5.373±0.010 Mev for Cm^{246} were obtained by using Po^{210} (5.301 Mev), Po^{214} (7.680 Mev), Po^{218} (5.998 Mev), Em^{222} (5.486 Mev), Cm^{242} (6.110 Mev), and Cm^{244} (5.798 Mev) as standards.

The spontaneous fission half-life of Cm^{248} was determined by fission-counting the curium sample after exhaustive separations from californium. When a correction of 3% was applied to allow for the spontaneous fission of Cm^{246} (assuming its half-life to be 3×10^7 yr),[10] a value of $(4.6\pm0.5)\times10^6$ yr was obtained. The results of this investigation are summarized in Table I.

[10] W. J. Swiatecki, Phys. Rev. **100**, 937 (1955).

PHYSICAL REVIEW VOLUME 103, NUMBER 4 AUGUST 15, 1956

Decay of Neutron-Deficient Isotopes of Pd and Rh†

SEYMOUR KATCOFF AND HENRY ABRASH*
Brookhaven National Laboratory, Upton, New York

(Received May 2, 1956)

Pd isotopes of masses 98, 99, and 101 were prepared by α-particle bombardment of ruthenium. The radiations of these isotopes and of their Rh daughters were investigated with a scintillation coincidence spectrometer. Rh^{98} decays with a half-life of 8.7±0.1 minutes by emission of 2.5±0.2 Mev positrons in coincidence with 650±10 kev γ rays. Its Pd^{98} parent was shown by successive daughter extractions to decay with a half-life of 17.5±0.5 minutes. Rh^{99} (4.7±0.1 hours) showed 0.74-Mev positrons (10%) in coincidence with 335±10 kev γ rays (70%). Other γ rays were observed at 615±15 kev (20%), 890±20 kev (weak line coincident with 335-kev γ), 1.26±0.03 Mev, and 1.41±0.04 Mev (weak). Pd^{99} (21.6±0.6 minutes) was identified by successive Rh daughter extractions. Its maximum positron energy is 2.0±0.1 Mev and the following γ rays were observed: 140, 275, 420, and 670 kev. The 140-kev γ ray is the most intense and the 420-kev peak represents two γ rays of about the same energy in cascade. Rh^{101} (4.7±0.2 days) decays by electron capture followed by emission of 312±10 kev γ rays. Pd^{101} (8.5±0.3 hours) emits 0.58 ±0.04 Mev positrons (4%) by decay to the ground state of Rh^{101}. The γ rays observed were at 288 kev (15%), 590 kev (15%), 720, 1190, and 1280 kev (last three less intense). The photopeak at 288 kev represents two coincident γ rays of about the same energy. Tentative spin and parity assignments were made for the ground states of the nuclides investigated. Mass assignments were confirmed by a study of the relative yields of the various radioactive isotopes as the bombarding α-particle energy and isotopic composition of the Ru target were varied.

INTRODUCTION

IN order to help identify the mass number of a long-lived Tc isotope,[1] assumed to be Tc^{98}, its γ-ray spectrum was compared with that of Rh^{98}. Since both of these nuclides decay to stable Ru^{98}, some of the γ rays might be identical from both. When this work was begun, a 9-min Rh^{98} had been reported[2] but its mass assignment also was uncertain and no information was given about any γ rays. Therefore a study of this isotope was undertaken and extended to include other Rh and Pd isotopes produced in the same bombardments. During the course of these measurements, a new paper appeared[3] in which additional data are presented on Rh^{98}, Rh^{97}, Pd^{98}, and Pd^{99}. Some of these data were confirmed and extended in the present investigation.

EXPERIMENTAL PROCEDURE

Small samples of $RuCl_3$ or of Ru metal powder were irradiated with α particles at a series of energies between 17 and 40 Mev. When necessary, the 40-Mev α particles from the cyclotron were degraded to the desired energy by the proper thickness of Al foil. Irradiation times varied between 5 and 30 minutes; the beam current was usually 0.5–1.0 μa.

