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Radioactive Isotopes of Iodine

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Five radioactive iodine isotopes are described, three of which are new. These were produced, by use of 8 Mev deuterons, 16 Mev helium ions and neutrons. Evidence is presented for the following isotopic assignments and properties:

REACTION	HALF-LIFE	PARTICLE RANGE GRAMS/CM² AL	GAMMA—HALF-THICKNESS GRAMS/CM² PB
$I^{127}(n, \gamma)I^{128}$ $Te^{128}(d, 2n)I^{128}$	25±1 min. (−)	1.05	3.5
$I^{127}(n, 2n)I^{126}$ $Sb^{123}(\alpha, n)I^{126}$ $Te^{125}(d, n)I^{126}$	13.0±0.3 days (−)	0.44	4.5
$Sb^{121}(\alpha, n)I^{124}$	4.0±0.3 days (+)	—	—
$Te^{128 \text{ or } 130}(d, n)$ $I^{129 \text{ or } 131}$	8.0±0.2 days (−)	0.50	3.3
$Te^{130}(d, 2n)I^{130}$	12.6±1.0 hr. (−)	0.40	6.0

It is shown that the 8-day activity is also produced by the radioactive decay of $Te^{129 \text{ or } 131}$. Chemical identification has been made in every case. A table of approximate yields is given.

INTRODUCTION

THE radioactive isotopes of iodine studied in this paper were produced by the bombardment of iodine, tellurium, or antimony by neutrons, deuterons, or helium ions. Fig. 1 shows the percent abundance of the stable antimony, tellurium, iodine and relevant xenon isotopes, as compiled by Livingston and Bethe.[1]

[1] Livingston and Bethe, Rev. Mod. Phys. 9, 380 (1937).

The bombardments were made with the Berkeley cyclotron. The deuteron currents were about 50 microamperes at 8 Mev, the helium ion currents 0.6 microampere at 16 Mev. In the final experiments, tellurium metal was melted onto a copper plate and bombarded in an atmosphere of helium at reduced pressure. The projectiles entered the chamber through a thin aluminum window. In the preliminary work, to

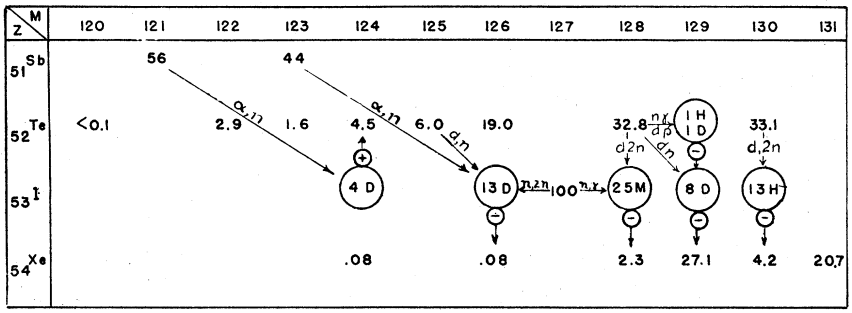

FIG. 1. Percent abundance of stable isotopes in the region of iodine. The half-lives of radioactive isotopes are encircled, and the transmutations are indicated by arrows.

avoid possible contamination of copper, sticks of metallic tellurium were supported outside the aluminum window and were bombarded in air. Antimony was activated in vacuum. Slabs of the metal were soldered to water-cooling pipes and the activated side was then filed off.

The signs of the disintegration products were determined by magnetic deflection in air. Activities were observed on a Lauritsen-type quartz-fiber electroscope. The background activity, about 0.001 division per second, was subtracted from all data.

All of the measurements were made upon chemically analyzed samples. In many cases, the activated tellurium was dissolved in nitric acid after the addition of a small quantity of sodium iodide to act as a carrier. The iodine which was liberated during the solution process was distilled into a reducing solution of sodium sulfite. The iodide was then precipitated as silver iodide upon the addition of silver nitrate solution. The same procedure was followed after alpha-particle bombardment of antimony and fast neutron activation of sodium iodide. On several occasions, a liter of ethyl iodide was exposed to neutrons and then shaken with an aqueous solution of sodium sulfite and sodium bicarbonate. The iodine was separated from the aqueous phase as silver iodide. For the study of the growth of radioactive iodine from tellurium, the iodine was separated by successive distillations during two experiments, while in a third more quantitative investigation, the procedure was followed of successive precipitations as silver iodide directly from the tellurium solution.

