# Half-Life of $Pd^{109}$ and Neutron Activation Cross Section of $Pd^{102}$

W. WAYNE MEINKE
*Department of Chemistry, University of Michigan, Ann Arbor, Michigan*
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The half-life of $Pd^{109}$ has been determined to be 13.6±0.1 hours by careful decay measurement on a chemically purified fraction of metallic palladium irradiated in a nuclear reactor. From the same irradiation the activation cross section of $Pd^{102}$ was found to be 4.8 barns for pile neutrons. This cross section value is based on the literature value (corrected for the new half-life determination) of 11.7 barns reported for the thermal neutron activation cross section of $Pd^{108}$.

## I. INTRODUCTION

METALLIC palladium is in many ways an ideal beta-ray source.[1] $Pd^{109}$ is formed with high cross section in the nuclear reactor; it decays with a half-life of slightly more than half a day, thereby reducing the problems of contamination and decontamination; and its metabolic inertness reduces the general personnel hazard of its use. Furthermore, the palladium serves as a pure beta-ray source except for some low energy gamma-rays associated with the decay of the isotopes of palladium formed in neutron irradiation.

Despite this use of palladium as a beta-ray source and its availability from various reactor groups, a survey of the literature reveals discrepancies in nuclear characteristics of some of the radioactive isotopes obtained in a neutron irradiation of pure palladium metal. Since this laboratory has had access to the multi-curie $Pd^{109}$ sources obtained by the Fission Products Laboratory of the University of Michigan Engineering Research Institute,[2] a study has been made of the characteristics of some of the palladium isotopes occurring in these sources.

The stable isotopes of palladium and the isotopes formed in the neutron irradiation of palladium are shown with their decay products in Fig. 1. The abundances of these palladium isotopes were reported by Sampson and Bleakney[3] with an accuracy of one part in one hundred. The half-life values indicated for $Pd^{103}$ and $Ag^{111}$ are considered to be the best literature values. The half-life indicated for $Pd^{109}$ is the value reported in this paper. The thermal neutron activation cross sections for $Pd^{108}$ and $Pd^{110}$ were reported by Seren, Friedlander, and Turkel[4] in their general survey of activation cross sections. The $Pd^{108}$ cross section reported has been changed to incorporate the new half-life value for $Pd^{109}$. Other pertinent data found on the chart were obtained from the Bureau of Standards Compilation of Nuclear Data.[5]

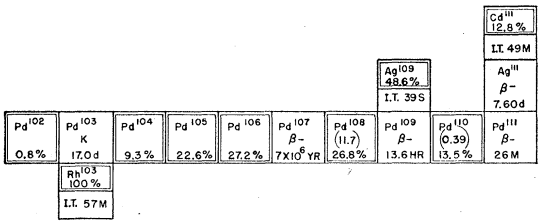


FIG. 1. Isotope chart.

A complete literature search was made for data on the half-lives of $Pd^{103}$, $Pd^{109}$, and $Ag^{111}$. A 17.0-day half-life for $Pd^{103}$ was reported by Mathews and Pool[6] who followed the activity of samples from both proton and deuteron bombardments of rhodium for 10 half-lives. They report that the activity of the isotope was due solely to x-ray emission following orbital-electron capture and they assigned the activity to $Pd^{103}$. Brosi,[7] Gunlock and Pool,[8] and Lindner and Perlman[9] observed the 17-day $Pd^{103}$ but give no further information on the half-life. An evaluation of these values establishes the half-life of $Pd^{103}$ at 17.0 days with a maximum error of 0.1 or 0.2 of a day.

