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Radioactive Isotopes of Palladium and Silver from Palladium

J. D. KRAUS AND J. M. CORK
University of Michigan, Ann Arbor, Michigan
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The bombardment of palladium with 6.3 Mev deuterons yields radioactive periods of 17 min. and 13 hr. (both negative) in palladium and periods of 26 min. (positive) and 180 hours (negative) in silver. The palladium activities are due to neutron capture and are probably attributable to Pd^{111} and Pd^{109}, respectively. Fast neutron bombardment of silver shows that there are undoubtedly two distinct long-period negative activities of about 180 hours half-life in the silver due to Ag^{106} and Ag^{111}. The positive 26 min. silver activity is also associated with Ag^{106}, being an example of the existence of isomeric isotopes. The active Ag^{111} can be formed from Pd^{110} directly by proton capture and indirectly by a chain reaction through Pd^{111}. The two 180-hour silver activities differ in that there is a strong gamma-radiation accompanying the beta-emission from Ag^{106} and little or no gamma-radiation from Ag^{111}. The upper limits of energy for the beta-spectra of the 26-min. (Ag^{105}), 13-hr. (Pd^{109}), and 180-hr. (Ag^{111}) activities are 2.24 Mev, 1.08 Mev, and 0.80 Mev, respectively. The excitation functions for the 13-hr. (Pd^{109}) and 180-hr. (Ag^{111}) activities are determined by bombarding a stack of 25 palladium foils. These results are compared, since the processes involved correspond to the escape of a neutron or a proton from similar excited nuclei.

INTRODUCTION

IN a preliminary report[1] it was noted that palladium bombarded by deuterons (6.3 Mev) yields radioactive isotopes of both palladium and silver. This study has been continued to determine which particular isotope is responsible for each radioactive period and to evaluate the upper limits of the beta-spectra. In addition, the excitation functions for certain of the processes are shown, indicating how certain of the activities vary with the energy of the bombarding deuterons.

The radioactive isotopes in palladium are produced with deuteron bombardment by the capture of a neutron and the ejection or rejection of the proton. The radioactive silver isotopes may be produced either by the capture of a proton with the ejection of a neutron or by a secondary process from a radioactive palladium isotope. There is evidence that both processes are present.

Fermi[2] and his collaborators have observed in palladium, as a result of slow neutron bombardment, radioactive periods whose half-lives are 15 min., 12 hr., and 50 hr. These same periods should be expected also with deuteron bombardment. Actually only the first two, slightly modified (17 min. and 13 hr.), are found in the palladium. The longer (50-hr.) period is due to a radioactive silver isotope built up from the palladium in a chain reaction. Although it was intended to study only the palladium in this work, it became necessary to include silver to understand the processes involved.

[1] J. D. Kraus and J. M. Cork, Phys. Rev. **51**, 383 (1937).

[2] E. Amaldi, D'Agostino, E. Fermi, B. Pontecorvo, F. Rassetti, and E. Segrè, Proc. Roy. Soc. **149**, 522 (1935).

