Proton Activation of Indium and Cadmium

S. W. BARNES
University of Rochester, Rochester, New York
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Indium activated by 7.2-Mev protons exhibits two activities, one due to In^{115*} and one due to Sn^{113}. The other two activities previously reported have been shown to be due to impurities. Sn^{113} has a half-life of about 100 days and decays by K-electron capture emitting In K x-rays. An active In of 105 ± 10 min. half-life with a γ-ray of 0.39 Mev has been chemically separated from an aged Sn^{113} sample. An exactly similar In has been formed by proton bombardment of Cd which suggests the assignment of this activity to an excited state of stable In^{113}. The absence of Cd K x-rays indicates that In^{113*} decays to In^{113} with the emission of the 0.39-Mev γ-ray. In addition bombarded Cd shows the expected In isotopes In^{111} 20 min. (+), In^{114} 48 days (−) and In^{116} 54 min. (−). A positron emitter of 65 ± 5 min. half-life with a maximum positron energy of 1.6 ± 0.3 Mev is found and tentatively assigned to In^{110}. Activities of 72 sec. (−) and 2.7 ± 0.2 days (−) the latter accompanied by γ-rays of 170 and 250 kev are ascribed to isomeric states of In^{112}.

I. INTRODUCTION

THE various radioactive indium isotopes known at the present time have been formed in several ways, i.e., from the n-γ,[1] the n-$2n$,[2] the d-n,[2] the d-p[2] and the p-n[3] reactions. In addition one isotope In^{115*}[4] has been formed from stable In^{115} by nuclear excitation with neutrons,[5] protons,[6] x-rays,[7] and α-particles.[8]

A description of the known radioactive In isotopes which could be formed by proton bombardment of Cd is given in Table I. Of these radioactivities all but the 13 sec. have been found by proton activation of Cd. In addition activities ascribed to In^{110} and In^{113*} are found and described in the present report.* Proton bombardment of In gives a new period assigned to Sn^{113} which decays to In^{113*}.

II. APPARATUS

The Cd and In foils of 1 mil and 4 mils thickness, respectively, were bombarded within the cyclotron chamber with $\frac{1}{2}$ to 1μa beams of 7.2-Mev protons. In one instance a thick layer of indium was mounted on the probe for thirteen hours where it received a proton current of 20 to 30μa.

Activities were measured with an ionization chamber connected to a d.c. amplifier.[9] The chamber is closed with a window of Al of 3.4 mg/cm² thickness and 1″ diameter and is filled with Freon 12 to a pressure slightly more than one atmosphere. The system was calibrated with a standard source of U_3O_8 placed in a standard position above the window. With a grid resistance of 10^{11} ohms a deflection of 1 cm corresponds to a U_3O_8 source which emits 185 ± 10 β/sec. in all directions. When a floating grid is used, a rate of drift of 1 cm/sec. corresponds to a source emitting 110 ± 10 β/sec. in all directions. During the measurements on the long-lived decays the sensitivity was determined for each set of readings by means of a Ra E substandard. "Single sign" decay curves were obtained by mounting a small deflecting magnet over the window of the ionization chamber with the

TABLE I. *Some known radioactive isotopes of indium.*

ISOTOPE	HALF-LIFE	SIGN	e_{MAX}	γ	ACTIVITY RATIO	THRESHOLD
In^{111}	20 min.	+	1.75 Mev			
In^{112}	72 sec.	−				
In^{114}	50 days	−	2.15			
In^{115*}	4.1 hr.	γ		0.34		1.35 ±0.1 Mev
In^{116}	13 sec.	−	3.1			
	54 min.	−	1.4	1.4 Mev	1.12 ±06	

[1] Amaldi, D'Agostino, Fermi, Pontecorvo, Rosetti and Segrè, Proc. Roy. Soc. **149**, 522 (1935); A. C. G. Mitchell, Phys. Rev. **53**, 269 (1938).
[2] J. L. Lawson and J. M. Cork, Phys. Rev. **52**, 531 (1937); J. M. Cork and R. L. Thornton, Phys. Rev. **51**, 608 (1937).
[3] S. W. Barnes, Phys. Rev. **55**, 241 (1939).
[4] Szilard and Chalmers, Nature **135**, 98 (1935).
[5] Goldhaber, Hill and Szilard, Phys. Rev. **55**, 47 (1939).
[6] S. W. Barnes and P. W. Aradine, Phys. Rev. **55**, 50 (1939).
[7] Pontecorvo and Lazar, Comptes rendus **208**, 99 (1939); Collins, Waldman, Stubblefield and Goldhaber, Phys. Rev. **55**, 507 (1939).
[8] Lark-Horovitz, Risser and Smith, Phys. Rev. **55**, 878 (1939).
* These results supersede those of the preliminary report, reference 3.
[9] S. W. Barnes, Rev. Sci. Inst. **10**, 1 (1939).