RADIOACTIVE ISOTOPE I^{128}: FROM $I+n$ AND $Te+D$; HALF-LIFE 25 ± 1 MINUTES $(-)$

This well-known activity, produced strongly by slow neutrons on iodine, was first observed by Fermi and his associates.[2] The work of Aston, and more recently, that of Nier,[3] shows only one stable iodine isotope, I^{127}, so that this activity has been assigned definitely to I^{128} through $I^{127}(n, \gamma)I^{128}$.

We have produced this same active isotope by bombarding tellurium with 8 Mev deuterons. It cannot have been formed by the usual (d, n) reaction, since Te^{127} is not stable. Consequently the transmutation must be either $Te^{126}(d, \gamma)I^{128}$ or $Te^{128}(d, 2n)I^{128}$. At present, the choice between these alternatives must be made from theoretical

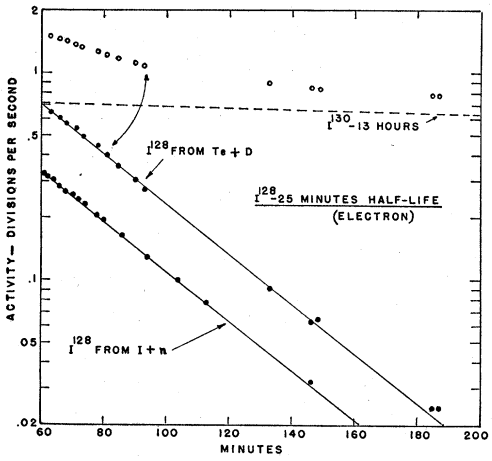

FIG. 2. Decay curves of I^{128}.

[2] Amaldi, d'Agostino, Fermi, Pontecorvo, Rasetti, Segrè, Proc. Roy. Soc. **A149**, 522 (1935).
[3] Nier, Phys. Rev. **52**, 933 (1937).

considerations, and the second alternative appears more likely.

The reactions leading to I^{128} are therefore,

$$_{53}I^{127}+{}_{0}n^{1}\rightarrow{}_{53}I^{128}+\gamma$$

and either

$$_{52}Te^{126}+{}_{1}D^{2}\rightarrow{}_{53}I^{128}+\gamma$$

or

$$_{52}Te^{128}+{}_{1}D^{2}\rightarrow{}_{53}I^{128}+2{}_{0}n^{1}.$$

The decay is

$$_{53}I^{128}\rightarrow{}_{54}Xe^{128}+{}_{-1}e^{0}\ (25\ \text{minutes}).$$

Figure 2 shows the 25-minute half-life of a neutron-activated sample of iodine and the initial portion of the iodine from a deuteron bombardment of tellurium (with correction for the longer-lived iodines also produced).

The absorption curves in aluminum and lead for the 25-minute I^{128} are given in Fig. 3, after correction for decay. The negative electrons have a maximum range of 1.05 ± 0.05 gram per cm² Al, corresponding to an energy of 2.2 Mev, according to the relation of Widdowson and Champion:[4] Energy (Mev) = [Range (gram/cm²) + 0.165]/0.536. The downward curvature of the plot suggests the presence of another component, possibly a monokinetic line from an internally converted gamma-ray. Cloud-chamber observations by others agree remarkably well with these measurements: Alichanian, Alichanow and Dzelepow[5] and Bacon, Grisewood and van der Merwe[6] find the maximum energy at 2.1 Mev, while the latter workers have further evidence for another group at 1.2 Mev.

The gamma-ray is absorbed to half-value by 3.5 grams per cm² Pb, which indicates an energy of 0.4 Mev, according to Gentner's correlation.[7] This is not in agreement with the difference of the maximum energies of the two electron groups quoted above.

