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Transmutation of Mercury by Fast Neutrons

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The products of the transmutation of mercury by fast (Li+D) neutrons were investigated. Three radioactive gold isotopes were found to be formed by the $n-p$ reactions: 65-hour Au^{198}, 78-hour Au^{199}, and a new unassigned electron emitter with a period of 48 min. Three radioactive platinum isotopes were observed and attributed to $n-\alpha$ reactions in mercury: 29-min. Pt^{199}, 19-hour Pt^{197}, and a new activity with an 80-min. half-life. The latter period was also observed in deuteron and fast neutron bombardment of platinum, but was not found in slow neutron irradiation of platinum. Radioactive mercury isotopes with 43-min., 25-hour, and 50-day periods were observed in the fast neutron irradiation of mercury. The 43-min. and 25-hour activities were also formed by bombarding platinum with α-particles. Difficulties in the assignment of the 80-min. Pt and the 43-min. and 25-hour Hg activities are discussed.

INTRODUCTION

THE bombardment of nuclei by fast neutrons may result in radioactive nuclei following simple neutron capture or transmutations involving the emission of neutrons, protons and α-particles. The emission of neutrons has been observed for practically every element. Radioactivity following the emission of heavy charged particles has been observed for the light and medium weight nuclei, but the number of such observations falls rapidly with the increasing atomic number of the elements bombarded.[1] This trend can be attributed largely to the effect of the increasing potential barrier on emission of protons and alpha-particles. The energy requirement for an $n-p$ reaction is

$$E_n = E_{\max} - 0.8 \text{ Mev}$$

where E_n and $E_{\max}$ represent the neutron threshold energy and the energy of the radioactive transition, respectively, and 0.8 Mev is the energy equivalent of the difference in mass between the neutron and proton. The $n-p$ reaction will ordinarily be slightly endoergic. The $n-\alpha$ reactions will in general be exoergic because of the high internal binding energy of the α-particle. Despite the fact that the energy considerations favor the $n-\alpha$ and $n-p$ reactions over the $n-2n$ reaction, the effective cross sections for the former are greatly reduced by the high potential barriers.

As a consequence of the rapid decrease of cross section with increasing atomic number, the $n-p$ and $n-\alpha$ reactions require very high neutron energies and large quantities of target material to produce sufficient activity for experimental study. The most energetic neutrons available are those produced in the bombardment of lithium

[1] G. T. Seaborg, Chem. Rev. **27**, 199 (1940).

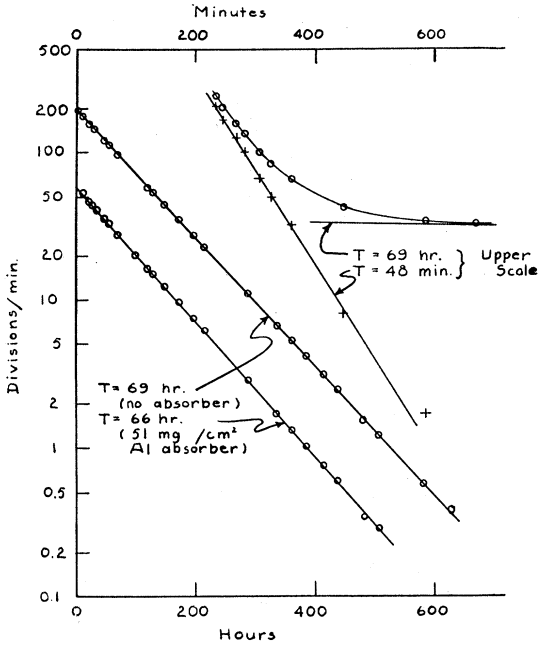

FIG. 1. Radioactive gold from Hg+Li neutrons.

by deuterons. However, even for these, the cross sections are very small and large quantities of material must be used. This, in general, complicates the chemical treatment required to identify the resulting radioactive nuclei. In this respect mercury is an ideal element, since it has been shown that microscopic quantities of gold and platinum can be easily separated from mercury by distilling the mercury in vacuum.[2]

METHOD

Three to four hundred grams of mercury were bombarded with fast neutrons from the Li+D reaction. The deuterons used were produced by the Harvard cyclotron with an energy of 11 Mev and currents of 5–15μa. After irradiation, the mercury was placed in a vacuum distilling flask with the addition of 50 mg of gold. The latter readily formed an amalgam with the mercury and served as a carrier for the radioactive gold and platinum. The residue of the distillation was then treated chemically to separate the gold and platinum activities.