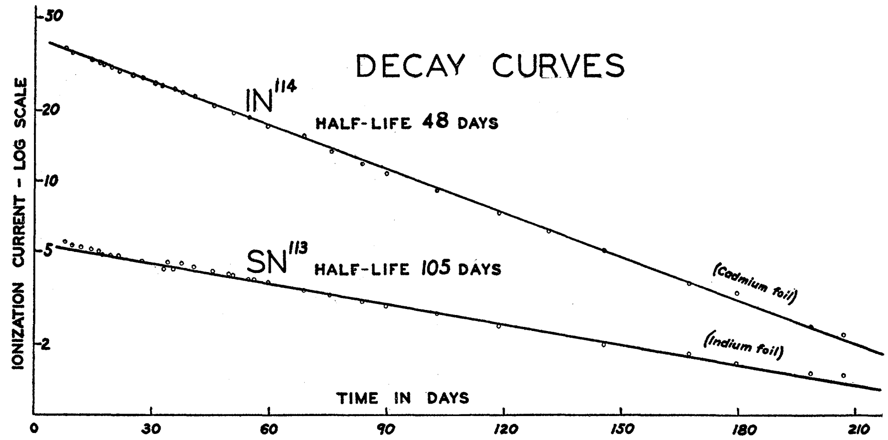

FIG. 1. Long-lived decay curves for Cd and In targets.

radioactive sample placed at the edge of the gap between the poles. For no magnetizing current the ionization current is produced by the γ-rays emitted by the source, though a contribution to this current of a few percent is made by β-particles scattered into the window from the air and from the cardboard covered poles of the magnet. About half of either the positive or negative electron distributions could be deflected into the window with the magnet used. The ordinates of the e^- and e^+ curves of Fig. 8 are the ionization currents observed with the magnetic field direct and then reversed, each reading then being reduced by the corresponding γ-ray ionization currents.

Measurements of β-ray and γ-ray energies were carried out with two β-ray spectrographs in one of which a photographic film and in the other a small ionization chamber was used as detector. These were of low resolving power because of the limited intensity of the thin Cd and In foils. The maximum and minimum radii of the spectrograph are 3.1 and 1.5 cm. The radius of the spectrometer is 7.8±0.1 cm. The window of its small ionization chamber is 0.15 cm wide and the source widths were between 1 and 2 mm.

The magnetic field of the instruments was calibrated with a fixed 50-mh mutual inductance supposedly good to 0.1 percent and a 1″ diameter flip coil constructed with care. ThB deposited on a 5-mil (0.005″) wire was used as a source of known lines. The $H\rho$ values of the five lines found on the film agreed within the experimental error with Ellis'[10] $H\rho$ values for the lines J, I, F, B and A. The energy range covered by this group, from 0.025 to 0.234 Mev, was very convenient for the present work.

III. Activities Observed After Proton Bombardment of Indium

A. The long-lived activity

Preliminary work showed activated indium to have a long-lived decay.* Since the 50-day half-life value of In^{114} is known fairly accurately a sample of this activity was produced by the p-n reaction with Cd and its decay followed for seven months along with the decay of activated indium. The curves shown in Fig. 1 indicate a half-life of 105 days for the aged In. This period has been given as 70 days[11] by Livingood and Seaborg. A small amount of a longer-lived activity in the present sample would account for this difference. Therefore, the study of the decay of the sample will be continued for at least another year.

The results of a chemical separation are shown in Fig. 2. Since the long-lived activity appears in the Sn fraction it is assigned to Sn^{113} (Sn^{115} being stable), formed by a p-n reaction with In^{113}.

After a two-week aging period a portion of the indium used in the probe bombardment was separated into Sn and In fractions.† Both

[10] C. D. Ellis, Proc. Roy. Soc. **138**, 318 (1932).
* Given in reference 3 as 50 days.
[11] J. J. Livingood and G. T. Seaborg, Phys. Rev. **55**, 667 (1939).
† The indium was dissolved in concentrated HCl, a few milligrams of stannic chloride were added, and the solution