Shortly after each irradiation the target was dissolved in diluted HCl,[4] Pd^{+2} carrier was added, and then Pd dimethylglyoxime was precipitated. This was dissolved in hot HNO_3, and then a few successive $Fe(OH)_3$ and AgCl scavenging precipitations were made. The Pd was precipitated again as the dimethylglyoxime. When an active Pd sample was desired for study this purification cycle was repeated. When an active Rh daughter activity was wanted, the second Pd precipitate was allowed to stand for an appropriate growth time. Then it was dissolved, Rh^{+3} carrier was added, and the Pd was removed by a series of precipitations with cupferron. Finally the Rh was precipitated as $K_3Rh(NO_2)_6$. In the last few experiments an ion exchange method was used for separating the Rh from the Pd parent. This will be described below in the section on Pd^{98}.

Decay of gross β rays and of gross γ rays was followed with end window proportional counters and with NaI(Tl) scintillation detectors, respectively. The β- and

† Research carried out under the auspices of the U. S. Atomic Energy Commission.

* Summer visitor (1955), now at the Department of Chemistry, California Institute of Technology, Pasadena, California.

[1] S. Katcoff, Phys. Rev. **99**, 1618 (1955).

[2] A. H. W. Aten Junior and T. De Vries-Hamerling, Physica **19**, 1200 (1953).

[3] A. H. W. Aten Junior and T. De Vries-Hammerling, Physica **21**, 597 (1955).

[4] For the Ru metal targets a preliminary fusion in a mixture of NaOH and NaO_2 was necessary.

γ-ray spectra were investigated with a single-channel pulse-height analyzer and with a coincidence gray-wedge scintillation spectrometer. Among the sources used for γ-ray energy calibration were Te^{123m} (159 kev), Eu^{152} (344 kev), Cs^{137} (662 kev), and Na^{22} (0.511 and 1.28 Mev).

8.7-Min Rh^{98}

In several runs the positron activity was observed to decay through ten half-lives with a half-life of 8.7±0.1 minutes; the γ-ray activity was observed to decay through eight half-lives with the same period (Fig. 1). The end point of the positron spectrum was investigated with the aid of an anthracene scintillator (1×1½ inches) connected to the gray-wedge spectrometer. A value of 2.5±0.2 Mev was obtained for this end point by comparison with the 1.89-Mev positrons of Ga^{68}. This value is considerably lower than the 3.3±0.4 Mev which was obtained by Aten and De Vries-Hamerling[3] from absorption measurements.

These authors reported a γ ray of 650±20 kev and an indication of higher energy γ rays. In the present work, only one γ ray was found, the one at 650±10 kev. Its energy was identical, within the experimental uncertainty, with the energy of one of the Tc^{98} γ rays.[1] The end point of the positron spectrum observed in coincidence with this γ ray was identical with the 2.5-Mev end point of the noncoincident spectrum. Therefore there are no β transitions to the ground state, for the end point of positrons to the ground state would be 0.65 Mev higher than the end point of positrons to the 650-kev excited state. The probable decay schemes of Rh^{98} and Tc^{98} are shown in Fig. 2. Experiments which established the mass assignment are described below in a separate section.

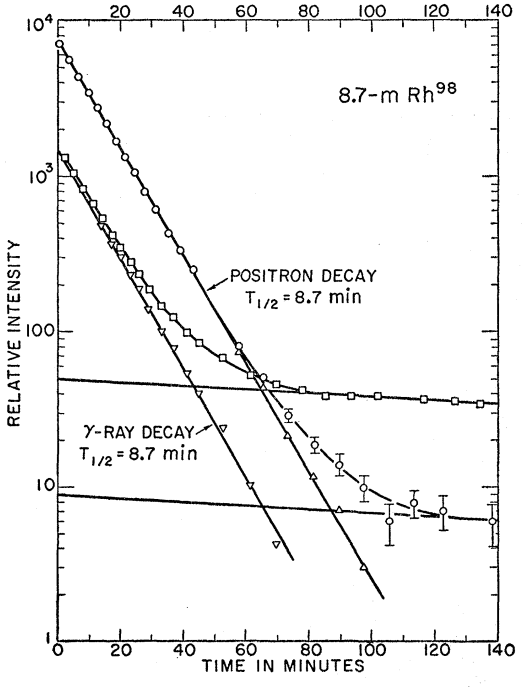

FIG. 1. Decay of β^+ and γ radiation from 8.7-min Rh^{98}.