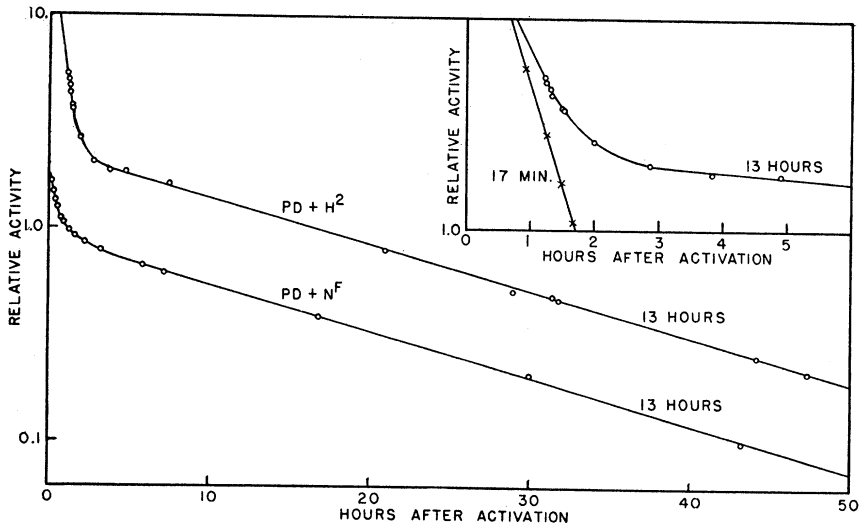

FIG. 1. Decay curves for palladium precipitate following deuteron bombardment (upper) and for palladium following fast neutron bombardment (lower). The inset shows the resolution of the decay curve into the 17-min. and 13-hr. half-life palladium periods.

APPARATUS

In this investigation targets of palladium and silver were bombarded in several ways. Deuterons of energy about 6.3 Mev were produced and allowed to fall directly on the targets in the cyclotron. For slow neutron exposures the palladium or silver specimen was imbedded in a mass of paraffin and placed close to a beryllium target, which in turn was bombarded by the deuteron beam. For fast neutron exposures, the 6.3 Mev deuterons were allowed to fall on a lithium target. The palladium specimen was wrapped so as to exclude any scattered deuterons and was placed close to the lithium.

The intensities of the radioactivities produced were measured with Lauritsen quartz fiber electroscopes and a Wulf string electrometer equipped with ionization chamber. A Wilson cloud chamber was used to determine the sign and energy of the beta-particles.

TABLE I. *Stable and radioactive isotopes of palladium and neighboring elements. Radioactive isotopes with their half-life periods are indicated by circles.*

AT. NO.	AT. WT.	102	103	104	105	106	107	108	109	110	111	112	113
45	RH		100%	(4.2 MIN.)									
46	PD	.8%		9.3%	22.6%	27.2%		26.8%	(13 HR.)	13.5%	(17 MIN.)		
47	AG					(26 MIN.) (180 HR.)	52.2%	(2.3 MIN.)	47.5%	(22 SEC.)	(180 HR.)		
48	CD					1.5%		1.0%		15.2%	15.2%	21.8%	14.9%

PROCEDURE AND RESULTS

After bombardment, the palladium targets were dissolved and a small quantity of solutions of the neighboring elements, silver, rhodium, and ruthenium, were added. The precipitates were taken down separately and examined. Only the palladium and silver showed radioactivity.

The silver was precipitated from the nitric acid solution as the chloride and then redissolved in ammonium hydroxide and reprecipitated. The palladium was next precipitated by the addition of dimethylglyoxime to the slightly acid solution.

From Table I it appears that radioactive isotopes of palladium corresponding to the addition of a neutron might be expected for the masses 107, 109, and 111. These should be produced by bombardment with deuterons or slow neutrons and in part by very fast neutrons.

The decay curve for the palladium precipitate for deuteron bombardment is shown in Fig. 1 (upper curve and inset). Periods of 13 hr. and 17 min. half-life are apparent. Both are negative active. Similar curves are obtained for slow neutron bombardment. For fast neutron bombardment (lower curve of Fig. 1) the decay curve is similar except the ratio of the initial activities of the 17-min. to the 13-hr. periods is less than it is for slow neutron or deuteron bombardment. Although the fast neutron bom-