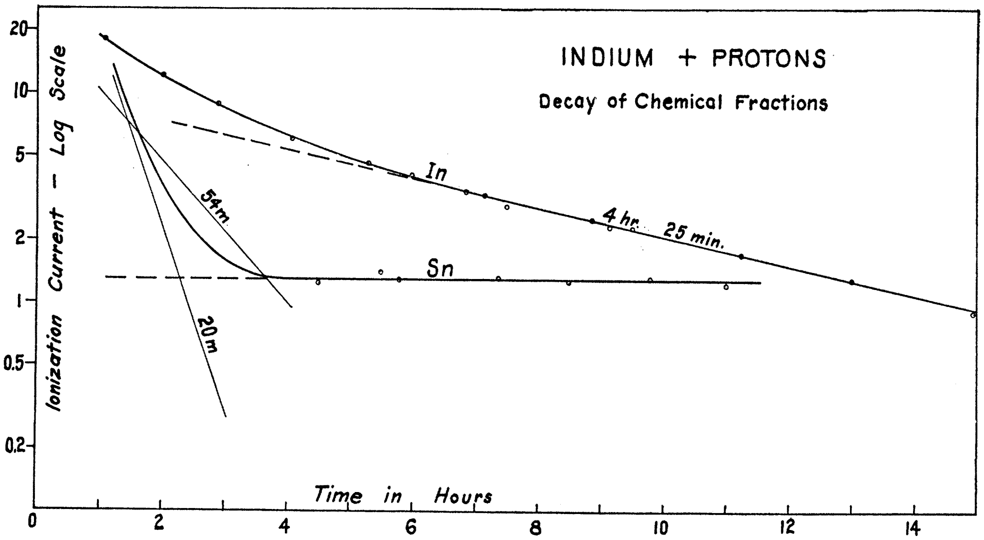

FIG. 2. Decay of In and Sn fractions from In target. (54-min. period is in In and 20-min. period in Sn.)

fractions showed activity, that of the indium decayed with a half-life of 105 minutes while that of the Sn grew with the same half-life. The data are plotted in Fig. 3. The broken line gives the intensity reached by the Sn fraction after two days. Time is reckoned from the moment of shaking the benzene water mixture.

Since Sn^{115} is stable and the In^{115*} 4-hr. activity is well known, the indium activity of 105 min. found in this experiment must be ascribed to In^{113*} formed by decay of Sn^{113} (105 days).

A well-aged proton-activated foil of indium was mounted close to the 1-mil Al window of a hydrogen filled cloud chamber. Some 400 negative electron tracks were measured and the histogram of Fig. 4 obtained. The absence of positron tracks indicates that Sn^{113} decays rarely if ever by positron emission. The shape of the distribution indicates that conversion electrons rather than negative β-particles are emitted, so that the decay is largely by K-electron capture.

A further investigation was made with a similarly aged but much stronger source in the β-ray spectrograph. Film 1 of Fig. 5 is a spectrogram obtained with an exposure of five days, beginning three weeks after bombardment. Electron lines corresponding to the K and L conversion electrons from a 0.39-Mev γ-ray are evident. In addition two weak lines are found on the original film which correspond to the K and L conversion of an 0.085-Mev γ-ray. There is no evidence either from the spectrograph or the cloud chamber of a continuous β-ray spectrum.

Sn^{113} must then decay entirely by K-electron capture and should emit In K x-rays. Evidence for these x-rays as well as for the γ-rays just

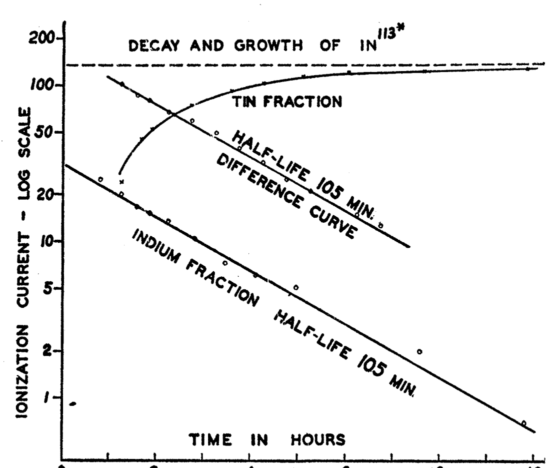

FIG. 3. Plots of the activity of Sn and In precipitates from an In target after three weeks' aging.

was diluted to about $\frac{1}{4}N$. Cupferron (ammonium nitrosophenylhydroxylamine) was then added to precipitate the tin, and the solution and precipitate were shaken up with benzene. The tin goes into the benzene layer, which was separated with a separatory funnel and evaporated to give the sample labeled Tin Fraction. The aqueous layer was neutralized, then made acid with acetic acid, and the indium was precipitated as the sulphide to give the sample labeled Indium Fraction.

mentioned is given by the absorption curve in Ag shown in Fig. 6. A 2-mm plate of beryllium placed between the source and the window stopped all electrons. The broken line is drawn through points corresponding to greater thicknesses of silver absorber than are shown on the plot. A comparison between the μ/ρ values obtained from the absorption curve and those calculated using the energy of the lines obtained from the spectrogram is given in Table II.