The mercury had been subjected to a number of distillations before use and gave results in complete quantitative agreement with samples of C.P. analyzed mercury, indicating a high state of purity. Furthermore, the same mercury was used repeatedly with concordant results, indicating that the distilling process carried out at the rate of approximately 50 cc per hour was indeed removing at least 99 percent of the added gold. Control tests made by bombarding Pt and Au simultaneously with the mercury showed that the amounts of Pt and Au which would have to be present in the mercury as impurities were of the order of 10 mg of Au and 250 mg of Pt. Impurities of this magnitude are impossible, as shown by the fact that a second distillation yielded an activity of less than 0.1 div./min., while the first distillation gave 30 div./min. for the gold fraction and 6 div./min. for the platinum fractions.

The activities were observed with a Lauritsen electroscope of the conventional type. The radioactive samples were placed on aluminum holders and covered with Cellophane tape. The aluminum holders could be inserted into a series of shelves directly below the 1-mil Al electroscope window. The samples were as thin as possible to prevent self-absorption, but, in general, the radiations had to penetrate a thickness of 15 mg/cm^2 (window+tape). The sign of the emitted electrons was determined by deflecting them by means of a magnetic field into a Geiger counter.

It should be noted that any radioactivity with half-life values less than 15 min. would have been difficult to observe, since the distillation and the chemical separation required about two hours.

TABLE I. *Stable isotopes of platinum, gold, and mercury.**

Z	ELEMENT	A	% ABUNDANCE	Z	ELEMENT	A	% ABUNDANCE
78	Pt[a]	192	0.8	80	Hg[c]	196	0.15
		194	30.2			198	10.1
		195	35.3			199	17.0
		196	26.6			200	23.3
		198	7.2			201	13.2
						202	29.6
						204	6.7
79	Au[b]	197	100				

[2] A. Miethe, Naturwiss. **12**, 597 (1924); **13**, 635 (1925); M. W. Garrett, Proc. Roy. Soc. **A112**, 391 (1926).

* [a] M. B. Sampson and W. Bleakney, Phys. Rev. **50**, 732 (1936); [b] A. J. Dempster, Nature **136**, 65 (1935); [c] A. O. Nier, Phys. Rev. **52**, 933 (1937).

Chemical Procedure[3]

The residue (gold) of the mercury distillation was dissolved in aqua regia and platinum carrier added. The solution was adjusted to 1.2 N in hydrochloric acid after complete removal of nitric acid. Gold was precipitated with hydroquinone and removed by filtration. Platinum was reduced by hydrazine.

For a more critical study of the platinum activities, the following methods were used. After removal of the gold by hydroquinone, the latter was destroyed by nitric acid. Platinum and iridium carriers were separated by the use of sodium bicarbonate with bromphenol blue indicator.[3] Iridium hydroxide precipitated out at this point. (Other metals of the platinum group, except platinum, would also be precipitated.) Platinum was then recovered from the solution by the use of hydrazine. This method was also used to prepare "spectroscopically pure" platinum. An alternative method was the addition of potassium chloride solution to precipitate potassium chlorplatinate and potassium chloriridate in the presence of other metallic carriers (iron, calcium, sodium, mercuric and gold chlorides.) The potassium chlorplatinate and chloriridate were reduced to the metals by hydrazine. After a sodium peroxide fusion, the resulting hydroxides were dissolved in hydrochloric acid. Platinum and iridium were then separated by the sodium bicarbonate method (above).

