Radioactivities Produced by Proton Bombardment of Palladium

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Proton bombardment of Pd produces the three known Ag periods of 2.3 min. (Ag^{108}), 25 min. and 8 days (Ag^{106}), and three new periods of 16.3 min. (+), 73 min. (+) and 45 days (*K* capture). The latter are assigned tentatively to Ag^{102}, Ag^{104}, and Ag^{105}, respectively. Excitation functions for the isomeric 25-min. and 8-day periods give *p-n* thresholds of 3.8 and 3.9 Mev, respectively. The former agrees with the known positron spectrum of the short period. The higher threshold for the 8-day period indicates that this state lies above the 25-min. state. This activity is accompanied by probably three gamma-rays of which two of 0.62 and 1.06 Mev have been identified with a beta-ray spectrograph. Since no positrons accompany this activity it must decay mainly by *K*-electron capture to Pd^{106} rather than to the lower isomeric state. Four gamma-rays of about 0.29, 0.42, 0.50, and 0.69 Mev accompany the 45-day period.

BOMBARDMENT of Pd by fast protons produces six radioactive periods. Decay curves from which these are obtained are shown in Figs. 1 and 2. The half-lives are 2.3 min., 16.3 min., 25 min., 73 min., 8 days, and 45 days. All of them have been assigned to Ag isotopes. Fig. 3 shows the radioactive Ag isotopes (circles) and the Rh, Pd, Ag, and Cd stable isotopes (squares).

The 2.3-min., 25-min., and 8-day periods have been reported by previous observers[1] and assignments fixed by several reactions. The 2.3-min. activity (e^-) is due to Ag^{108} while the 25-min. and 8-day periods have been assigned to isomers of Ag^{106}. The 25-min. state emits positrons whose maximum energy is 2.0 Mev.[1,2] The 8-day activity has been reported[1,2,3] as emitting electrons, a much smaller number of positrons, and gamma-rays. A further study of its radiations will be reported in this paper.

It should be possible to produce the 22-sec. period assigned to Ag^{110} by proton bombardment of Pd^{110} (Fig. 3). No trace of this period was found even when observations began within 2 min. after bombardment. It is easily observed after slow neutron bombardment of Ag. If the cross section of this activity at 6.5 Mev were of the order of magnitude of other short period cross sections, the period would have been observed. The *p-n* threshold, therefore, must be quite high.

THE 16.3-MIN. AND 73-MIN. PERIODS

These periods have been assigned to Ag^{104} and Ag^{102}, respectively, since they are probably formed by *p-n* reactions. The assignments were made in this way since the abundance of these isotopes gave yield values of the order of magnitude of those for the 25-min. period. These periods have not been produced by other reactions. Ag^{102} cannot be produced by any other known type of reaction but Ag^{104} could be formed by Cd^{106} (d, α).

The activities probably consist of positrons though no accurate sign determinations could be made because of the other short periods of both signs.

The 16.3-min. activity is too strong to be assigned to an impurity. There is however some doubt concerning the assignment of the 73-min. period. This activity appeared in the Pd foils obtained from the American Platinum Works. The purity was given as 99.8 percent and the impurities listed as Pt, Rh, and Ir. None of these elements show *p-n* reactions at an energy of 6.6 Mev. The period also appeared in Pd obtained from another source. If the period is due to impurities, it must be composed of several periods from low atomic number elements occurring as 0.1 percent or less in all Pd targets.

[1] M. L. Pool, Phys. Rev. **53**, 116 (1938).
[2] N. Feather and J. V. Dunworth, Proc. Roy. Soc., **168**, 566 (1938).
[3] J. R. Richardson, Phys. Rev. **55**, 609 (1939).

The 8-day and 45-day Periods

Analysis of the long period activities of the Ag produced from Pd gives an 8-day and a 45-day activity. The 45-day period was determined from the curves of Fig. 2 and from other measurements extending over three months.

Considering the possible products of p-n reactions, Ag^{105} was the unassigned isotope of odd mass number closest to the stable Ag isotopes (Fig. 3). Hence the longest of the periods was assigned to it. It should be possible to produce this period from Pd^{104} by a d-n reaction. The activity would be small and may have escaped observation.