Although the agreement is only rough the difference in the absorption shown in Fig. 6 by the weighed Pd and Rh foils is satisfactory evidence for presence of In K x-rays. The nearly identical values of absorption shown by the Rh and Mo foils precludes the existence of Cd K x-rays, which could arise if In^{113*} decayed to Cd^{113} by positron emission or K-electron capture. This is confirmed by the absence of positrons in the cloud-chamber pictures.

Since (from Fig. 3) the activity to be ascribed to the Sn fraction at the instant of separation is very small the process must be the following. Sn^{113} captures a K electron, emits an In K x-ray quantum and becomes In^{113} in an excited state. It falls to the metastable state In^{113*} through the emission of the 0.085-Mev γ-ray. This metastable state has a half-life of 105 minutes and decays to the ground state of In^{113} with the emission of the 0.39-Mev γ-ray. From the ratio of intensity of the γ-ray ionization to that produced by the conversion electrons it is found that the coefficient of internal conversion for this 0.39-Mev line is about 70 ± 10 percent.

It may well be pointed out here that if this activity is correctly assigned to In^{113*} it should also be formed when Cd is bombarded by protons. This is found to be the case as will be shown in Section IV. Also it should be possible to raise In^{113} to the excited state In^{113*} with neutrons, protons, x-rays and α-particles as has been done for In^{115*}. The small relative abundance, five percent, may explain why In^{113*} has not been observed before.[12]

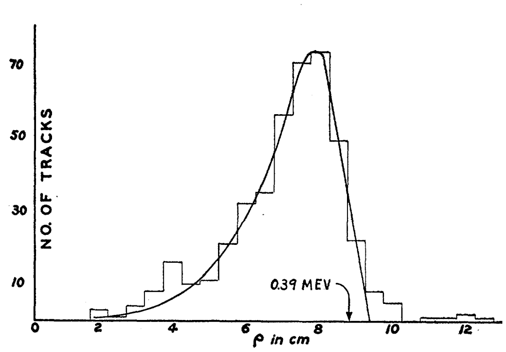

FIG. 4. Distribution of negative electron tracks in cloud chamber from In target one month after bombardment.

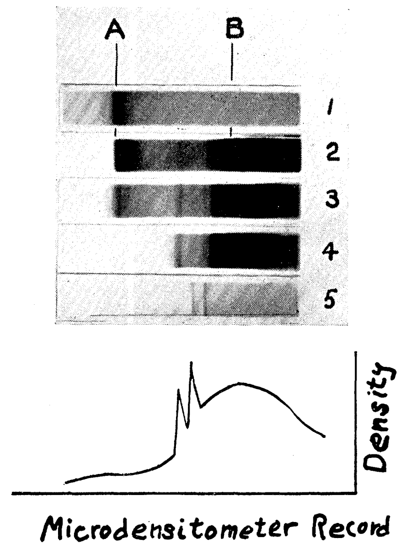

FIG. 5. β-ray spectrograms for Cd and In targets. Film 1 is for In target, 5-day exposure beginning 20 days after bombardment. Films 2–5 are for a Cd target the exposure intervals in terms of time after bombardment being as follows: film 2, 0 to 3.5 hr.; film 3, 3.5 to 15.5 hr.; film 4, 15.5 to 45 hr.; film 5, 30 to 34 days. The microdensitometer record below is for film 5 (not to same scale in the reproduction).

B. The short-lived activities

An early investigation showed a greater number of short-lived activities in proton activated indium than could easily be explained, unless some were assigned to impurities. The length of time required for good chemical separations together with the low initial activities made the solution of this problem by the ordinary chemical means difficult. Finally a sample of fairly pure indium* together with a quantitative statement of its impurities was acquired. The impurities consisted of 0.004 percent Cu and 0.005 percent Sn with negligible traces of a few

[12] G. T. Seaborg (private communication) has just reported finding a period of around 90 min. with a 0.4-Mev converted gamma-ray in fractions containing indium separated from old samples of Sn^{113} produced by the reaction Sn^{112} (d-p). This makes the assignment of this activity to In^{113*} almost certain.

* Indium Corporation of America, Utica, New York.

