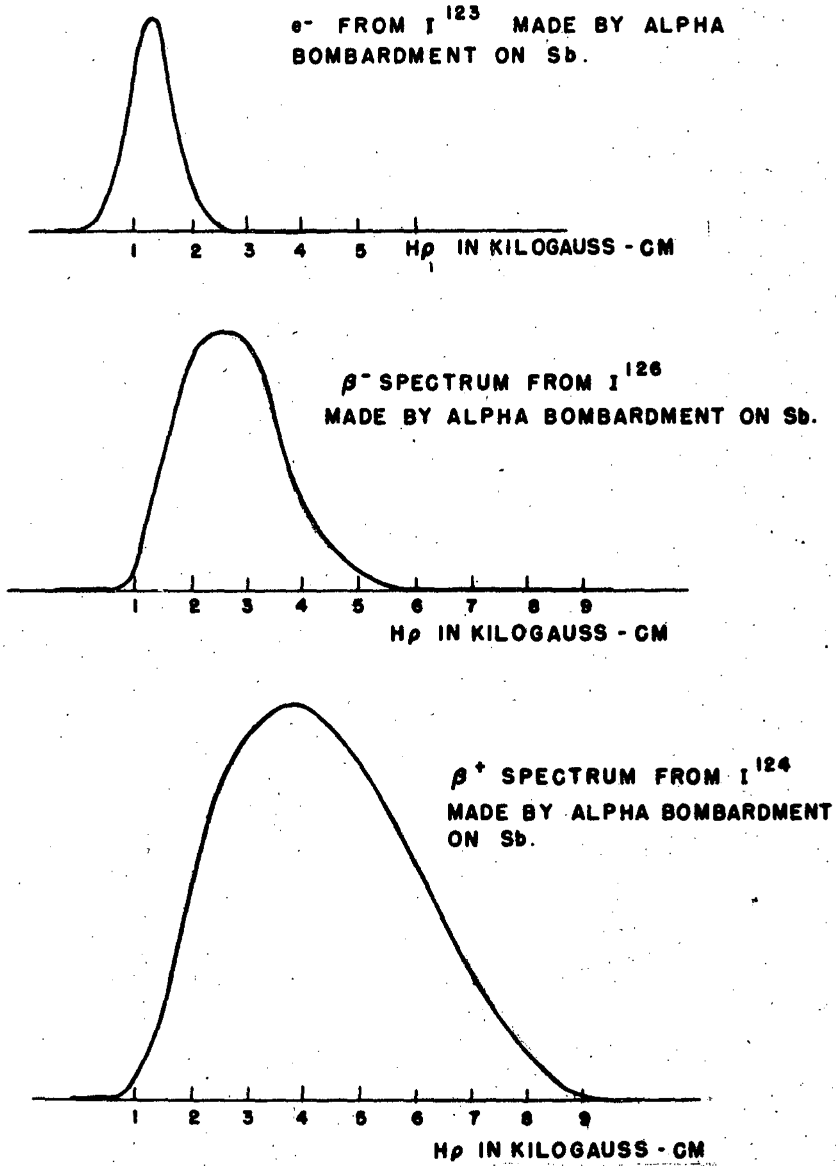


FIG. 2

Mu 146

14738-1

β^- IODINE ACTIVITY FROM 8 HR BOMBARDMENT OF Sn WITH 350 MEV PROTONS.