Radioactive Isotope I^{126}: From I+Fast n, Te+D and Sb+He; Half-Life 13.0 ± 0.3 Days (−)

The existence of an activity with 13 days' half-life after irradiating iodine with fast neutrons from the (Li+D) reaction was discovered by Tape and Cork.[8] We have shown[9] the isotope to be chemically identifiable with iodine. The majority of the particles are electrons, but the method of sign determination does not rule out the existence of additional positron decay to stable Te^{126}. There can be no doubt that it is I^{126} formed by $I^{127}(n, 2n)I^{126}$.

We have found that the bombardment of antimony with 16 Mev helium ions gives rise to two radioactive iodine isotopes, with half-lives 4 days and 13 days. The appearance of the latter affords a good check on the assignment of the 13-day activity to I^{126}, since here it is formed by $Sb^{123}(\alpha, n)I^{126}$. (The interpretation given to the 4-day period will be discussed later.)

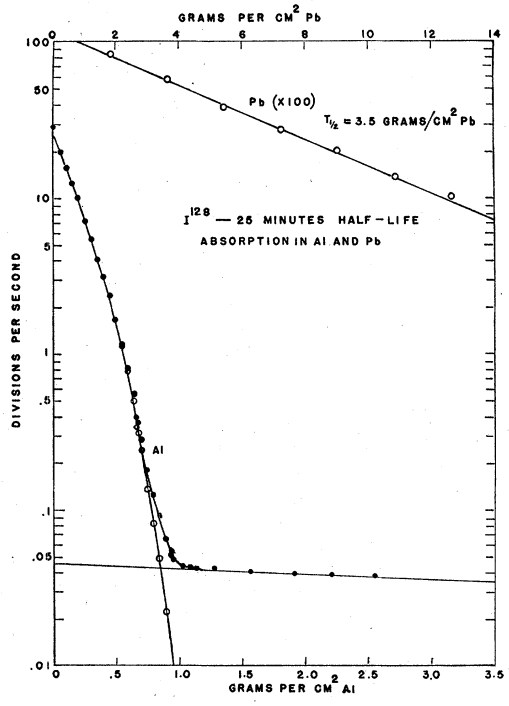

FIG. 3. Absorption curves of I^{128}.

Figure 4 shows decay curves of the 13-day activity obtained in these two reactions.

It is to be expected that the transmutations $Te^{125}(d, n)I^{126}$ or $Te^{126}(d, 2n)I^{126}$ should occur, but direct evidence for this is masked by another activity.

[4] Widdowson and Champion, Proc. Phys. Soc. **50**, 192 (1938).
[5] Alichanian, Alichanow and Dzelepow, Physik. Zeits. Sowjetunion **10**, 78 (1936).
[6] Bacon, Grisewood and van der Merwe, Phys. Rev. **54**, 315 (1938).
[7] Gentner, J. de phys. et rad. **6**, 274 (1935).
[8] Tape and Cork, Phys. Rev. **53**, 676 (1938).
[9] Livingood and Seaborg, Phys. Rev. **53**, 1015 (1938).

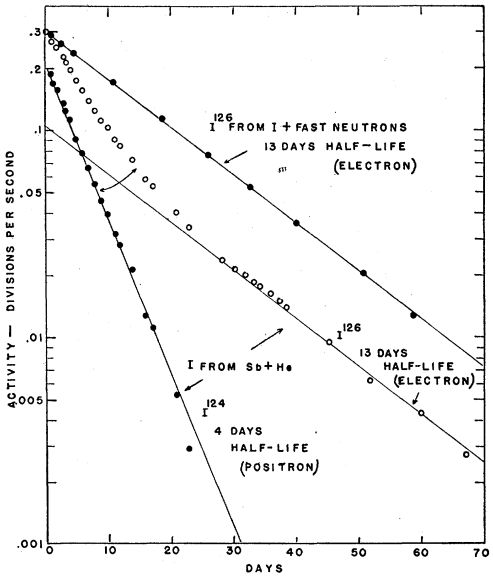

FIG. 4. Decay curves of I^{124} and I^{126}.