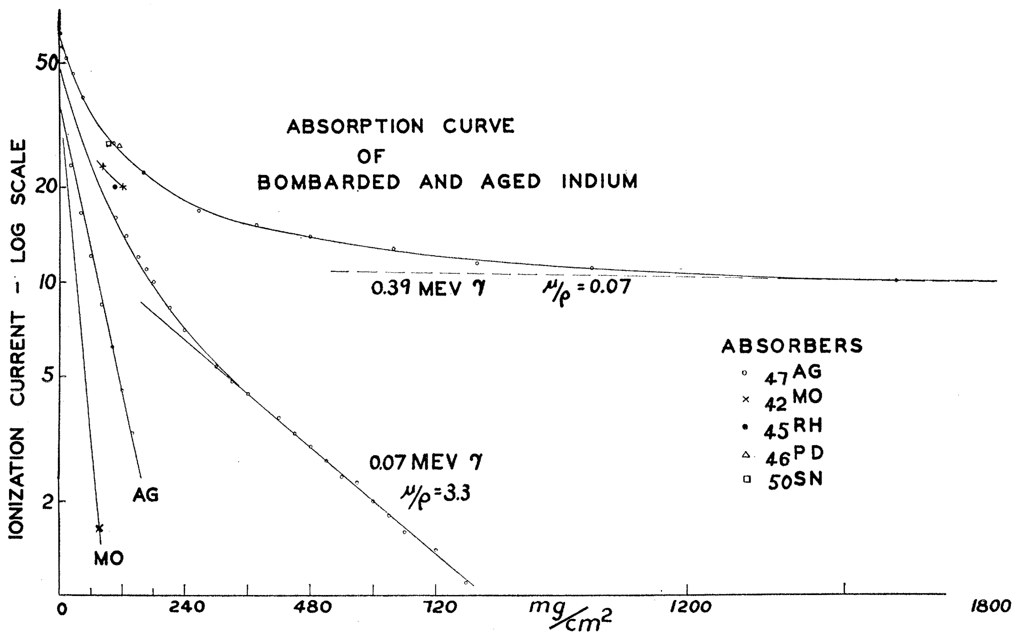

FIG. 6. Absorption curves for radiations from an aged In target. The β-rays were removed by a Be plate. The marked difference in absorption between the Pd and Rh foils shows the soft component to be In K x-rays.

other elements. The decay of two foils rolled from this indium is shown in Fig. 7. The curve In 1 is for a foil which faced the proton beam and received both protons and the neutrons present in the chamber. The In 2 curve is for a foil separated from foil 1 by a 10-mil sheet of lead and which therefore received only the neutrons.

The activity of foil 2 is seen to consist of the 54-min. period of In^{116} produced by In^{115} (n, γ) (the isomeric period of 13 sec. would not have been detected in this experiment) and the 4.1-hr. period of In^{115*} produced by neutron excitation. Presumably the same amounts of these two activities were produced by neutrons in foil 1. The excess 50-min. activity of foil 1 has been shown by single-sign decay curves to be a positron activity. The 0.005 percent of Cu present would give approximately this amount of 40-min. Zn^{63} activity through a p-n reaction with Cu^{63} so this period may be disregarded. Furthermore when the new and purer indium was substituted for the old—all other conditions of the experiment remaining the same, as indicated by the same initial activity of either the 4.1-hr. or 105-day activity—both the 18-min. and the 50-min. periods were found to have roughly one-tenth of their former activity. Thus the 18-min. activity which has previously been assigned[3] to an isomeric state of Sn^{113} is also due to an impurity (and is doubtless the 18-min. (e^+) activity of Sb^{120} formed[13] by a $(p$-$n)$ from Sn^{120}). Therefore the activities to be ascribed to indium following proton bombardment are In^{115*} formed by a p-p reaction with In^{115}, Sn^{113} formed by a p-n reaction with In^{113} and In^{113*} formed when Sn^{113} decays by K capture.

IV. ACTIVITIES RESULTING FROM PROTON BOMBARDMENT OF CADMIUM

All the activities observed in a bombarded Cd foil have been found in the indium precipitate after a chemical separation.

TABLE II. *Absorption coefficients for x- and γ-rays.*

Source	Line Mev	Absorber	μ/ρ Observed	μ/ρ Calculated
In^{113*} γ-ray	0.39	Ag	0.07	0.07
Sn^{113} γ-ray	0.085	Ag	3.3	4.4
Indium K x-rays	0.024	Ag	17	10
	0.024	Mo	40	50

[13] G. T. Seaborg, private communication.