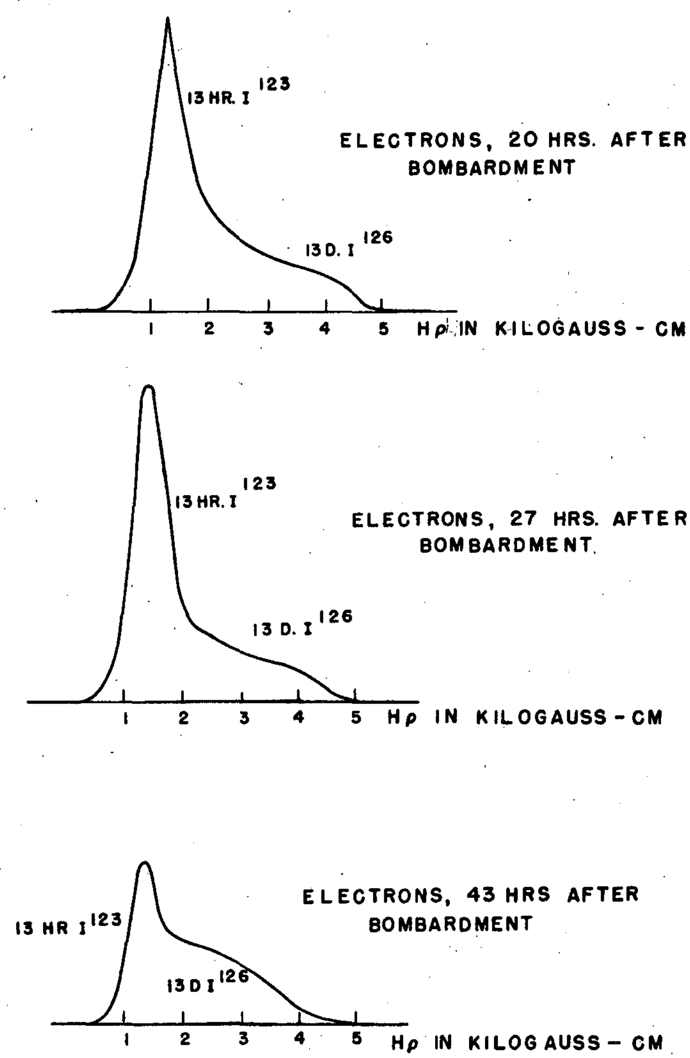


FIG. 3

Mu 147

β^+ IODINE ACTIVITY FROM 8 HR BOMBARDMENT OF Sn WITH 350 MEV PROTONS.