Both long period activities are composed of x-rays, γ-rays, and conversion electrons. These were separated and identified from absorption measurements. The electrons were stopped by covering the target with 2 mm. of Be. From absorption measurements in aluminium the x-ray and γ-ray components were separated and are plotted in Fig. 2.

The x-ray absorption coefficient of the 45-day activity is equal to that expected for Pd $K\alpha$. This isotope probably decays by K-electron capture followed by γ-ray emission in a manner similar to that assigned to the 8-day period in Fig. 6.

The 8-day period x-ray absorption curves also have the Pd $K\alpha_1$ slope. The activity was too small however to make definite differentiation from x-rays of neighboring elements.

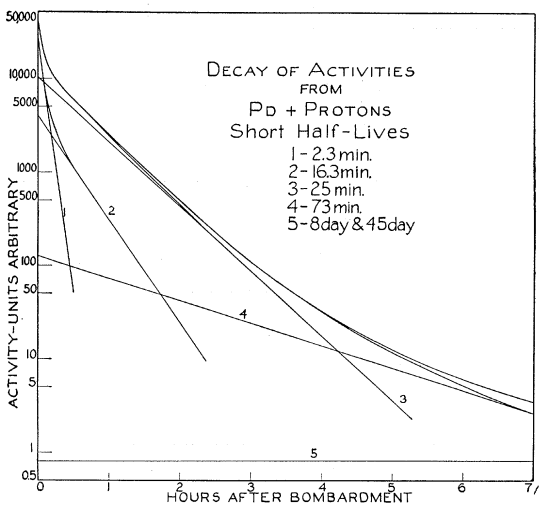

Fig. 1. Short period decay curve for activated Pd.

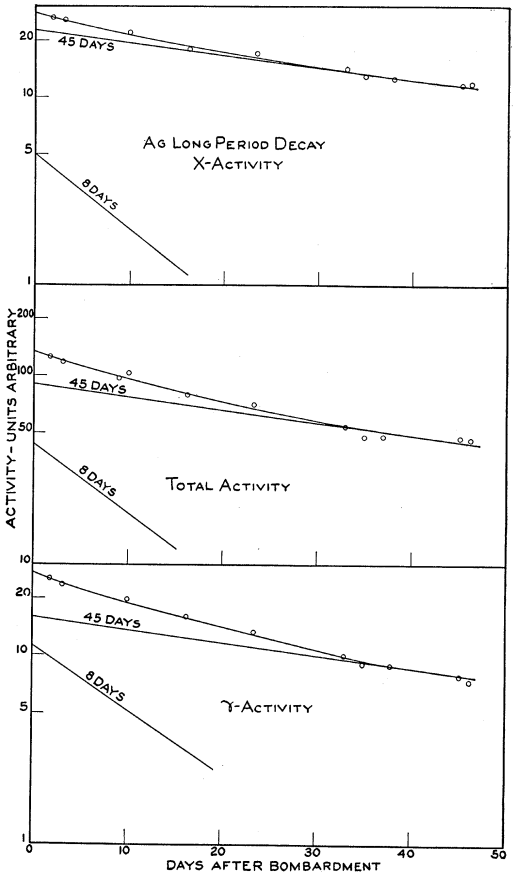

Fig. 2. Long period decay curve for activated Pd.

From the curves of Fig. 2 and the sensitivity ratio, the ratio of electrons to x-ray quanta was found to be 1 : 5 for the 8-day period and 1 : 10 for the 45-day period. The low energy electrons recently reported by Richardson[3] for the 8-day period are undoubtedly secondaries produced by the x-rays.

Gamma-ray spectra of the two long periods were obtained by covering the sample with lead foil and mounting in a β-ray spectrograph to obtain spectra of the photoelectrons. One 6-day

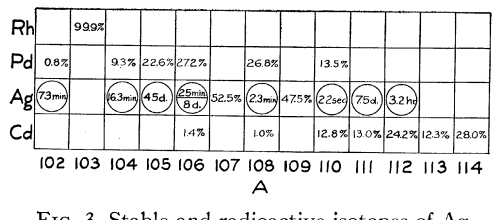

Fig. 3. Stable and radioactive isotopes of Ag and neighboring elements.