Results

(a) Gold

A glance at the isotope chart (Table I) for this region of the periodic table indicates that at least six radioactive gold isotopes may result by $n-p$ reactions in mercury: 198, 199, 200, 201, 202, and 204 (the abundance of Hg^{196} is so small that it could scarcely yield an observable activity under the fast neutron bombardment conditions of this experiment). Au^{198} is the well-known activity of half-life, 2.7 days.[1] Au^{199} has a half-life of 3.3 days and has been observed by McMillan, Kamen and Ruben,[4] as the product of the decay of the 31-min. Pt^{199}. Cork and Halpern[5] have reported a gold period of 164 days which they assigned to Au^{199}. The gold isotopes of higher mass number have not been observed.

The gold fraction in the present experiment gave the decay curve shown in Fig. 1. Two activities are apparent, one of 48±2-min. half-life and one with a 69-hour half-life. However, when the longer period activity was observed through a 50-mg/cm² aluminum absorber, a value of 66 hours was obtained. This indicated

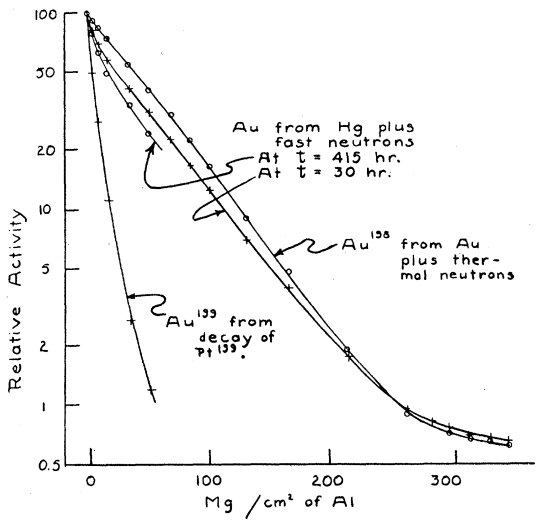

Fig. 2. Absorption in aluminum of 69-hour gold from Hg+Li neutrons, 65-hour Au^{198} and 78-hour Au^{199}.

that the 69-hour activity was actually complex, despite the fact that the decay was linear within the limits of accuracy over a factor of 500 in intensity. This complexity was also indicated by the absorption data in Fig. 2, which show that the radiation was softer at $t=415$ hours after irradiation than at $t=30$ hours. Absorption curves for the 65-hour Au^{198} and 78-hour Au^{199} were obtained by bombarding gold with slow neutrons, and platinum with slow neutrons. In the latter case, the gold was separated after allowing the platinum to decay for a number of hours. These absorption curves are also shown in Fig. 2. It was found that both the 69-hour decay curve (Fig. 1) and the absorption curve at 30 hours (Fig. 2) of the gold obtained from the mercury could be fitted by a mixture of 75 percent Au^{198} (65 hours) and 25 percent Au^{199} (78

[3] See Beamish, Russell, *et al.* Ind. Eng. Chem. (1936–37).
[4] McMillan, Kamen, and Ruben, Phys. Rev. **52**, 375 (1937).
[5] J. M. Cork and J. Halpern, Phys. Rev. **58**, 201A (1940).

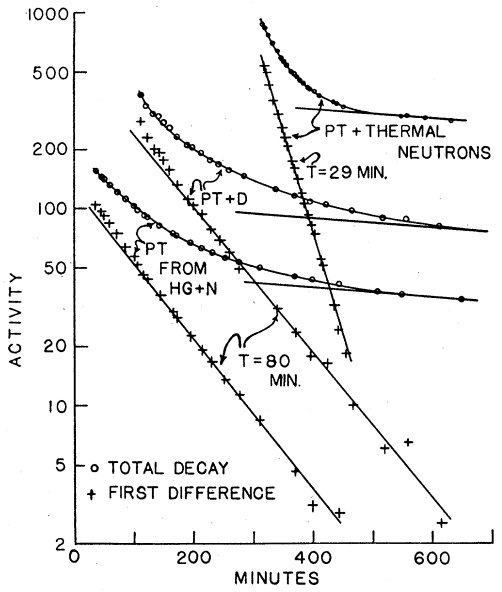

FIG. 3. Short period platinum activities produced by Hg+Li neutrons, Pt+D and Pt+slow neutrons. In each "total decay" curve, the background is due to the 19-hour +longer periods. The departure of points from the 80-min. lines is due to the 29-min. activity.

hours). Actually, the intensities of the 65-hour and 78-hour activities would be roughly equal if one corrected for the absorption in the material covering the samples (15 mg/cm^2). Both isotopes emit negative electrons. The 65-hour electrons have a maximum energy of 0.8 Mev, while the 78-hour electrons have an energy of several hundred kilovolts.