Literature values for the half-life of $Pd^{109}$ vary from 12.7 to 14.1 hours. Early work by Kraus and Cork[10] established a value of 13 hours, but the decay was followed for little more than three half-lives. Seiler[11] made chemical separations of $Pd^{109}$ from fission product mixtures and followed the decay for about eight half-lives. He reports a half-life of 13 hours, but some of his points scatter considerably, and it appears that a somewhat longer half-life line would better represent his data. Perlman and Friedlander,[12] Mock *et al.*,[13] and Wäffler and Hirzel[14] report values of 12.7 hours, 14.1

[1] Meinke, Emmons, and Nehemias, Nucleonics **10**, No. 12, 54 (1952).
[2] L. E. Brownell *et al.*, Atomic Energy Commission Reports COO-90, COO-91, 1952 (unpublished).
[3] M. B. Sampson and W. Bleakney, Phys. Rev. **50**, 732 (1936).
[4] Seren, Friedlander, and Turkel, Phys. Rev. **72**, 888 (1947).
[5] *Nuclear Data*, National Bureau of Standards Circular No. 499 (1950).
[6] D. E. Mathews and M. L. Pool, Phys. Rev. **72**, 163 (1947).
[7] A. R. Brosi, reported in Oak Ridge National Laboratory Classified Reports Mon N-150 (August, 1946); Mon N-229 (January, 1947).
[8] H. F. Gunlock and M. L. Pool, Phys. Rev. **74**, 1264 (1948).
[9] M. Lindner and I. Perlman, Phys. Rev. **73**, 1202 (1948).
[10] J. D. Kraus and J. M. Cork, Phys. Rev. **52**, 763 (1937).
[11] J. A. Seiler, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1950), Paper 119, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[12] M. L. Perlman and G. Friedlander, Phys. Rev. **74**, 442 (1948).
[13] Mock, Waddel, Fagg, and Tobin, Phys. Rev. **74**, 1536 (1948).
[14] H. Wäffler and O. Hirzel, Helv. Phys. Acta **21**, 200 (1948).

±0.3 hours, and 13.1 hours, respectively, for the half-life of $Pd^{109}$ obtained in $(\gamma, n)$ bombardments using a betatron. Recently Bergström *et al.*[15] have made an assignment of the "13-hour" palladium isotope to $Pd^{109}$ with a mass spectrograph. Hummel and Hamermesh[16] used a 13-hour half-life value in calculating the activation cross section of $Pd^{108}$ with 0.03-Mev neutrons. Neither of the latter two papers, however, add further critical information to the value of the half-life.

There is good agreement among the values reported in the literature for the $Ag^{111}$ half-life. In early work Kraus and Cork[17] and Pool[18] reported a value of 7.5 days for this isotope. Steinberg and Glendenin[19] in their fission product work followed the $Ag^{111}$ activity for 12 half-lives and report a half-life of 7.6 days. Duffield and Knight[20] obtained a 7.6-day decay for $Ag^{111}$ produced in betatron bombardments. Recently two other workers investigated the properties of $Ag^{111}$: Storruste[21] reports a 7.5-day decay for a chemically separated silver fraction of a neutron irradiation of palladium; and Johansson[22] reports a 7.5-day or 180±3 hour half-life for $Ag^{111}$ after following a sample for 6 half-lives. An evaluation of these reports establishes the half-life of $Ag^{111}$ at 7.60 days with a maximum error of 0.10 day or less.

$Pd^{107}$ with a half-life of $7\times10^6$ years and $Pd^{111}$ with a half-life of 26 minutes will also be formed in a neutron irradiation of palladium metal, but their half-lives are of such nature that they will have negligible effects a week or so following irradiations of one or two weeks duration.