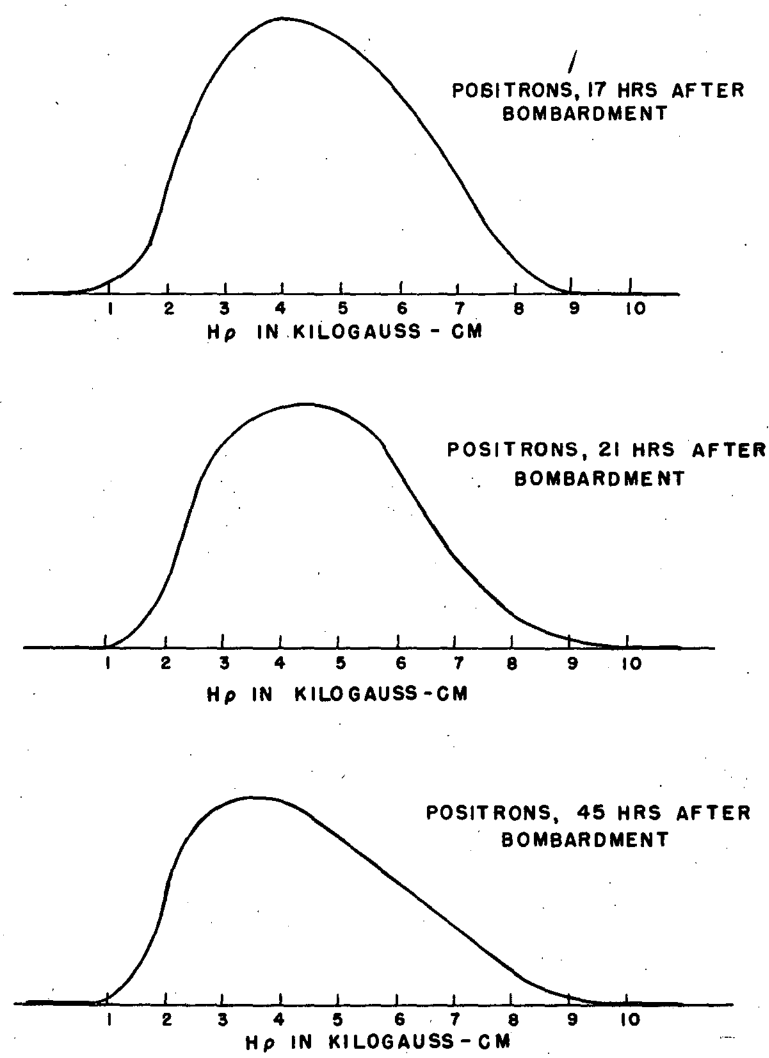


FIG. 4

Mu 148

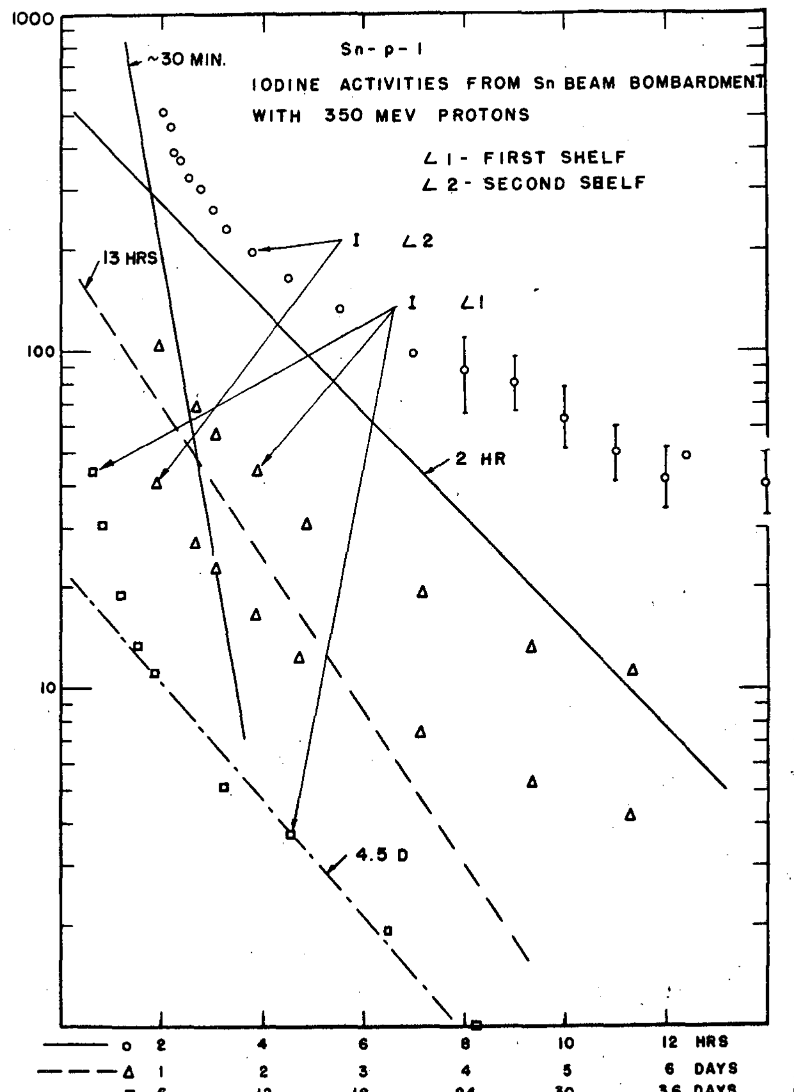