The 48-min. activity emits negative electrons with a maximum energy of about 2.5 Mev. Its absorption curve is shown in Fig. 6. It is observed to be much more energetic than the 43-min. Hg activity and, therefore, could not be confused with the latter. This gold activity must be assigned to a gold isotope of mass number >199. On reducing the observed activities to the activities for infinite bombardment time, one finds that all three periods are formed with approximately the same intensity. No evidence for a 164-day period was found, although it would have been observable if its formation had a comparable cross section. The energy ascribed to this activity is 0.45 Mev[5] and the emitted particles would not have been absorbed to an appreciable extent by the electroscope window and Cellophane tape.

(b) Platinum

The platinum fraction from the mercury bombardment gave a decay curve with four observable periods. A long period (>2 days) of weak intensity was observed but its relative intensity was erratic, indicating that it might be due, at least in part, to a slight gold contamination. The other periods observed were 19 ± 1 hours, 80 ± 5 min. and 27 ± 5 min., with approximately equal intensities (estimated for infinite bombardment time). All three emit negative electrons.

The isotope chart shows that the following mass numbers in Pt can result from an $n-\alpha$ reaction in mercury: 195, 196, 197, 198, 199, 201. McMillan, Kamen, and Ruben[4] found and assigned a 31-min. period to Pt^{199} and an 18-hour period to Pt^{197}, by neutrom bombardment of Pt. The same periods are found in platinum activated by deuterons.[6] A deuteron bombardment was made in the present work and an absorption curve for the observed 19 ± 1-hour period was obtained. By comparison with the absorption curve of the 19-hour Pt resulting from the Hg+n reaction, it was established that they were the same. The reaction is ascribed to Hg^{200}. A satisfactory identification of the 27 ± 5-min. activity by absorption measurements was not possible, since its intensity was too small by the time the activity could be measured (about 2 hours after bombardment). However, it seems probable that it is the 31-min. Pt^{199}, formed by an $n-\alpha$ reaction from Hg^{202}.

No 80-min. period in Pt has previously been reported. In the course of this work it was found that this activity could be produced by bombarding Pt with deuterons (Fig. 3) and with fast neutrons. It is surprising that slow neutron

TABLE II. *Relative intensities of three platinum activities as produced by different reactions.*

REACTION	SUPERFICIAL DENSITY OF SAMPLE	29 ± 1 MIN.	80 ± 3 MIN.	19 ± 1 HR.
Hg+Li neutrons	15 mg/cm^2	1	0.8	0.9
Pt +D	20 mg/cm^2	1	1.4	3.7
Pt +Li neutrons	150 mg/cm^2	1	3.5	6.0
Pt +slow neutrons	150 mg/cm^2	1	not observed (<0.1)	0.3

[6] J. M. Cork and E. O. Lawrence, Phys. Rev. **49**, 788 (1936).

bombardment failed to produce the 80-min. activity (Fig. 3). A number of chemical tests (above) were made before and after irradiation in order to establish the chemical identity of this period. The results indicated that the 80-min. activity was due to a Pt isotope.