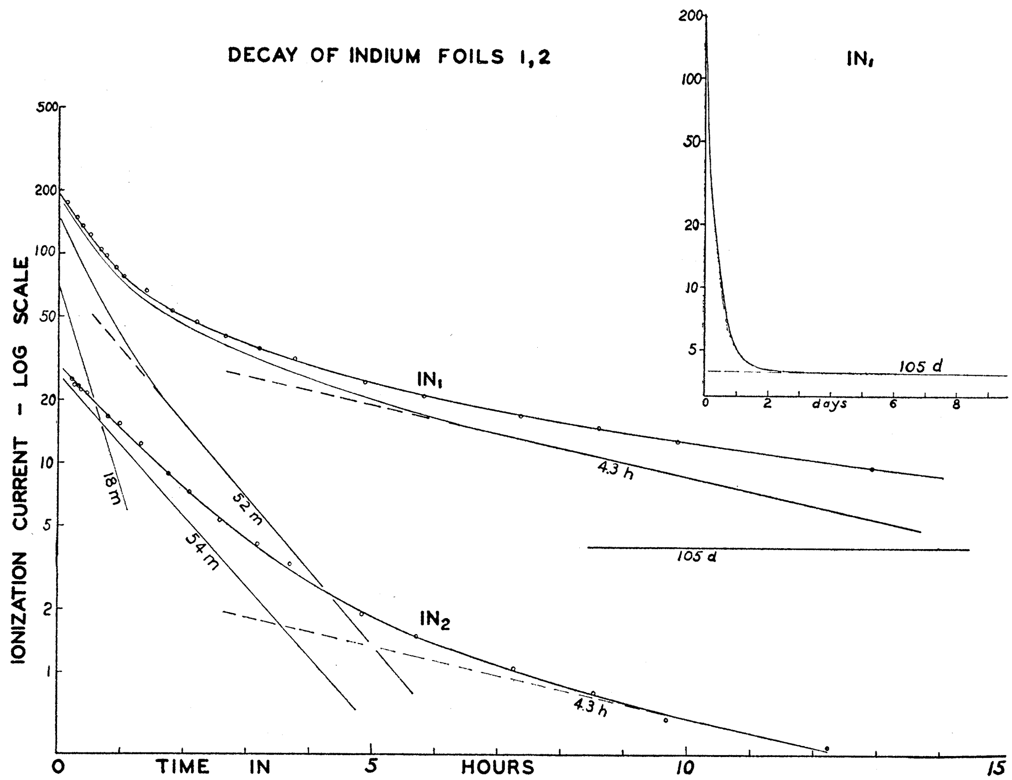

FIG. 7. Decay curves for bombarded In. In_1 is for a foil receiving both protons and neutrons; In_2 is for a foil receiving neutrons only.

In Fig. 8 are shown the single sign and γ-ray decay curves of the various activities observed in a proton-bombarded Cd foil. Fig. 5 shows spectrograms of the negative electrons obtained at the times indicated with the β-ray spectrograph. These are to be compared with the e^- decay curve of Fig. 8. The components of this e^- curve which may be immediately assigned are the 54-day activity which is due to In^{114} formed by Cd^{114} (p-n) and the 51-minute which is due to In^{116} formed by Cd^{116} (p-n). Film 5 of Fig. 5 and the microdensitometer record immediately below belong to In^{114} (50 day.) Two electron lines are evident which are assumed to be K and L conversion lines of a 0.195-Mev γ-ray although the L conversion is unusually strong.

The activity with the half-life of 105 ± 5 minutes seemed to be associated with the highly converted highest energy (0.39 Mev) γ-ray seen at A on films 2 and 3 of Fig. 5. This could not be proved, however, until the data of Fig. 9 were obtained, which represents three out of nine runs made at successive times with one sample on the spectrometer. Only two lines (marked A and B on Figs. 5 and 9) were intense enough to be measured. Line B which is plainly evident on the original film 2 unfortunately does not show on the print of Fig. 5. At the left of Fig. 9 are decay curves of these two lines, the ordinates represent line intensities, at the various times, obtained by subtracting from the peak of the line a reasonable base line. The value of the half-life of line A (115 ± 15 min.) definitely associates this conversion line with the 105-min. period of the e^- decay curve of Fig. 8. This activity is seen to be identical in its half-life and γ-ray* with the activity which grew from the Sn^{113} obtained from an indium target. It is ascribed therefore to In^{113*} formed by a p-n reaction with Cd^{113}.

The low energy (0.16 Mev) γ-ray of Fig. 9 with a half-life of 23 min. is undoubtedly to be

* On the basis of the assumed level scheme the 0.085 γ-ray should not be present here; however this could not be checked because of the presence of other strong lines in this region.