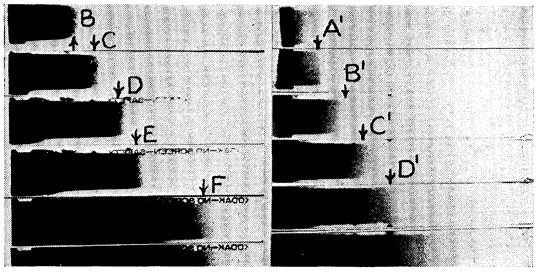

FIG. 4. Long period γ-ray spectra.

exposure was begun 3 days after bombardment and a second 7-day exposure began 38 days after bombardment. The sample was the same one used to get the decay curves of Fig. 2. Exposure 1 should show spectra of both periods, but exposure 2 should show only the 45-day period. Lack of contrast in the films made high contrast prints necessary for their analysis. Prints were made on Kodalith film and developed in a special high contrast developer prepared by Mr. T. W. Finucane of the Institute of Optics. The edges in the spectra were brought out by varying the exposures of the Kodalith film. No edge was assigned unless it appeared on several prints of different exposures. Normal contact prints of some of the Kodalith films are shown in Fig. 4. The high energy edges are on the right side of the prints.

Table I gives the $H\rho$ values of the observed edges and the corresponding γ-ray energy values. It is seen that the 45-day spectrum consists of four gamma-rays of 0.29 Mev, 0.42 Mev, 0.51 Mev, and 0.62 Mev. The 8-day spectrum shows two gamma-rays of 0.69 Mev and 1.06 Mev.

EXCITATION DATA

Table II gives the thick target yields and thin target cross sections for the reactions observed. The yields are expressed as radioactive atoms formed per proton for a thick target of the pure isotope with incident protons of 6.3 Mev energy. The cross sections were calculated from thin film excitation measurements using stacked foils with incident protons of 6.63 Mev.

The 8-day period and 45-day period cross sections are calculated for K-electron capture, the number of radioactive atoms decaying being taken equal to the number of x-ray quanta. The fraction of the initial activity of 8-day and 45-day periods caused by x-rays was obtained from the data of Fig. 2, and the same ratios assumed for the thin targets. The ionization chamber was calibrated for x-rays by observing the deflection caused by a known number of In x-ray quanta from radioactive[4] Sn^{113}. The ionization produced by one x-ray quantum was found to be 1/25 of that for one β-ray.

The excitation curves of Fig. 5 were obtained from observations on stacked Pd foils 0.2 mil thick. The energy intervals were increased in some cases by placing Al foils between the targets. To make possible a closer examination of the long period thresholds, a bombardment was made on 0.1-mil foils without intermediate Al foils.

Energy determinations were made by measuring the visible range in air of the proton beam. This value was corrected for temperature and pressure, and assumed to be the extrapolated range. The mean range was calculated and the energy read from the curves given by M. S. Livingston and H. A. Bethe.[5] The energy values used were thus maxima rather than average

TABLE I. *Gamma-rays from 8-day and 45-day periods of Ag.*

EXPOSURE	LINE NO.	$H\rho$ IN GAUSS-CM	$h\nu$ IN MEV
1	A	1650	0.29±0.03
	B	2260	0.43± .04
	C	2620	0.52± .04
	D	2980	0.62± .04
	E	3270	0.69± .05
	F	4560	1.06± .05
2	A'	1610	0.29± .03
	B'	2140	0.40± .04
	C'	2470	0.49± .04
	D'	2990	0.62± .04