FIG. 5

MU 149

14743-1

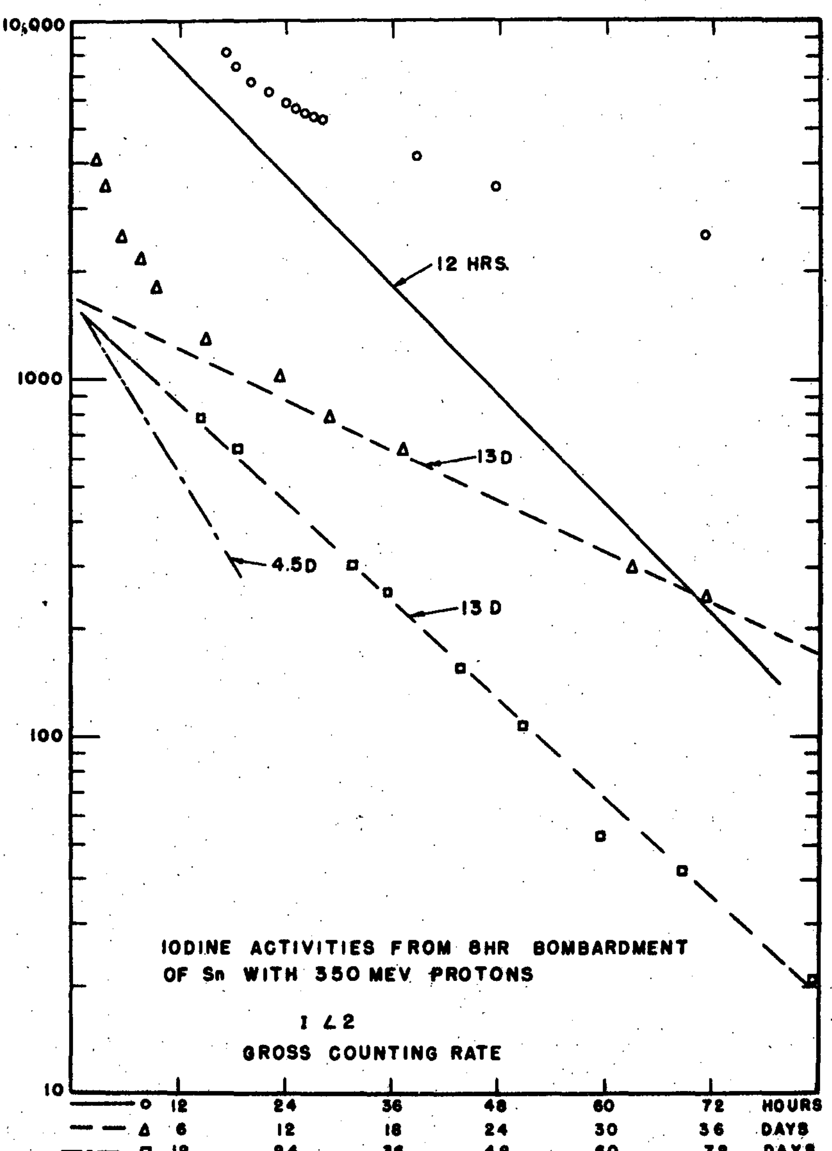


FIG. 6 Mu 150 14741-1