In Fig. 5 may be seen the decay of a first separation of iodine from a deuteron bombardment of tellurium; the observed half-lives are 25 minutes, 13 hours, and about 8.5 days. We will show later on that one can interpret the observed 8.5-day figure as the combination of an 8.0-day period found with independent evidence and a comparatively weak activity with the expected 13-day half-life. The intensity of this latter component, as indicated in Fig. 5, is what should be expected on the basis of the abundance of the tellurium isotopes, the half-lives and ionizing powers of the 8- and 13-day radiations.

We may therefore write as the transmutations leading to I^{126}:

$$_{53}I^{127}+{}_0n^1\rightarrow{}_{53}I^{126}+2{}_0n^1,$$

$$_{51}Sb^{123}+{}_2He^4\rightarrow{}_{53}I^{126}+{}_0n^1,$$

$$_{52}Te^{125}+{}_1D^2\rightarrow{}_{53}I^{126}+{}_0n^1,$$

followed by

$$_{53}I^{126}\rightarrow{}_{54}Xe^{126}+{}_{-1}e^0 \text{ (13 days)}.$$

Figure 6 shows absorption curves of the 13-day period, obtained from a sample of iodine activated with fast neutrons from a Li+D source. The negative electrons have a range of 0.44 ± 0.02 gram per cm^2 Al, corresponding[4] to an energy of 1.13 Mev. The gamma-ray is reduced to half-value by 4.5 grams per cm^2 Pb, which indicates[7] an energy of 0.5 Mev.

RADIOACTIVE ISOTOPE I^{124}: FROM Sb+He; HALF-LIFE 4.0 ± 0.3 DAYS (+)

As mentioned above, the bombardment of antimony with 16 Mev helium ions causes the formation of two active iodine isotopes, one emitting electrons with a half-life of 13 days and the other positrons with a 4-day half-life. There are but two stable antimony isotopes, Sb^{121} and Sb^{123}, which by the (α, n) reaction would lead to radioactive I^{124} and I^{126}. Since prolonged exposure of iodine to fast neutrons yields only the 13-day period [through $I^{127}(n, 2n)I^{126}$] but not the 4-day activity, it is certain that this latter must be due to I^{124}, the disintegration being:

$$_{51}Sb^{121}+{}_2He^4\rightarrow{}_{53}I^{124}+{}_0n^1,$$

followed by

$$_{53}I^{124}\rightarrow{}_{52}Te^{124}+{}_{+1}e^0 \text{ (4 days)}.$$

The decay of the 4-day activity is shown in Fig. 4.

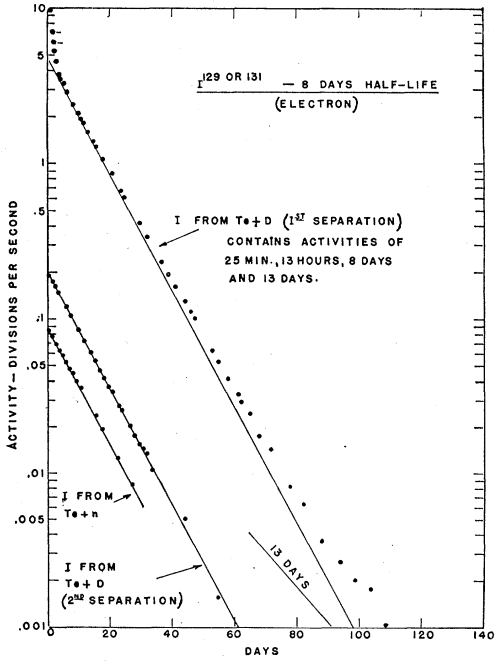

FIG. 5. Decay curves of I^{129} or I^{131}.

RADIOACTIVE ISOTOPE I^{129} OR I^{131}: FROM Te+D AND $Te^{129, 131} \rightarrow I^{129, 131}$; HALF-LIFE 8.0±0.2 DAYS (−)

We have briefly reported[9] the existence of an iodine isotope with 8 days' half-life, emitting negative electrons, that can be produced directly by bombarding tellurium with deuterons and which grows in activated tellurium. The evidence for this is now reported in detail. The 8-day period has been observed by Tape and Cork,[8] but was erroneously ascribed by them to a tellurium isotope.