The relative intensities for infinite time of irradiation (but not corrected for absorption) are shown in Table II, for the various reactions studied. The results for fast and slow neutrons are consistent with the assignment of the 29-min. period to Pt^{199} and the 19-hr. period to Pt^{197}, since the latter can be formed by an $n-2n$ reaction from Pt^{198}, whereas the former can be formed only by neutron capture. The assignment of the 80-min. period is difficult. If the failure to observe it with slow neutrons is taken to indicate that it cannot be formed by simple capture, then it must be assigned to Pt^{198*} and would have to be formed by deuterons by a reaction $Pt^{198}(D, ?)$ Pt^{198*}; that is, by some form of excitation. In order to check this possibility, a stack of 6 Pt foils (1 mil) was irradiated with deuterons and the activity of each foil analyzed. Since a number of long period gold activities are formed (the 37-min. positron emitter reported by Cork and Halpern[5] in Au was not found), only the 29-min. and the 80-min. periods were compared (Table III). The first three foils gave the same ratio of

TABLE III. *Stacked foil bombardment of platinum. The energy values are the mean energy of the deuterons. The "energy thickness" of each foil is about* 1.7 *Mev.*

FOIL	1	2	3	4	5	6
Mean energy within foil	10 Mev	8.3	6.5	3.9	1.5	0
29-min. activity*	3.5×10^4	1.3×10^4	1.0×10^3	~ 7	~3.6	3.6
80-min. activity	4.6×10^4	1.4×10^4	1.3×10^3	~14	~7.2	7.2

* The activities listed here correspond to a time of 120 min. after irradiation. These data were obtained with an ionization chamber and cannot be directly compared with the data of Table II which were observed with the electroscope, because of differences in the sensitivity of the two instruments.

the 80-min. to 29-min. period within 25 percent, while the relative activity of the foils decreased by a factor of 30. The low activity of the fourth foil shows that the yield drops very rapidly below 6 Mev. The fourth foil had an activity about twice that of the fifth and sixth foils, which show the neutron background activity. This, incidentally, shows that fast neutrons, which are always present during deuteron bombardment, can be responsible for only a fraction

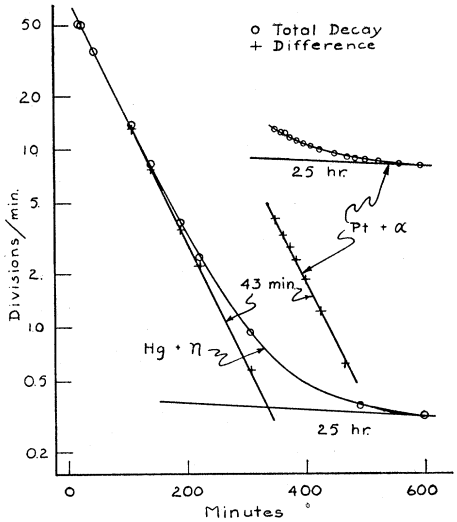

FIG. 4. 43-min. Hg activity as produced by mercury+lithium neutrons and platinum+α-particles.

of a percent of the observed 80-min. activity. This experiment indicates that the 80-min. and the 29-min. activities are formed by similar reactions, although it is, perhaps, possible that the cross section for some type of excitation process might have the same energy dependence as that for a (D, P) reaction. The high yield at relatively low deuteron energies makes it improbable that the 80-min. period is being formed by a (D, H^3) or $(D, 2n, p)$ reaction.

It is possible that the cross section for the production of the 80-min. period by slow neutron capture is finite but small. If its cross section were only a few percent of that for the formation of the 29-min. period it could easily escape observation, because of masking by the 29-min. and 19-hour activities. Attempts to find the 80-min. period by bombarding osmium with α-particles and iridium with deuterons were unsuccessful. These results suggest that the 80-min. period may be assigned to Pt^{196*} or Pt^{197}.

Absorption data show that the maximum electron energies of the 19-hour and 29-min. activities are 0.65 Mev, and 1.8 Mev, respectively. The absorption curve for the 80-min. period as produced by deuterons is concave in shape and indicates that it is probably composed of a line or lines, with an energy of several hundred kilovolts.

McMillan *et al.*[4] observed a 3.3-day Pt period of equal intensity with the 19-hour Pt^{197}, when platinum was irradiated with slow neutrons. We have observed a long period of small intensity, but it is possible that the electroscope window and Cellophane tape absorbed most of the 3.3-day electrons.