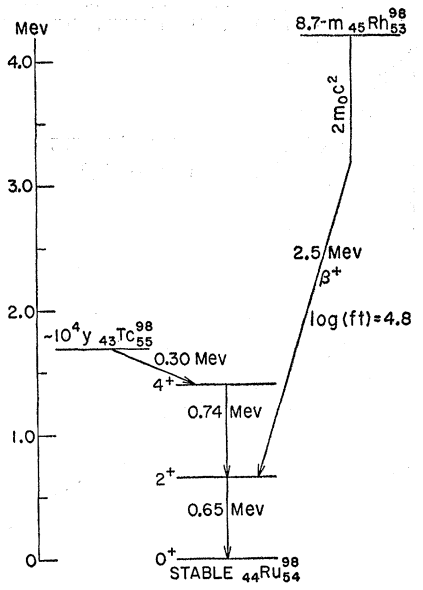

FIG. 2. Probable disintegration schemes of Rh^{98} and Tc^{98}.

17.5-Min Pd^{98}

The radiations of Pd^{98} were not investigated. Its half-life was measured by successive 8.7-min Rh^{98} daughter extractions. The Pd was first separated from the Ru target and purified by the method described above. After the third precipitation with dimethylglyoxime the Pd was put into a few ml of 3*N* HCl. This was then poured in at the top of a Dowex-1 anion exchange column 7 mm in diameter and 4 cm long.[5] It was found that Rh^{+3} is eluted quantitatively from such a column with 4 ml of 6*N* HCl while 60 ml of 6*N* HCl moves the Pd^{+2} down the column only 2–3 cm. Therefore twelve successive Rh daughter extracts were "milked" from the column at 10-minute intervals by passing through 4.5 ml portions of 6*N* HCl. About 2 minutes was required for each portion to pass through the column. Each extract was placed in a 25-ml Erlenmeyer flask and the decay of its activity was followed with a 2 in.×2 in. NaI(Tl) crystal and single channel pulse-height analyzer. The channel was made wide enough to include both the 511- and 650-kev photo lines. Each decay curve showed a satisfactory 8.7-minute component and a longer-lived tail. A plot of the initial Rh^{98} activities against their times of separation from the column shows the decay of the Pd^{98} parent (Fig. 3). The observed half-life, 17.5 ±0.5 minutes, is in good agreement with the most recent literature value[3] of 17±3 minutes.

[5] This procedure was suggested by the review of anion exchange studies by K. A. Kraus and F. Nelson, *International Conference on the Peaceful Uses of Atomic Energy*, Paper No. 837, Geneva, 1955. Proceedings, Vol. 7, p. 113, United Nations, New York (1956).

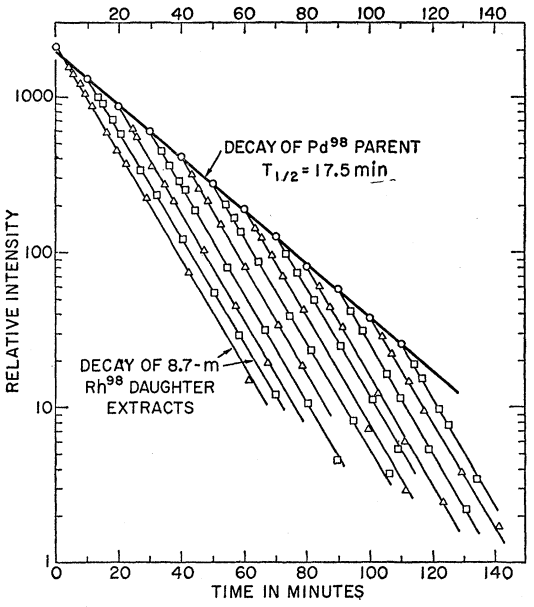

FIG. 3. Successive Rh^{98} daughter extractions showing decay of 17.5-min Pd^{98} parent.

4.7-Hr Rh^{99}

According to Scoville *et al.*,[6] Rh^{99} emits 0.74-Mev positrons and no γ rays. However Fultz *et al.*[7] report a γ ray of 286 kev. In the present work, the maximum positron energy of 0.74 Mev was confirmed by measurement of the maximum pulse size produced in an 8-mm thick anthracene crystal. The γ-ray spectrum (Fig. 4) showed at least three lines: 335 ± 10 kev, 615 ± 15 kev, and 1260 ± 30 kev. The decay of the gross γ-ray activity in several samples was followed through a factor of about 400; a half-life of 4.7 ± 0.1 hours was obtained.