In the iodine separated from a Te+D bombardment there is found a strong activity with 8 days' half-life, in addition to the other periods reported in this paper. If further extractions of iodine are made from the tellurium solution, the 8-day activity is again obtained in each, but not the other periods. This indicates a double decay from tellurium to iodine to stable xenon, and study of the isotopes shows that the chain must be either $Te^{129} \rightarrow I^{129} \rightarrow Xe^{129}$ or $Te^{131} \rightarrow I^{131} \rightarrow Xe^{131}$. The much greater intensity of the 8-day period in the first extraction suggests that a direct transmutation from tellurium to the active iodine also occurs, and it is seen that the expected reactions $Te^{128}(d, n)I^{129}$ or $Te^{130}(d, n)I^{131}$ lead to the same iodine isotope involved in the double decay.

To clinch the argument for a double decay, tellurium was irradiated with slow neutrons, which by no known reaction possibly could cause a direct transmutation to iodine. Nevertheless, the 8-day period was found in the chemically separated iodine fraction, thus proving conclusively that it grows from active tellurium.

Figure 5 gives examples of the decay curves obtained. A first extraction of iodine from deuteron-activated tellurium gives a multiplicity of periods, including that of 8 days' half-life, while a second extraction shows the 8-day period only. This also is the only activity found in iodine removed from neutron activated tellurium.

It has not been possible to determine the period of the parent tellurium by observations on the tellurium activity before and after iodine extractions, because of the overwhelming intensity of other tellurium activities. Recourse

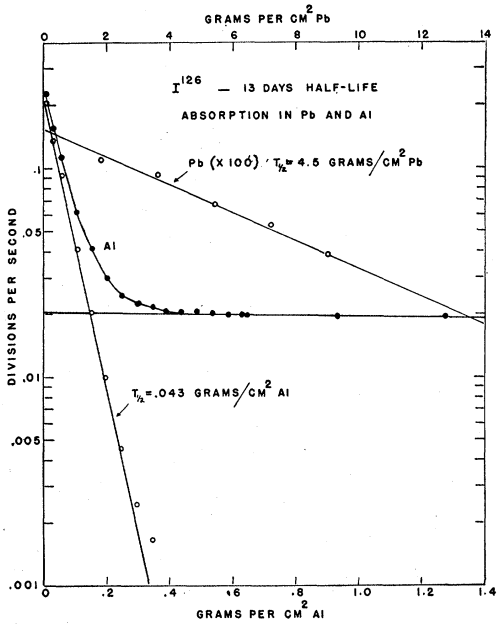

FIG. 6. Absorption curves of I^{126}.

therefore has been made to the procedure of observing the growth of the 8-day iodine and noting the time when this activity reaches its maximum value. This was done by making successive extractions of iodine from the same solution of deuteron-activated tellurium and by following each sample for a number of days in order to establish with precision the intensity of its 8-day period. The growth curve was then obtained by adding to the intensity of each sample (extrapolated back to the time of its separation) the intensities of all previously removed specimens, each also as of that moment. The resulting curve shows the total amount of 8-day iodine present as a function of time, zero being taken at a preliminary separation which removed all iodine directly produced by the $Te(d, n)I$ reaction.

Figure 7 shows such a curve, obtained from a run in which every precaution was taken to make the separations quantitative. It is seen that the 8-day activity reaches a maximum value in about 4 days' time.