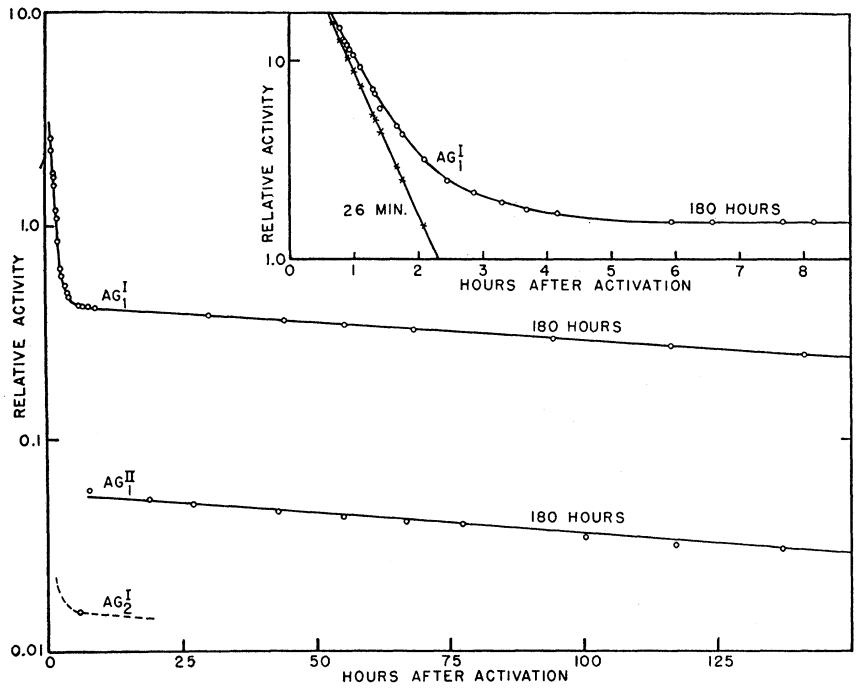

FIG. 2. Decay curves for successive silver precipitates from palladium bombarded by deuterons, showing the production of active silver (Ag^{111}) from active palladium (Pd^{111}). The inset shows the resolution of the silver activity into the 26-min. and 180-hr. half-life periods.

bardment giving the activity shown in Fig. 1 (lower curve) was about twice as long as that with deuterons, giving the activity of the upper curve, the ratio of the 17-min. to the 13-hr. activities is sufficiently smaller in the case of the fast neutron bombardment to indicate a difference in the reactions. The possibility of an additional shorter palladium period is not excluded as several minutes are required for making the chemical separation.

If one of the observed periods is due to the isotope of mass 111 then by beta-decay it should produce a radioactive silver since there is no stable silver of mass 111. Subsequent precipitation of silver from the palladium precipitates proved this to be the case. Moreover, by making these subsequent separations at different times after the original bombardment, it appears to be quite certain that this silver activity (180-hr. half-life) must be built up from the 17-min. palladium and not the 13-hr. palladium. If this assignment is correct, it may seem surprising that this 17-min. activity should appear as strongly as it does by fast neutron bombardment. In the fast neutron reaction a neutron should be ejected and the mass decreased by one, and no activity whatever should be associated with Pd^{111}. Actually the Pd^{111} activity is about half as strong when activated by fast neutrons as by slow neutrons. This may be due to a part of the fast neutron beam being slowed by elastic collisions or to energetic relations existing in the palladium nucleus such that the process of neutron ejection is relatively difficult.

The 13-hr. palladium activity could be due to either isotopes 107 or 109. The assignment to isotope 109 is preferred when one considers the ratio of activities for fast and slow neutrons and the relative abundance of the parent isotopes.

Figure 2 shows the activity of the silver precipitate from palladium bombarded by deuterons. There are two periods evident, one of 26-min. half-life and the other of about 180-hr. The 180-hr. silver activity has been followed for over 500 hours. The 26-min. silver emits positrons, while the 180-hr. silver is negative active. Radioactive silver isotopes of mass numbers 103, 105, 106, and 111 could be made from the stable