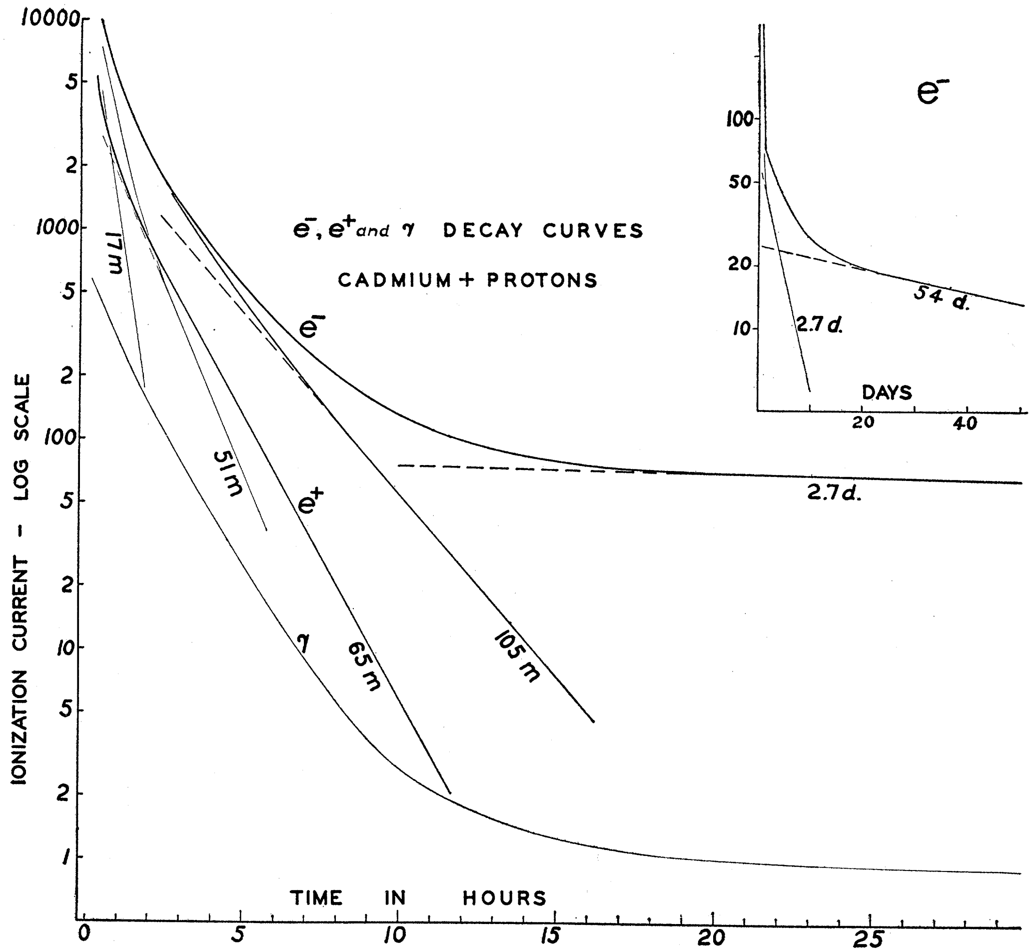

FIG. 8. Single sign and γ-ray decay curves for bombarded Cd.

associated with the 18- to 20-min. positron component always found in the e^+ decay curve. This component is not shown in Fig. 8 but arises when the 65-min. component is subtracted from the upper portion of the e^+ curve. This activity has been found by Cork[2] and assigned to In^{111}.

The 17-min. component of the e^- curve is thought to be due to the conversion electrons of In^{111} rather than to any new β-ray emitter. This is reasonable in view of the geometry of the apparatus even though the e^- 17-min. curve is more intense than the corresponding e^+.

The positron activity with half-life of 65 ± 5 min. has not been previously reported. A rough value of the maximum energy of the positrons has been determined by using both the spectrograph and spectrometer and is 1.6 ± 0.3 Mev. It has been found impossible to assign with certainty any of the conversion electron lines of Fig. 5 to this activity. In^{106}, In^{108} and In^{110} must be positron emitters, and since Cd^{110} is ten times as abundant as either Cd^{106} or Cd^{108} this activity is tentatively assigned to In^{110}. If this assignment is correct the activity should be produced by an α-n reaction with Ag.

The 2.7-day component of the e^- curve has two conversion electron lines (see film 4 of Fig. 5) corresponding to γ-rays of 0.253 and 0.168 Mev. Cork[14] has reported a similar activity after fast neutron bombardment of indium. If his activity is the same as the one found here it can be

[14] Cork and Lawson, Washington Meeting 1939. (Verbally reported; figures are not given in the abstract, Phys. Rev. 55, 1136 (1939)).