## II. EXPERIMENTAL METHODS

A small amount of high purity palladium metal which had been irradiated for a period of about two weeks in the high flux position ($5.4\times10^{13}$ $n/cm^2/sec$) of the NRX reactor at Chalk River, Ontario, Canada was partially dissolved in hot nitric acid and the solution subjected to a chemical separation to purify the palladium isotopes for characterization. Since the separation was performed one day after removal of the sample from the reactor, no $Pd^{111}$ remained in the palladium. Spectrographic analysis of this "high purity" palladium metal showed the presence of the following impurities: 0.002 percent Ag, 0.1 percent Au, 0.003 percent Ca, 0.005 percent Cu, 0.02 percent Fe, 0.002 percent Mg, 0.2 percent Pt, 0.2 percent Rh, and 0.01 percent Si.[23]

[15] Bergström, Thulin, Svartholm, and Siegbahn, Ark. Fysik **1**, 281 (1950).
[16] V. Hummel and B. Hamermesh, Phys. Rev. **82**, 67 (1950).
[17] J. D. Kraus and J. M. Cork, Phys. Rev. **52**, 763 (1937).
[18] M. L. Pool, Phys. Rev. **53**, 116 (1938).
[19] E. P. Steinberg and L. E. Glendenin, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1952), Paper 123, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[20] R. B. Duffield and J. D. Knight, Phys. Rev. **75**, 1613 (1949).
[21] A. Storruste, Phys. Rev. **79**, 193 (1950).
[22] S. Johansson, Phys. Rev. **79**, 896 (1950).
[23] Reference 1 gives a list of activities that these contaminants would produce upon activation in the reactor.

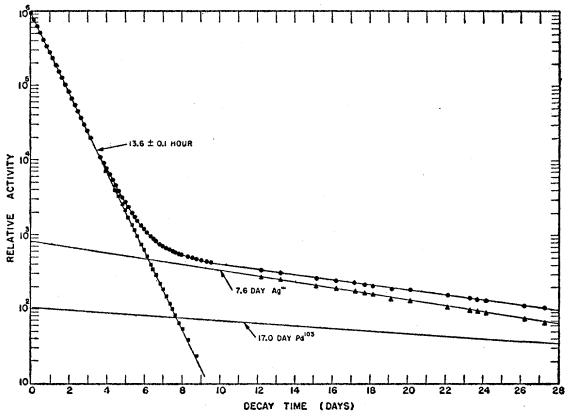


FIG. 2. $Pd^{109}$ half-life.

Silver and palladium carriers were added to the solution of palladium. The silver was then removed by silver chloride precipitations while further decontamination was achieved by precipitating the palladium with dimethylglyoxime in a solution which was 0.5 *N* in HCl. After three complete cycles, portions of the palladium dimethylglyoxime precipitate were placed on a 1-inch diameter aluminum counting plate. The dried samples were finally sealed with Scotch tape.

The decay of the samples was followed on an end-window chlorine-quenched argon-filled Geiger-Muller tube (Amperex 100C) with a window of 3.5-mg/cm² thickness used in conjunction with a scale of 128 counting circuit. The counts were automatically recorded with a Streeter-Amet Scientific Recorder modified to record regularly at any desired interval. Coincidence corrections were applied where the counting rates required them.

The decay of the samples is shown in Fig. 2. Two samples of different activity levels were required to follow the decay over the entire range. The decay of the first sample is shown in the first two cycles of the graph, while the second more active sample was used for the remainder of the data. Counts were recorded automatically for the first ten days, but subsequent data were taken manually.

After most of the $Pd^{109}$ had decayed out, it was discovered that the chemical separation had not been thorough enough to completely decontaminate from the $Ag^{111}$. (Later measurements showed that about 98 percent of the $Ag^{111}$ had been eliminated.) Therefore a third sample was followed for about 45 days through one g/cm² of beryllium absorber to obtain the decay of the x-rays from the orbital electron capture decay of the $Pd^{103}$ activity.

## III. RESULTS AND DISCUSSION

The value for the half-life of $Pd^{109}$ was found to be 13.6±0.1 hours over a period of 15 half-lives as shown in Fig. 2. Sample counts were taken every hour for the first eight days, but only every fourth count is shown on

the graph. After ten days all data are presented on the graph. The decay of the samples was followed for more than 60 days with consistent agreement between the resolved lines and the points.