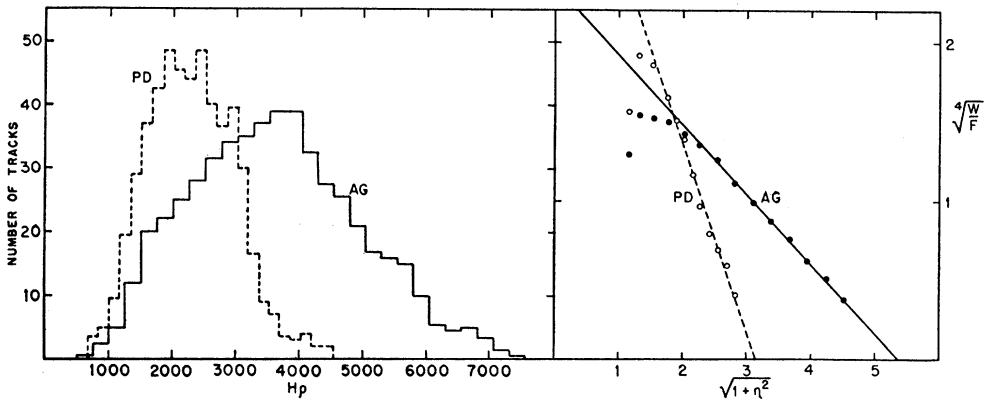

FIG. 3. Beta-ray histograms for the negative 13-hr. Pd^{109} and the positive 26-min. Ag^{106} deuteron induced activities. $H\rho$ in gauss-cm. At the right are the corresponding Kurie plots made according to the Konopinski-Uhlenbeck theory.

palladium isotopes by proton capture. Isotopes 103 and 105 should be radio-positive, 106 either positive or negative, and 111 should be radio-negative. To aid in the correct assignment of the observed activities, silver was bombarded with fast and slow neutrons. The slow neutron bombardment yields the well-known Fermi periods of 22 sec. and 2.3 min. The fast neutron[3] bombardment yields periods of 2.3 min., 26 min., and another period of approximately 180 hr. This fixes the 2.3-min. period as due to isotope 108 and the 26-min. positive activity with isotope 106.

The long negative activity in the silver of half-life 180 hours offers an interesting puzzle. Since it is made by fast neutrons it might be due to either isotopes 106 or 108. Slow neutron bombardment of silver has so far failed to produce this activity, hence it is ascribed to isotope 106. On the other hand, the subsequent separations of silver from the palladium precipitate makes it appear to be due to isotope 111, which could also be formed from palladium by direct proton capture. The activity of this later silver separation from the palladium precipitate is shown in Fig. 2 and is labeled Ag II/1. The first silver precipitate made immediately after bombardment, is called Ag I/1 and this is followed immediately by two additional separations called Ag I/2 and Ag I/3, to make sure that the active silver is completely removed. The separation Ag I/3 was too weak to be indicated in Fig. 2. The separation Ag II/1, made several hours after bombardment, shows many times the activity found in the earlier identical separation Ag I/3, and several times the activity of Ag I/2. A second precipitate, called Ag II/2, made immediately after Ag II/1, shows almost no activity. The half-life of the precipitate Ag II/1 is about 180 hours. It thus appears that there must be two radioactive silver isotopes of approximately the same half-life (180 hr.), both negative active, one being an isomer of mass 106 and the other an isotope of mass 111.

To establish this more definitely, a calculation was made of the ratio of initial activities (immediately after bombardment) for the 26-min. and the 180-hr. periods when fast neutron bombardment of the silver was used and when deuterons were used. For a two and one-half hour exposure, with fast neutrons, the ratio of the initial activities of the 26-min. to the 180-hr. was 990 to 1. When corrected for the periods and length of exposure, the value of the branching ratio for the 26-min. to 180-hr. activities in isotope 106 is about 10 to 1. For the case of proton capture (i.e., the silver precipitate from bombarded palladium), the ratio of the 26-min. to the 180-hr. initial activities for a corresponding exposure is only 60 to 1. It thus appears that the final activity in the silver precipitate from palladium is in large part due to isotope 111 and only slightly due to isotope 106.