In addition to the three most intense γ rays, a weak line was seen at 1.41 ± 0.04 Mev, and another one appeared at 890 ± 20 kev in coincidence with the most intense γ ray, at 335 kev. No other γ-γ coincidences were found in appreciable intensity. The only β-γ and γ-β coincidences observed were those between the 0.74-Mev positrons and 335-kev γ rays. It appears, therefore, that the positron emission is to the first excited level of Ru^{99} at 335 kev and that the total disintegration energy is 2.10 Mev.

The relative probabilities of some of the modes of decay were estimated by measuring the relative numbers of positrons, K x-rays, and 335-kev γ rays. The positron intensity was estimated from the counting rate obtained with an end-window proportional counter. Corrections were made for geometry, absorption, and the contributions of the x-rays and γ rays. The K x-ray intensity was measured with a 19-mm square of NaI(Tl), 2 mm thick, attached to a photomultiplier and single-channel analyzer. Corrections were made for geometry and for the small contribution of the γ rays to the counting rate. To convert from K x-ray intensity to the total electron capture rate, the fluorescence yield (0.80) and the $K/(K+L)$ capture ratio (0.90) were taken into account. The γ-ray intensities were estimated from the photopeak intensities produced in a standard 1 in.$\times1\frac{1}{2}$ in. NaI(Tl) crystal with the source in a known geometry. These measurements showed that the abundances per disintegration of Rh^{99} are approximately 0.10 for the positrons, 0.90 for orbital electron capture, 0.70 for the 335-kev γ rays, and 0.20 for the 615-kev γ rays.

21.6-Min Pd^{99}

The Pd parent of 4.7-hr Rh^{99} was identified by successive daughter extractions. The method used was the same as for 17.5-min Pd^{98} described above, except that the channel of the pulse-height analyzer was set at the 335-kev photopeak of Rh^{99}. On the resulting Pd^{99} decay curve (Fig. 5), the last few points are less reliable than the others because of some interference by the 312-kev γ ray of 4.7-day Rh^{101}. However the observed half-life value agrees well with that obtained from direct decay of the positrons (Fig. 5). The average value based on three runs is 21.6 ± 0.6 minutes. While this work was in progress Aten and De Vries-Hamerling reported[3] this isotope with a half-life of 24 ± 7 minutes.

In order to obtain 21.6-min Pd^{99} free of 17.5-min Pd^{98}, ruthenium was irradiated with α particles whose energy (17 Mev) was too low for the $(\alpha,2n)$ reaction required to make Pd^{98}. The maximum positron energy of these Pd^{99} samples was determined by comparison with the 1.89-Mev positrons of Ga^{68}. A value of 2.0 ± 0.1 Mev was obtained from scintillation spectrometer measurements and also from aluminum absorption measurements. A gray-wedge photograph of the γ-ray spec-

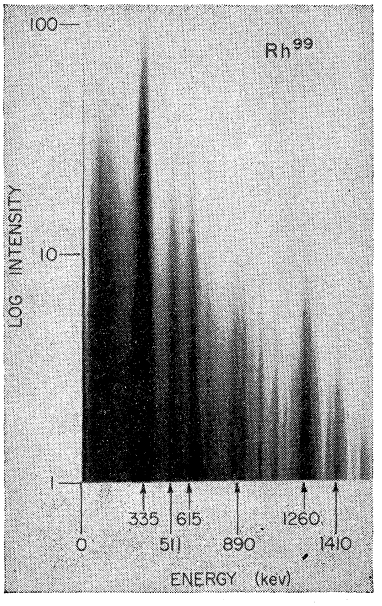

FIG. 4. Gray-wedge photograph of γ-ray spectrum of 4.7-hr Rh^{99}.

[6] Scoville, Fultz, and Pool, Phys. Rev. **85**, 1046 (1952).
[7] Fultz, Nash, Woodward, and Pool, Phys. Rev. **88**, 170 (1952).

trum is shown in Fig. 6 and a summary of all the γ-ray measurements is presented in Table I. The photopeak at 420 kev must be from two different γ rays of about the same energy since 420–420 coincidences were observed. The end point of the positron spectrum observed in coincidence with the 140-kev or 670-kev γ rays was the same as that of the noncoincident spectrum.