The activity of the samples shown in Fig. 2 was followed down to background to obtain the 17.0-day component. The decay of the third sample, measured through beryllium absorber to eliminate the $Ag^{111}$ beta-particles, gave a 17.0±0.4 day half-life for $Pd^{103}$. As mentioned above, a literature search showed that the half-lives of the $Pd^{103}$ and $Ag^{111}$ were known accurately. Establishment of a 17.0-day line through the low activity points of $Pd^{103}$ to give exactly a 7.60-day line for the $Ag^{111}$ component gave the best resolution of the curve. This procedure is justified in light of the well-established literature values for the two half-lives and the chemical separation which eliminated contaminants.

The sum of the 7.60-day line and the 17.0-day line was then subtracted from the gross decay curve to obtain the half-life for $Pd^{109}$. Because of the internal consistency of all of the decay data the error presented for the half-life of $Pd^{109}$ is the upper limit for the total error of the experimental determination.

The thermal neutron activation cross section for $Pd^{108}$ has been measured by Seren *et al.*[4] and found to be 11.2 barns with a probable error of 20 percent. A linear change in this cross section value to 11.7 barns has been made in view of the change in the $Pd^{109}$ half-life value from 13.0 to 13.6 days. In the present work the neutron activation cross section for $Pd^{102}$ with pile neutrons (neutrons from the high flux position of the Chalk River NRX reactor) was found to be 4.8 barns by resolution of Fig. 2 using the half-lives of 17.0 days, 7.60 days, and 13.6 hours, and knowing the total time and flux of the irradiation. This value is based on the above thermal neutron cross section of 11.7 barns for $Pd^{108}$.

Several assumptions were made in arriving at this cross-section value. First it was assumed that the contribution to the $Pd^{108}$ and $Pd^{102}$ cross sections by resonance neutron activation is either negligible or equal for the two isotopes. Resonance absorption peaks which may be assigned to $Pd^{108}$ have been reported recently[24] for palladium and may invalidate this assumption to some extent. Since the $Pd^{103}$ decays by orbital electron capture the relative counting efficiencies of the *K* x-rays of rhodium and the beta-particles of $Pd^{109}$ must be evaluated. The ratio of the counting efficiencies of the $Pd^{109}$ beta-particles to the rhodium x-rays was found to be about 87:1 by calibrations involving counting of the $Rh^{103m}$ conversion electron in an internal counter and counting the $Pd^{103}$ x-ray in the Amperex chlorine-quenched tube.[25]

The error of the cross-section value of $Pd^{102}$ is dependent on the ±20 percent probable error of the value of Seren *et al.*[4] The decay data presented in this paper establish the ratio of the $Pd^{108}$:$Pd^{102}$ cross sections within an error of about 5 percent, if the two assumptions listed above are completely valid. Note, however, that any change in the established value of the $Pd^{108}$ activation cross section or any corrections involving resonance activation contributions to either the $Pd^{102}$ or $Pd^{108}$ cross sections or more accurate determinations of the relative counting efficiencies of the $Pd^{109}$ beta-particles and the rhodium x-rays in chlorine-quenched GM tubes would mean a linear change in the cross-section value of $Pd^{102}$.

The establishment of the half-life of $Pd^{109}$ as 13.6 ±0.1 hours may require the re-evaluation of certain yield and cross-section data based on a 13.0-hour half-life. For example the $Pd^{109}$ fission yield reported by Seiler[11] may be changed somewhat when the best 13.6-hour line is drawn through his experimental points. Indeed, as noted previously, even the 11.2-barn activation cross section for $Pd^{108}$ is changed to 11.7 barns when the new half-life value is used.

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[24] D. J. Hughes *et al.*, Atomic Energy Commission Report AECU-2040, 1952 (unpublished).

[25] I am grateful to Mr. A. H. Emmons of the Radiological Safety Department of the University of Michigan for making available his counting data to help in determining these counting efficiencies.