That these two periods are quite distinct may

[3] Pool, Cork, and Thornton, Phys. Rev. **52**, 239 (1937).

be shown in still another manner. The activity due to the fast neutron bombardment of silver (isotope 106) is only slightly reduced by placing a one-eighth inch thick aluminum plate between the specimen and the ionization chamber, indicating a very strong gamma-radiation. The activity of the silver precipitate (isotope 111) from palladium bombarded by deuterons was reduced almost to zero when the same aluminum plate was interposed, indicating little or no accompanying gamma-radiation.

The probable reactions are as follows:

$$_{46}Pd^{N}+{}_{1}H^{2}\rightarrow{}_{46}Pd^{N+1}+{}_{1}H^{1}$$

$$_{46}Pd^{N+1}\rightarrow{}_{47}Ag^{N+1}+{}_{-1}e \quad \text{(17 min., 13 hr.)}$$

$$_{46}Pd^{N}+{}_{1}H^{2}\rightarrow{}_{47}Ag^{N+1}+{}_{0}n^{1}$$

$$_{47}Ag^{N+1}\rightarrow{}_{48}Cd^{N+1}+{}_{-1}e \quad \text{(180 hr.)}$$

$$_{47}Ag^{106}\rightarrow{}_{46}Pd^{106}+{}_{+1}e. \quad \text{(26 min.)}$$

Beta-Radiations

Since it was readily possible to isolate certain of the activities, an attempt was made to determine the form and the upper limits of energy for the beta-spectra. For this work tracks were photographed in a hydrogen-filled Wilson cloud chamber using a deflecting magnetic field of about 400 gauss. Only those tracks satisfying arbitrary criteria as to length of visible path and freedom from collisions were measured. The results of these measurements for the positive active 26 min. silver period and for the negative active 13-hr. palladium period are shown in Fig. 3. Over 500 tracks were measured in each case. These data are shown both as histograms, on the left, and as Kurie[4] plots based on the Konopinski-Uhlenbeck[5] theory, on the right. For the higher energies the Kurie plot is linear but for the lower energies the number of tracks is evidently deficient, due probably to the selection criteria. The Kurie plot intercepts indicate an upper energy of 2.24 Mev for the 26-min. Ag^{106} and 1.08 Mev for the 13-hr. Pd^{109}. The upper limits actually observed for the two cases were 1.90 and 1.03 Mev, respectively. Only about 125 tracks were measured for the 180-hr. Ag^{111} activity. The maximum observed energy for this radiation was 0.80 Mev.

Excitation Function

In the bombardment of palladium with deuterons two different processes are involved, namely, proton capture and neutron capture, and it is of interest to compare the activation in each case as a function of the bombarding energy. The energy of the deuteron beam in the cyclotron is not perfectly homogeneous, but in this case it seems that any lack of homogeneity would be relatively unimportant since both activities are observed in the same specimens. Accordingly, a stack of 25 foils, each 0.00127 mm thick was exposed to the deuteron beam for 5 hours.

Corrections for the absorption in the platinum foil of the window of the cyclotron and a cover foil over the stack bring the average energy for the top foil to 5.8 Mev. The average energy at the center of each succeeding foil was determined by using a modified value of the stopping power as calculated by Mano.[6] After the bombardment

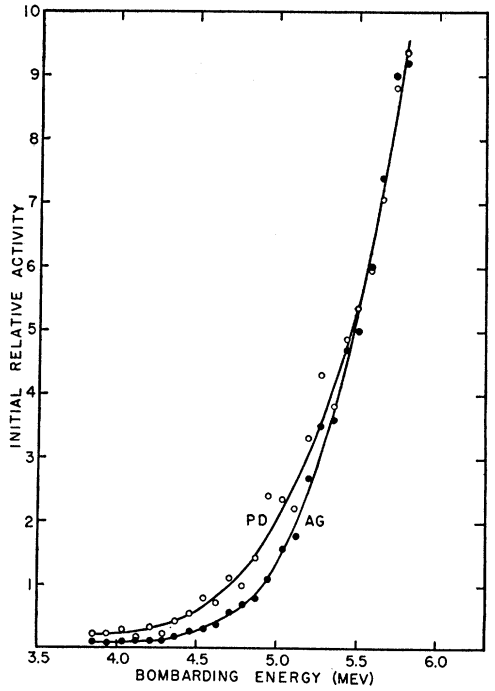

FIG. 4. Excitation curves for the 13-hr. Pd^{109} and the 180-hr. Ag^{111} activities.