The general equation describing the growth of a daughter substance from its parent is:

$$N_2 = N_1{}^0 \frac{\lambda_1}{\lambda_2 - \lambda_1}(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

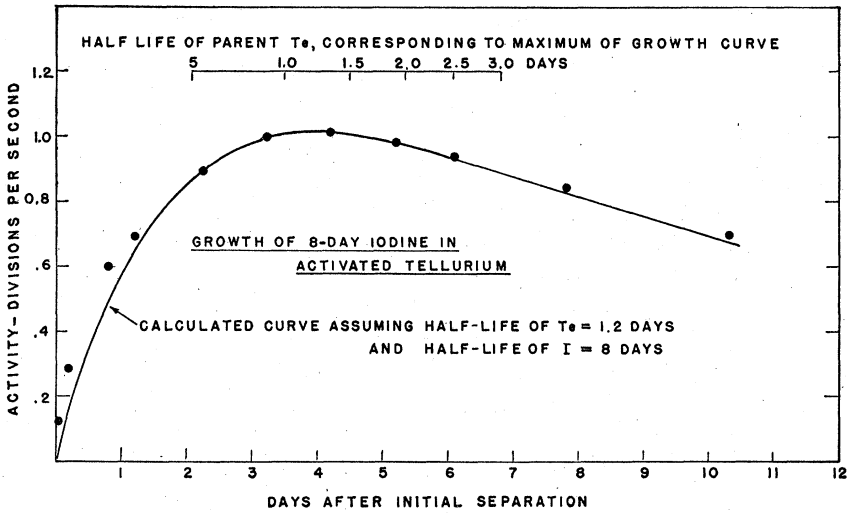

FIG. 7. The growth of 8-day iodine in tellurium. The ordinate of the calculated curve has been adjusted to fit at the maximum. See text for the possible significance of the abrupt initial rise of the experimental points.

where $\lambda = 0.693/T$ and T is the half-life. From this it follows by differentiation that the time required for the daughter to grow to a maximum amount is:

$$t_{(\max N_2)} = \frac{1}{\lambda_2 - \lambda_1} \ln \frac{\lambda_2}{\lambda_1} = 3.32 \frac{T_1 T_2}{T_1 - T_2} \log_{10} \frac{T_1}{T_2}.$$

With T_2 taken as 8 days and $t_{(\max N_2)}$ as 4 days, the half-life T_1 of the parent tellurium is found to be 1.2 days. This is admittedly rough, since the result is very sensitive to the chosen value for $t_{(\max N_2)}$.

The smooth curve in Fig. 7 was drawn from the general equation above, by using $T_1 = 1.2$ days and $T_2 = 8$ days, and adjusting the vertical scale to fit the experimental points at the maximum. It is seen that the observed rise at the beginning is somewhat faster than the calculated curve, which suggests that possibly the 8-day iodine is also growing from the well-known[2, 8, 10, 11] one-hour period of tellurium. This abrupt initial rise has been observed by us on other growth curves, and is more pronounced when the separations are made very soon after the bombardment ceases. This rapid growth at the beginning cannot be spurious because of incompletely separated iodine made directly through Te(d, n)I, since the other directly formed iodine activities (25 minutes and 13 hours) do not appear in the growth fractions.

If it is true that the 8-day iodine grows from the one-hour as well as the 1.2-day period, then these two activities must be isomers and may be assigned definitely to Te^{129}, since the one-hour activity has been produced by 17 Mev gamma-rays on tellurium, through $Te^{130}(\gamma, n)Te^{129}$. (Our unpublished work on radioactive tellurium has shown that the one-hour period must be assigned to Te^{129} rather than to Te^{127} or Te^{121}.)

Our decay curves of active tellurium, after neutron or deuteron bombardments, suggest confirmation of a tellurium activity with the predicted half-life of a little over a day, but the analysis is complicated and somewhat uncertain, because of the presence of the first growing and then decaying 8-day iodine period intermixed with many strong true tellurium activities.

To capitulate this discussion of the 8-day iodine: it is produced by direct transmutation of tellurium,

$$_{52}Te^{128 \text{ or } 130} + {}_1D^2 \rightarrow {}_{53}I^{129 \text{ or } 131} + {}_0n^1,$$

followed by

$$_{53}I^{129 \text{ or } 131} \rightarrow {}_{54}Xe^{129 \text{ or } 131} + {}_{-1}e^0 \text{ (8.0 days)}.$$

It can also be obtained as a double decay after

[10] Bothe and Gentner, Naturwiss. **25**, 191 (1937).
[11] Heyn, Nature **139**, 842 (1937).