TABLE II. *Yields for p-n reactions yielding silver isotopes.*

ISOTOPE AND EMITTED PARTICLE	HALF-LIFE	YIELD RAD. ATOMS PER 10^6 PROTONS	σ IN CM^2 $\times 10^{26}$
$Ag^{108}(e^-)$	2.3±0.2 min.	4.5±0.5	8.1 ±0.8
$Ag^{106}(e^+)$	25.0±0.5 min.	2.7± .3	5.5 ±0.6
$Ag^{106}(K)$	8.2±0.2 days		0.13±0.05
$Ag^{105}(K)$	45 ±5 days		3.7 ±1.0
$Ag^{104}(e^+)$	16.3±0.7 min.	2.6±1.0	1.8 ±0.9
$Ag^{102}(e^+)$	73 ±10 min.	2.1±1.0	

[4] S. W. Barnes, Phys. Rev. **56**, 414 (1939).
[5] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 245 (1937).

values. This was desirable as threshold determinations were the main object of the measurements.

The 25-min. threshold was established as 3.8 ± 0.1 Mev. The 8-day period and 45-day period thresholds are probably within 0.4 Mev of each other. A composite excitation curve taken within 4 days of the end of the bombardment time has been plotted. This shows a 3.9-Mev threshold. At 3.8 Mev the foils showed no activity. At 4.3 Mev the activity had a period of about 20 days. The 8-day period threshold was therefore between these limits.

The Isomers of Ag^{106}

Energy level diagrams for the decay of the 25-min. and 8-day Ag^{106} isomers to Pd^{106} are shown in Fig. 6. The Pd^{106} energy is taken as the ground level. The Ag^{106} energies are obtained by subtracting the n-p mass difference ($\sim$0.8 Mev) from the observed p-n thresholds.

The 25-min. period belonging to the lower state must decay directly to the ground state of Pd^{106} by positron emission, the threshold measurements being in agreement with the known positron spectrum. Since the 8-day period is accompanied by few if any positrons, and since the gamma-rays accompanying it are

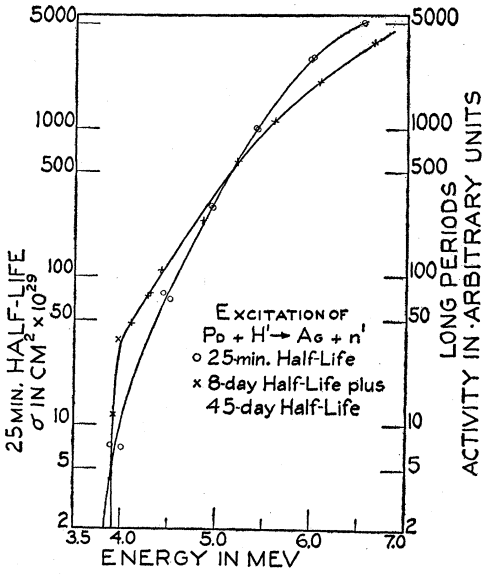

Fig. 5. Excitation curves for protons on Pd.

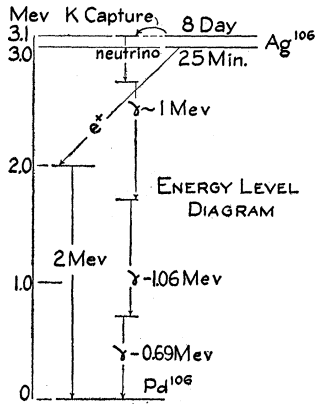

Fig. 6. Energy level diagram for Ag^{106} and Pd^{106}.

of much higher energy than the 0.1 to 0.2 Mev threshold difference, it is necessary to assume that this state decays by K-electron capture and gamma-ray emission directly to Pd^{106} rather than to the lower 25-min. state. Since a total of 3 Mev must be assigned to neurino and gamma-radiation and since two gamma-ray lines of 0.69 and 1.06 Mev have been identified it is evident that at least three gamma-ray quanta must be emitted in cascade in the process. Coincidence counter measurements by Feather and Dunworth[2] have previously indicated 3 to 5 quanta emitted simultaneously. It is probable that there are two quanta, unresolved on our plates, of about 1 Mev as shown in Fig. 6. Measurements of the relative ionization produced by the x-rays and gamma-rays also indicate about one x-ray quantum to three or four gamma-rays.