[4] F. N. D. Kurie, J. R. Richardson, and H. C. Paxton, Phys. Rev. **49**, 368 (1936).

[5] E. Konopinski and G. Uhlenbeck, Phys. Rev. **48**, 7 (1935).

[6] J. Mano, J. de phys. et rad. **5**, 628 (1934).

the activity of each foil was followed for over a week and a decay curve plotted for each foil. As explained earlier, the end activity in each foil was quite probably due mainly to the 180-hr. Ag^{111} isotope. The initial activity of the 180-hr. period in each foil was plotted as a function of the bombarding energy. These data for the 180-hr. silver activity are shown by the solid circles in Fig. 4. The curve through these points corresponds to a reaction involving proton capture (i.e., deuteron in, neutron ejected).

By subtracting the 180-hr. activity from the decay curve for each foil, a new family of curves is obtained, each ending in a straight line corresponding to the 13-hr. palladium activity. By extending these curves back to the time at which the bombardment was ended, the relative values of the initial 13-hr. Pd^{109} activities for each foil are obtained. These are shown as open circles in Fig. 4 and the curve through them corresponds to a reaction involving neutron capture (i.e., deuteron in, proton ejected).

While following the activity of the foils, the electrometer was continually calibrated in terms of a known uranium standard. The initial activity of the top foil for the 13-hr. palladium was about 1.70 microcuries and for the 180-hr. silver about 0.092 microcuries.

To facilitate comparison of the two excitation curves, they are arbitrarily adjusted to the same value at an energy of 5.8 Mev. Although too much significance should not be attached to these curves, it seems reasonably certain that as the energy of the incident particle decreases, the activity in the silver falls off somewhat more rapidly than in the palladium. This is contrary to what might be expected from the Bohr model of the nucleus, since it indicates less probability for neutron ejection from the excited nucleus than for proton ejection. It would, however, be more in accord with the Oppenheimer-Phillips[7] consideration, which is concerned with the probabilities of the particles entering the nuclei.

We are greatly indebted to B. R. Curtis for work with the cloud chamber and to D. W. Stewart and E. Rosenbaum for making the many chemical separations.

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[7] Oppenheimer and Phillips, Phys. Rev. **48**, 500 (1935).

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Neutron-Induced Radioactivity of Long Life in Cobalt

J. R. RISSER
*Palmer Physical Laboratory, Princeton University, Princeton, N. J.**

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When cobalt is bombarded by neutrons, an activity of long half-life is obtained. The radioactive element has been shown by a chemical analysis to be an isotope of cobalt. The radiations emitted in the disintegration of this element have been investigated, principally by the absorption method. A gamma-ray is observed with a mass absorption coefficient in Pb of about 0.047 cm^2/g, and a spectrum of soft beta-rays with an estimated limiting range of 30 mg/cm^2 of aluminum, representing an energy of 160 kv. A group of more penetrating particles completely absorbed only by about 0.65 g/cm^2 of aluminum is also indicated. This group may represent a relatively infrequent disintegration process, in which the total energy is carried off by the beta-particle and no gamma-ray is emitted. Results of a measurement on the half-life indicate that it is 2.0 ± 0.5 years.

AN activity of half-life over a year was discovered in cobalt bombarded with neutrons by Sampson, Ridenour and Bleakney.[1] Results of activation of only a few weeks with the neutrons of 55 mC of Ra+Be slowed down by water indicated that a very strong activity would be obtained if bombardment were continued for a time comparable to the half-life.

* Now at Purdue University, Lafayette, Indiana.

[1] Sampson, Ridenour and Bleakney, Phys. Rev. **50**, 382 (1936).