either of two primary processes:

$$_{52}Te^{128 \text{ or } 130} + {}_1D^2 \rightarrow {}_{52}Te^{129 \text{ or } 131} + {}_1H^1$$

or $$_{52}Te^{128 \text{ or } 130} + {}_0n^1 \rightarrow {}_{52}Te^{129 \text{ or } 131} + \gamma,$$

followed by

$$_{52}Te^{129 \text{ or } 131} \rightarrow {}_{53}I^{129 \text{ or } 131} + {}_{-1}e^0$$

(~1.2 days and perhaps one hour)

and

$$_{53}I^{129 \text{ or } 131} \rightarrow {}_{54}Xe^{129 \text{ or } 131} + {}_{-1}e^0 \text{ (8.0 days).}$$

If it is true that the iodine grows from the one-hour tellurium as well as from the 1.2-day tellurium, then the chain of decay can be definitely assigned to $Te^{129} \rightarrow I^{129} \rightarrow Xe^{129}$. An unambiguous check on this would lie in a deuteron bombardment of xenon, since the (d, α) reaction could lead to I^{129} but not to I^{131}, because stable Xe^{131} exists while stable Xe^{133} is unknown. We have little hope of accomplishing this with the 8 Mev deuterons available, since it is our experience that the (d, α) reaction becomes rapidly less efficient as one progresses up the periodic table, and we are not definitely sure of its occurrence even as far up as tin, four elements below xenon.

The characteristics of the radiations of the 8-day iodine are shown in Fig. 8. The electrons have a range of 0.50 ± 0.04 gram per cm² Al, or an energy[4] of 1.24 Mev. The gamma-ray is absorbed to half-value by 3.3 grams per cm² Pb, which indicates[7] an energy of 0.4 Mev.

Radioactive Isotope I^{130}: From Te+D; Half-Life 12.6 ± 1.0 Hours (−)

The bombardment of tellurium with 8 Mev deuterons gives rise to a chemically identified iodine isotope with half-life about 13 hours, as we have briefly reported.[9] The emitted particles are negative electrons with a normal type of absorption curve. Since this period is not obtained by very intense bombardment with fast or slow neutrons, it cannot be due to I^{126} or I^{128}, and since it is not produced by activating antimony with alpha-particles (see above) it cannot be assigned to I^{124} or I^{126}. The remaining iodine isotopes that could be electron emitters are I^{129}, I^{130} and I^{131}. If it is due to either the first or third of these, the 13-hour activity should be found growing from radioactive Te^{129} or Te^{131} and hence also should be produced by irradiating tellurium with neutrons. (We have found strong activities corresponding to both Te^{129} and Te^{131} in the tellurium fractions of our deuteron and neutron bombardments of tellurium.) We have not been able to find any trace of this 13-hour period growing in deuteron-activated tellurium, nor do we find it after neutron bombardments of tellurium. We therefore feel confident that it must be assigned to I^{130}, formed in either of these two ways:

$$_{52}Te^{130} + {}_1D^2 \rightarrow {}_{53}I^{130} + 2\,{}_0n^1$$

or $$_{52}Te^{128} + {}_1D^2 \rightarrow {}_{53}I^{130} + \gamma,$$

followed by

$$_{53}I^{130} \rightarrow {}_{54}Xe^{130} + {}_{-1}e^0 \text{ (13 hours).}$$

It will be recalled that it also was necessary to postulate the existence of either the $(d, 2n)$ or (d, γ) reaction to explain the production of the 25-minute I^{128} by deuteron bombardment of tellurium (see above).

Figure 10 gives the absorption curves of this 13-hour activity. These data were obtained by taking the difference between corresponding

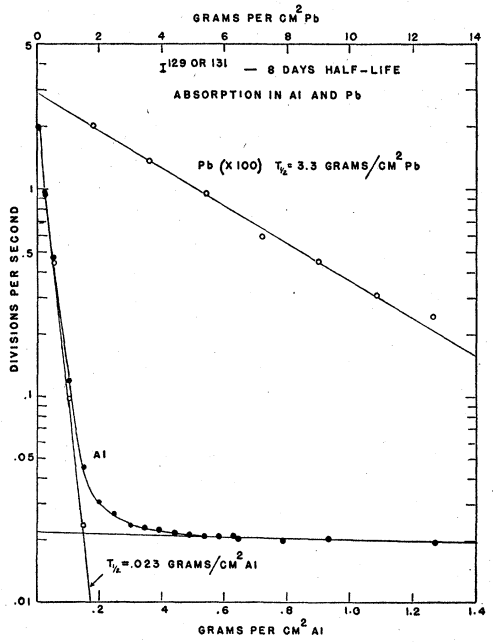

FIG. 8. Absorption curves of I^{129} or I^{131}.