Since the electrons which accompany this period can be assumed to be conversion electrons, and since no continuous negative β-ray spectrum has been established,[3] it is unnecessary to postulate that electron decay to Cd^{106} occurs to any appreciable extent.

Although our cloud-chamber observations showed no positrons accompanying this period a small number of positrons was reported by Pool. These may be due to occasional direct positron emission from this state or to decay from the 8-day to the 25-min. state followed by positron emission.

The p-n cross section for the 25-min. period is about 30 to 40 times as great as for the 8-day

period (Table II). Pool[1] reports that for the n-$2n$ reaction the 8-day period cross section is greater than the 25-min. period cross section.

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Disintegration of Beryllium by Electrons

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The beryllium nucleus has been disintegrated by fast electrons whose energies exceeded the photoelectric threshold. The source of the electrons was a 1.8-Mv Van de Graaff generator. As was predicted by Guth, neutrons were produced, and the radioactivity which these neutrons induced in silver was used as a measure of the rate of disintegration. The possibility that the observed disintegration was not due to the direct action of the electrons, but resulted from stray x-rays, or x-rays produced in the beryllium by the electrons was eliminated. The yield curve for disintegration by electrons was obtained, and fixed the threshold for this process at 1.63 ± 0.05 Mev. At 1.73 Mev the cross section was found to be 10^{-31} cm^2, in good agreement with theory.

The yield curve for photodisintegration by continuous x-radiation was also obtained, and the threshold was found to be identical with that for disintegration by electrons.

SEVERAL attempts have been made in the past to disintegrate atomic nuclei with electrons.[1] Cathode rays and positrons with energies up to 800 kev have been used to irradiate a wide variety of elements, but no evidence of nuclear disintegration has been reported. Recently, however, one of us[2] has presented a theoretical treatment of this problem and has indicated clearly the conditions under which disintegration by electrons may be expected to take place. His theory points out a similarity between this process and that of photodisintegration and states that before disintegration by electrons may occur the energy of the electrons must exceed the photoelectric threshold of the nucleus.

It was to be expected then, that disintegration by electrons would be most likely to succeed with the element beryllium, since it has the lowest known photoelectric threshold. The threshold has been estimated[3] to be about 1.6 Mev which fortunately is somewhat lower than the maximum potential of our Van de Graaff generator. The reaction predicted is the following

$$Be^9+e_v\rightarrow Be^8+n+e_{v'},$$

where v and v' indicate energies of electrons before and after disintegration. If Be^8 is unstable, on which point there is some disagreement,[4] it would soon disintegrate into two low energy helium nuclei. The cross section of this process for electrons having energies a few 100 kilovolts above the threshold is estimated[2] to be about 10^{-31} cm^2. It was decided to allow the neutrons produced by the above process to induce radioactivity in another element and to use this induced radioactivity as a measure of the rate of disintegration by electrons. Considera-

[1] J. J. Livingood and A. H. Snell, Phys. Rev. **48**, 851 (1935); W. B. Lewis and W. E. Burcham, Camb. Phil. Soc. Proc. **32**, 503 (1936); G. P. Thomson and J. A. Saxton, Phil. Mag. **23**, 241 (1937).

[2] E. Guth, Phys. Rev. **55**, 412 (1939).

[3] J. Chadwick and M. Goldhaber, Proc. Roy. Soc. **151**, 479 (1935).

[4] Allison, Skaggs, Smith, Phys. Rev. **56**, 288 (1939).

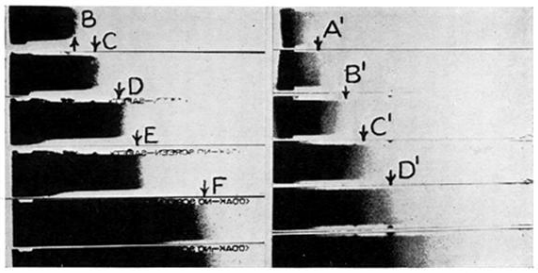

FIG. 4. Long period γ-ray spectra.