4.7-Day Rh^{101}

After decay of the shorter lived components in all of the Rh and Pd samples, Rh^{101} was observed to decay by electron capture with a half-life of 4.7±0.2 days. No positrons were observed and the only γ ray found was at 312±10 kev. The previously reported[6] γ rays at 148 kev could not be detected in the present investigation.

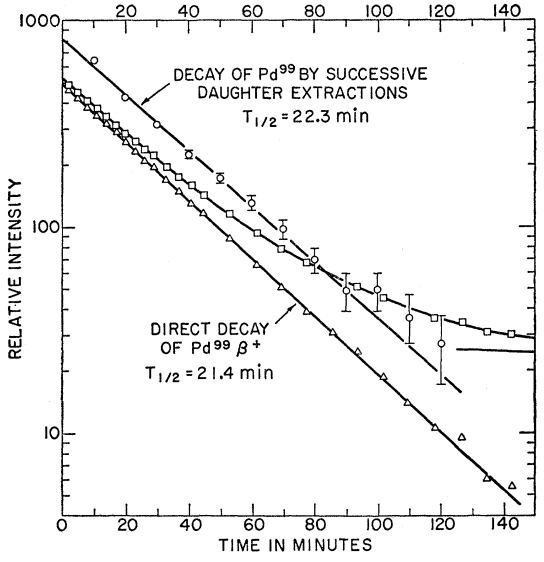

FIG. 5. Decay of 21.6-min Pd^{99} measured directly and by successive daughter extractions.

8.5-Hr Pd^{101}

There are conflicting data in the literature on this isotope. Lindner and Perlman[8] reported a maximum positron energy of 2.3 Mev and no γ rays. Eggen and Pool[9] reported a maximum positron energy of 0.53 Mev. In the present work the latter positron energy is confirmed and several γ rays are reported (Fig. 7). Our value for the maximum β^+ energy is 0.58±0.04 Mev and it is based on a comparison with the 0.54-Mev positrons of Na^{22}. Both gray-wedge scintillation spectrometer and aluminum absorption measurements were made. About 4% of the disintegrations is by positron emission; the remainder is by orbital electron capture.

The energies of the Pd^{101} γ rays are 288, 590, 720, 1190, and 1280 kev, each with a probable error of about 3%. The first of these was observed strongly in coincidence with itself; therefore it represents two γ transitions in cascade. The 590-kev γ ray is probably the cross-over transition. From relative intensities of the K x-rays, positrons, and the most intense γ rays (measured in the same way as described above for Rh^{99}), it was found that the abundance per disintegration is about 0.15 for the 288-kev γ rays and 0.15 for the 590-kev γ rays. The other γ rays are less abundant. Since none of the γ rays was found to be coincident with positrons, the latter must represent transitions to the ground state, and the total disintegration energy is 1.60 Mev.

A half-life of 8.5±0.3 hours for Pd^{101} was determined from three decay curves of total γ radiation and one decay curve of the 288-kev photoline.

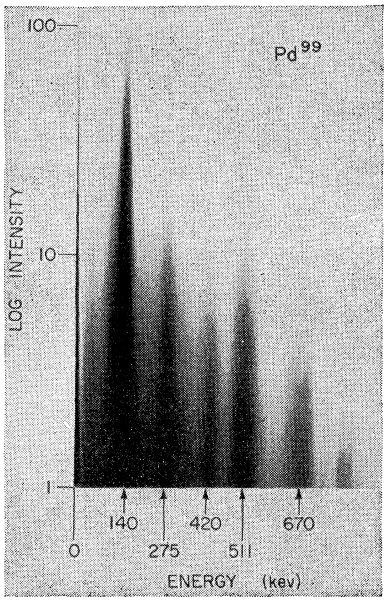

FIG. 6. Gray-wedge photograph of the γ-ray spectrum of 21.6-min Pd^{99}. The low-intensity peak near 800 kev is very likely from addition of the 670- and 140-kev γ rays and of the 420+420 kev γ rays.

Mass Assignments

Mass identification of the Pd and Rh isotopes discussed above was made by bombardment of natural and isotopically enriched Ru samples with alpha particles of various energies. Both the 21.6-min Pd and the 8.5-hr Pd were produced in good yield from natural Ru bombarded with 17-Mev alphas, below the $(\alpha,2n)$ threshold, but 17.5-min Pd was not produced in appreciable yield until the alpha energy exceeded 19 Mev. This observation indicates that the 21.6-min Pd and 8.5-hr Pd are

TABLE I. γ rays from 21.6-min Pd^{99}. Italics identify the more intense lines; parentheses denote weak lines.