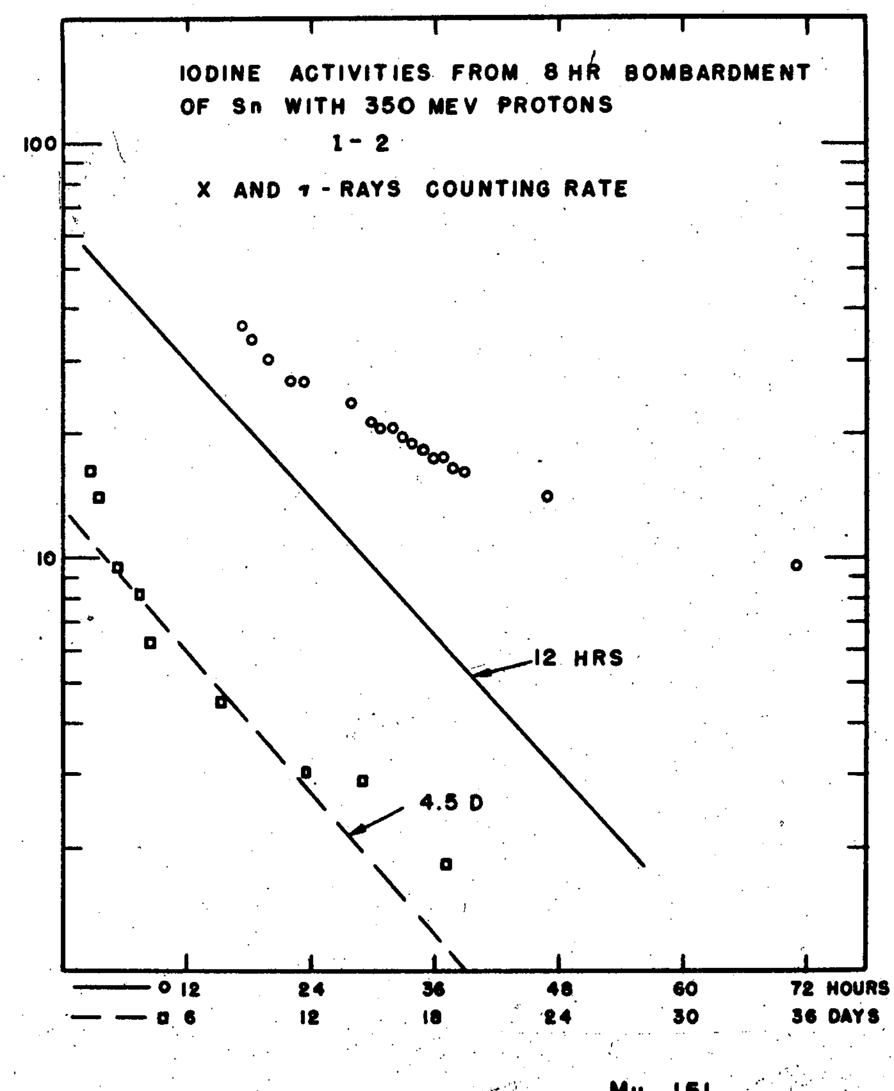
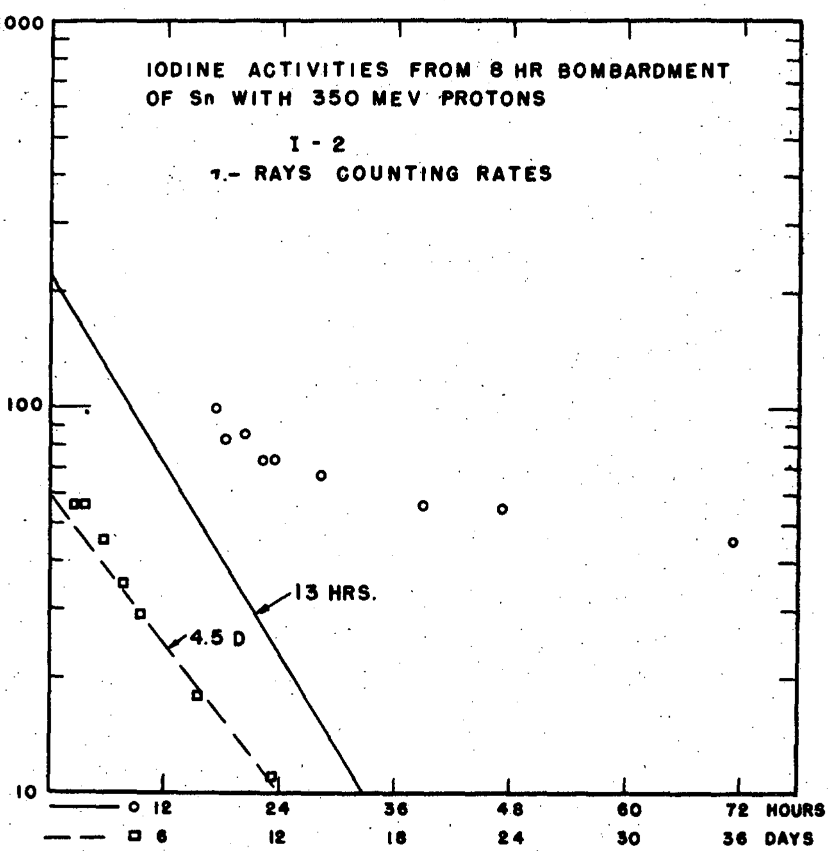