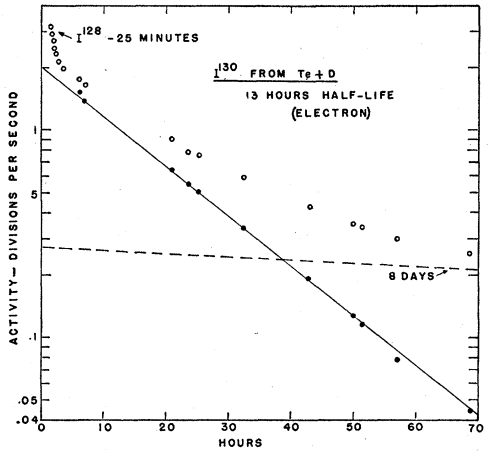

FIG. 9. Decay curve of I^{130}.

readings made (*a*) when the activity was composed of both the 13-hour and 8-day periods and (*b*) when only the 8-day period was existent; these latter figures were corrected for decay back to their appropriate values at the earlier date. The electrons show a maximum range of 0.40 $\pm$0.02 gram per cm^2 Al, or an energy[4] of 1.05 Mev, while the gamma-ray, showing a half-thickness of 6.0 grams per cm^2 Pb, has an energy[7] of 0.6 Mev.

TABLE I. *Transmutations and thick target yields.*

HALF-LIFE	REACTION	PROJECTILE ENERGY	PROJECTILES PER ACTIVE NUCLEUS
8 days	$Te^{128, 130}(d, n)I^{129, 131}$	8 Mev	5×10^6
25 min.	$Te^{128}(d, 2n)I^{128}$	8 Mev	2×10^7
13 hours	$Te^{130}(d, 2n)I^{130}$	8 Mev	1×10^7
4 days	$Sb^{121}(\alpha, n)I^{124}$	16 Mev	2×10^7
13 days	$Sb^{123}(\alpha, n)I^{126}$	16 Mev	1×10^7

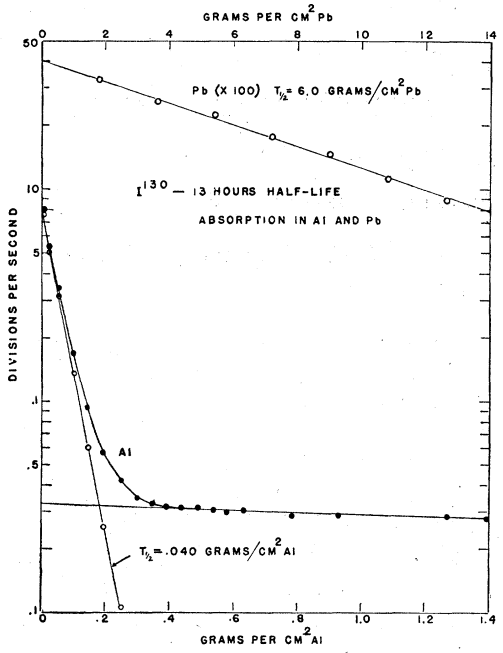

FIG. 10. Absorption curves of I^{130}.

YIELDS

In Table I are given the approximate yields for thick targets, corrected for the abundance in nature of the primary isotope involved. These figures have been derived from the duration and intensity of the bombardment, and the initial activity and an approximate calibration of the electroscope, and because of uncertainties in these factors, are necessarily rough.

We are happy to acknowledge the generous support of the Research Corporation and the Chemical Foundation which made this work possible. The work was facilitated by W.P.A. assistance.