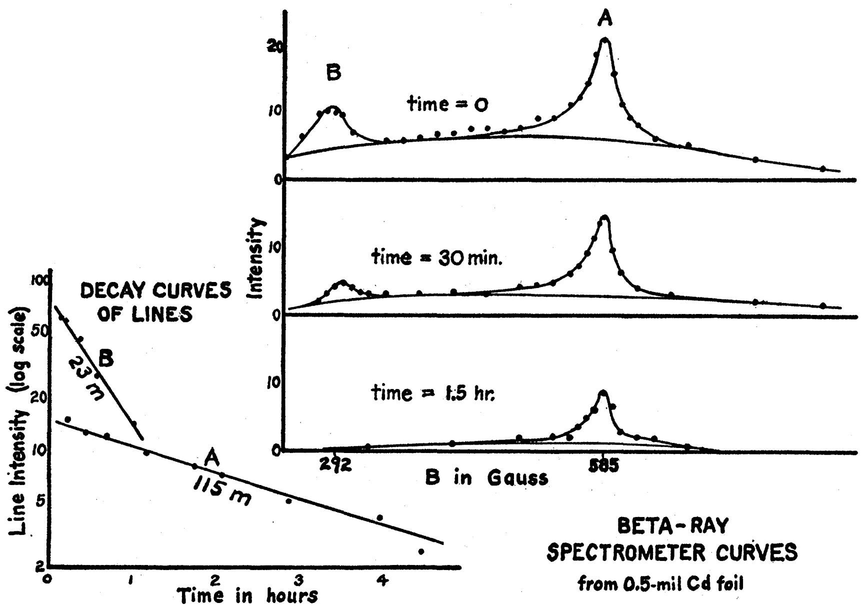

FIG. 9. Successive β-ray spectrometer curves for bombarded Cd, showing (lower left) decay curves for each line. Line *A* represents *K* and *L* conversion of a 0.39-Mev γ-ray (In^{113*}); line *B* for a 0.16-Mev γ-ray (In^{111}).

explained as an isomeric state of In^{112} (for which a period of 72 sec. has been reported)[2] formed in his case by an (*n*-2*n*) reaction and here by a (*p*-*n*) reaction. The 72-sec. activity has been found in the present work in total decay curves of cadmium targets.

TABLE III. *Summary of radioactive isotopes in* In *and* Cd *targets.***

ISO-TOPE	HALF-LIFE	SIGN	e_{MAX} IN MEV	γ IN MEV	PROTON CROSS SECTION FOR 6.9 MEV $CM^2 \times 10^{26}$
In^{110}	**66 ±5 min.**	e^+	**1.6 ±0.3**		**3***
In^{111}	20 min.	e^+	2.15	**0.16 ±0.01**	4
In^{112}	72 sec.	e^-			**3***
	2.7 days	e^-?	1.73?	**{0.253 0.168**	**0.4**
In^{113*}	**105 ±10 min.**	γ		**0.39 ±0.02**	**2**
In^{114}	48.5 ±2 days	e^-	1.75	**0.19 ±0.01**	**3**
In^{115*}	4.1 hr.	γ		0.34[19]	**0.003**
In^{116}	(13 sec.)				
	54 min.	e^-			**3***
Sn^{113}	**105 ±15 days**	**K** capture		**0.085 ±0.01**	**3**

* These values are least certain.
** Figures in **bold face** are those reported for the first time in this paper. Others have been previously reported and are here confirmed.

V. SUMMARY

The results of this work are summarized in Table III, together with the data contained in Table I. The cross section values are not precise; however, they do indicate a striking constancy for the various *p*-*n* reactions. The small cross section found for the 2.7-day activity thus becomes a justification for the assignment of this activity to one of two isomeric states. The cross section for the one *p*-*p* reaction (In^{115*}) is seen to be about a thousand times smaller than that for the others which are all *p*-*n* reactions. Since the incident protons of 7.2 Mev lose about 0.6 Mev in a 1-mil foil of Cd or In these cross sections correspond to a mean energy of the protons of about 6.9 Mev.

It is a pleasure to give acknowledgment here for the valued suggestions of Dr. S. N. VanVoorhis, Professor L. A. DuBridge, and Dr. V. Weisskopf. The work was financed in part by a grant from the Research Corporation.

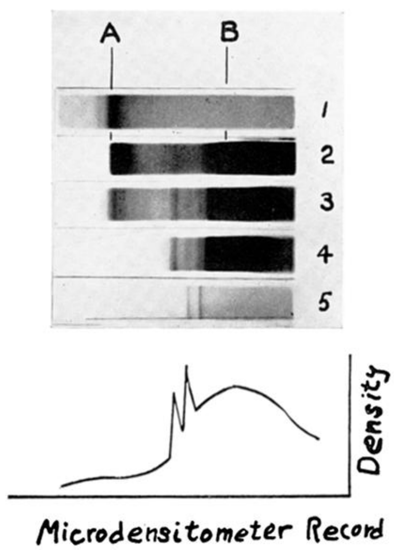

FIG. 5. β-ray spectrograms for Cd and In targets. Film 1 is for In target, 5-day exposure beginning 20 days after bombardment. Films 2–5 are for a Cd target the exposure intervals in terms of time after bombardment being as follows: film 2, 0 to 3.5 hr.; film 3, 3.5 to 15.5 hr.; film 4, 15.5 to 45 hr.; film 5, 30 to 34 days. The microdensitometer record below is for film 5 (not to same scale in the reproduction).