(c) Mercury

McMillan, Kamen and Ruben[4] reported 43-min. and 25-hour activities induced by fast neutrons on mercury. In the present work an additional weak period of about 50 days was observed but not studied. Krishnan[7] found by deuteron bombardment of mercury strong activities of 5 min. and 48 min., and weaker activities of 36 hours and 60 days. In addition, he found that deuteron bombardment of gold yielded a mercury activity of 36 hours. In a more recent publication,[8] he reports the mercury activity from deuteron bombardment of gold as having a period of 32 ± 2 hours, which he ascribes to Hg^{198*}, on the basis of selective absorption measurements on the γ-radiations, formed by $Au^{197}(D, n)$.

Since the present work gave a value of 25 ± 1 hours for the neutron-induced mercury activity, a deuteron bombardment of gold was made to see whether our 25-hour and Krishnan's 32-hour activities actually were different. The decay curve of the chemically separated mercury gave a period of 25 ± 1 hours and a longer weak period of 60–70 hours which was ascribed to a slight contamination of 65-hour Au^{198}.

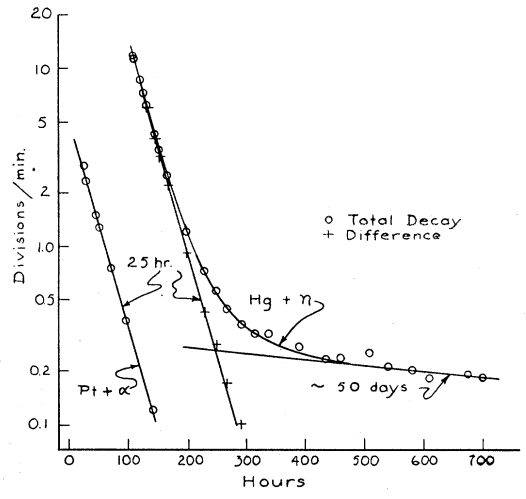

FIG. 5. 25-hour activity as produced by Hg+Li neutrons and Pt+α. A decay curve identical with the Pt+α curve was obtained with Au+D except for the presence of a very weak activity of longer life.

If the assignment of the 25-hour period to Hg^{198*} were correct, one might expect it to be formed by the decay of the 65-hour Au^{198}. A slow neutron bombardment gave a very intense 65-hour activity and, after several days, a small amount of mercury was added to the gold in solution and then precipitated. The mercury showed no detectable activity above the background indicating that the transition $Au^{198} \rightarrow Hg^{198*}$ occurred to less than 1 part in 14,000. This was surprising, since the gold has sufficient energy (0.8 Mev above the ground state of Hg^{198}) to decay to a 25-hour isomer, which has a much lower energy (0.2 Mev).

Bombardment of platinum with α-particles gave two periods of 43 min. (Fig. 4) and 25 hours (Fig. 5) (without chemical separation), with a corrected intensity ratio of 1 : 50. The absorption data for the 25-hour activity (Fig. 6) produced by Hg+n, Au+D, and Pt+α are in excellent agreement, and indicate an energy of approximately 200 kev (the Au+D data are not shown in Fig. 6).

Valley[9] examined the 25-hour activity produced in the Au+D reaction by means of a β-ray spectrometer and found that it was composed of a number of conversion lines whose energies agree very well with the x-ray level separations in gold rather than in mercury. These results indicate that the 25-hour activity is to be assigned to Hg^{197} formed by $Au^{197}(D, 2n)$, which decays by K capture to Au^{197}. Valley also found an activity of 62 hours in the mercury precipitate, consisting of very soft electrons. This period was not observed in the present work because of the thickness of the electroscope window. It is possible that Krishnan's value of 32 hours was the 25-hour period affected by the presence of the 62-hour activity.

The assignment of the 25-hour period to Hg^{197} is consistent with the present investigations

[7] R. S. Krishnan, Proc. Camb. Phil. Soc. **36**, 490 (1940).
[8] R. S. Krishnan, Proc. Camb. Phil. Soc. **37**, 186 (1941).
[9] G. E. Valley, private communication.

since this activity can be formed by the reactions

$$Hg^{198}(n, 2n)\ Hg^{197}, \quad Pt^{194}(\alpha, n)\ Hg^{197},$$

and explains the failure to observe the growth of the 25-hour activity from the decay of Au^{198}.