IODINE ACTIVITIES FROM 8 HR BOMBARDMENT
OF Sn WITH 350 MEV PROTONS
1-2
X AND γ-RAYS COUNTING RATE
100
10
12 HRS
4.5 D
12
24
36
48
60
72 HOURS
6
12
18
24
30
36 DAYS
Mu 151

FIG. 7



IODINE ACTIVITIES FROM 8 HR BOMBARDMENT
OF Sn WITH 350 MEV PROTONS
I - 2
γ - RAYS COUNTING RATES
1000
100
10
13 HRS.
4.5 D
12
24
36
48
60
72 HOURS
6
12
18
24
30
36 DAYS

Mu 152

FIG. 8

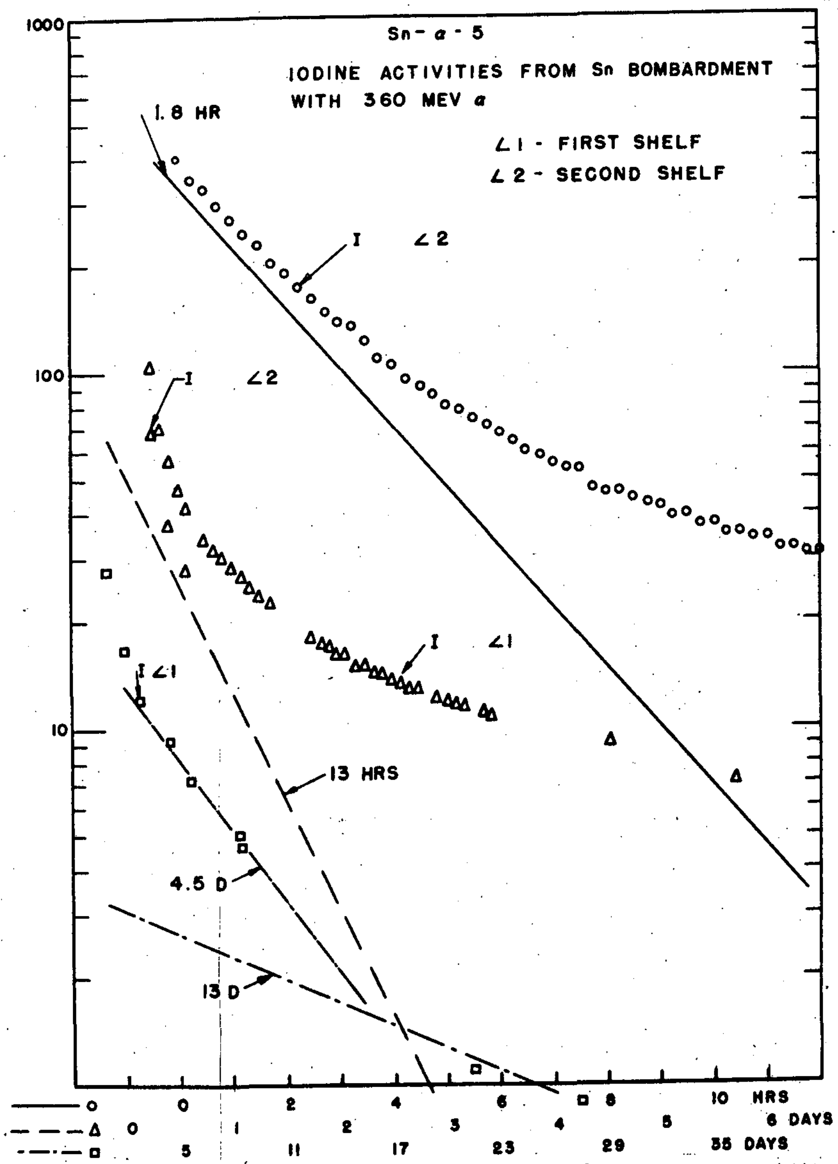
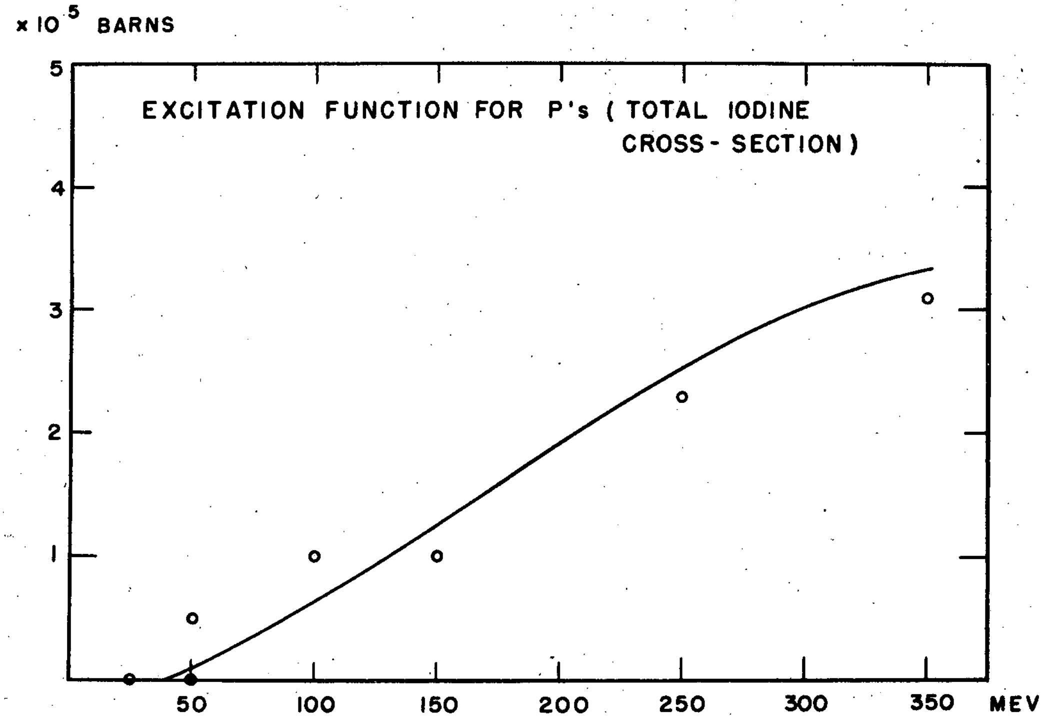


FIG. 9

Mu 153 14742-1



x 10^5 BARNS
5
4
3
2
1
EXCITATION FUNCTION FOR P's (TOTAL IODINE CROSS-SECTION)
50
100
150
200
250
300
350
MEV

Mu 155

FIG. 11