The 43-min. Hg activity has been assigned to Hg^{197}.[1,7] However, we did not observe it with bombardment of gold by deuterons. One would expect it to show up fairly strongly as a $(D, 2n)$ product if it were isomeric with the 25-hour period, and even more strongly if its correct assignment were Hg^{198*}, formed by a (D, n) reaction. Failure to observe this result from the bombardment of gold by deuterons would indicate a higher mass number than 198; the only numbers consistent with its production by $Pt+\alpha$ are 199 and 201, which can be formed from $Pt^{196 \text{ or } 198}$. This reasoning would, therefore, assign the 43 min. to an excited state of stable $Hg^{199,201}$. However, either of these should be formed by neutron capture in $Hg^{198,200}$. This slow neutron reaction has not been reported by other investigators. Observations made in the present work are obscured somewhat by the difficulty of eliminating all fast neutrons. Indications are that the cross section for slow neutrons is small.

Summarizing, the cross section for the formation of the 43-min. activity seems to be "small" for the reaction Hg+slow neutrons, "small" for $Pt+\alpha$ (1/50 of 25-hour activity), "large" for fast neutrons on Hg (25 times the 25-hour activity with Li neutrons), and "large" for its formation by $Hg+D$.[7] These changes cannot be attributed to the relative abundance of the initial nuclei concerned in these reactions.

It would be of interest to know the relative cross sections for the various fast neutron reactions. Unfortunately, the neutrons coming from the thick lithium target have an energy spread of 20–25 Mev. In view of this inhomogeneity, only rough measurements on the relative yields were made, corrected to infinite bombardment time and for absorption. The 29-min., 80-min., and 19-hour activities in platinum have equal intensities, each of which is 10^{-4} as large as the yield of the 43-min. Hg activity. The 48-min., 65-hour, and 78-hour gold activities have equal intensities, each being 10 times stronger than the platinum, and 10^{-3} as strong as the 43-min. mercury. The 25-hour intensity is 4 percent of the 43-min. mercury activity, which was 3000 div./min. per g of mercury for $10\mu a$ of deuterons at 11 Mev on Li. With Be neutrons, which have about half the energy, the yield of the 43-min. activity increases slightly, while the intensity of the gold yield drops by a factor of 7.

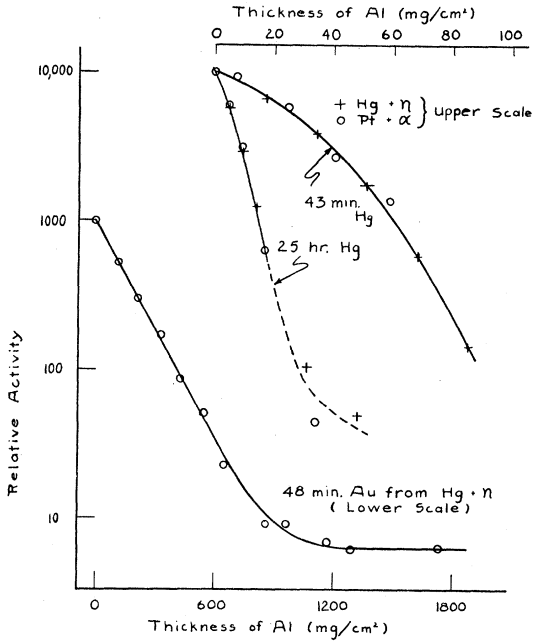

FIG. 6. Absorption in aluminum of 48-min. Au, 43-min. Hg and 25-hour Hg.

It is hoped that a further study of the cross sections for neutrons of various energies will help to clarify the identity of the 43-min. Hg and 80-min. Pt activities.

It is a pleasure to acknowledge the ready cooperation of Drs. B. R. Curtis, E. P. Clancy, and L. Fussell, Jr., and Mr. R. S. Bender and other members of the cyclotron laboratory.