Selecting γ ray (kev)	γ rays observed in coincidence (kev)
140	*275*, (420), 511, *670*
275	*140*, 511, (670)
420	140, *420*, 511
511[a]	*140*, 275, 511, *670*
670	140, 275, *511*

[a] Annihilation radiation.

[8] M. Lindner and I. Perlman, Phys. Rev. **73**, 1202 (1948).
[9] D. T. Eggen and M. L. Pool, Phys. Rev. **75**, 1464 (1949).

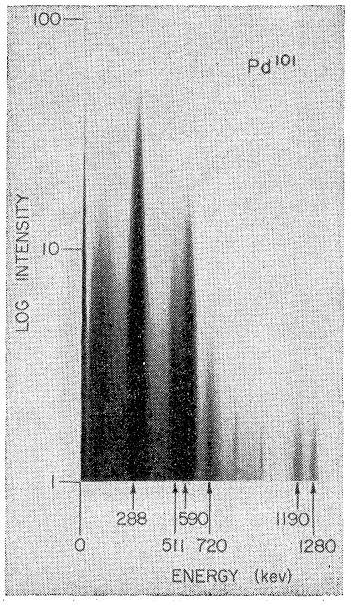

FIG. 7. Gray-wedge photograph of the γ-ray spectrum of 8.5-hr Pd^{101}.

produced by (α,n) reactions on Ru^{96} and Ru^{98} and that their masses are 99 and 101; the 17.5-min Pd is produced by an $(\alpha,2n)$ reaction, probably from Ru^{96}.

Definite assignment of the 21.6-min Pd to mass 99 was made when it was found that its yield was enhanced by use of a target enriched in Ru^{96} and diminished by use of a target depleted in Ru^{96}. The 8.5-hr Pd was assigned to mass 101 by the same kind of observations. The isotopic compositions of the various targets are shown in Table II. The values for the enriched Ru samples were supplied by the Stable Isotopes Research and Production Division of Oak Ridge National Laboratory. In the irradiations of ruthenium with 29-Mev alpha particles the ratio of 17.5-min Pd to 21.6-min Pd was the same (within 30%) whether the target was natural Ru or 13-fold enriched in Ru^{96}. Therefore it seems highly probable that both Pd isotopes were produced from the same Ru isotope: 21.6-min Pd^{99} by an (α,n) reaction and 17.5-min Pd^{98} by an $(\alpha,2n)$ reaction on Ru^{96}. Added confirmation of these assignments comes from a comparison, for 29-Mev alphas, of the change in the Pd^{98}/Pd^{101} activity ratio with the change in the Ru^{96}/Ru^{99} abundance ratio when an enriched Ru^{99} target was substituted for a natural Ru target. For 29-Mev alpha particles on these targets Pd^{98} and Pd^{101} should be produced mostly by $(\alpha,2n)$ reactions from Ru^{96} and Ru^{99}, respectively. Therefore the change in the activity ratio should approximately follow the change in the abundance ratio. The experiments showed that the Pd^{98}/Pd^{101} ratio decreased 370-fold when the Ru^{96}/Ru^{99} ratio decreased 200-fold. The discrepancy is not significant because of the large uncertainty in the abundance of Ru^{96} in the enriched Ru^{99} and because of other experimental errors. In these experiments the Pd activities were determined by measurements on their Rh daughters. The irradiation times, separation times, and growth times were approximately constant from run to run but some error was introduced owing to small variations in these times. The mass assignments determined here confirm previous tentative assignments.

DISCUSSION

Although the data presented here are insufficient to establish decay schemes for most of the nuclides investigated, certain tentative spin and parity assignments can be made. For example, in 8.5-hr Pd^{101} the 580-kev positron emission to the ground state of Rh^{101} corresponds to an allowed transition with $\log(ft)$ of 5.1. The only likely single-particle model assignment for the initial state is $d_{5/2}$ or $g_{7/2}$ and the only probable assignment for the final state is $p_{1/2}$ or $g_{9/2}$. A selection is made in favor of $g_{7/2}$ for the Pd^{101} ground state and $g_{9/2}$ for the Rh^{101} ground state because only this combination would lead to an allowed β^+ transition. The observed decay of Rh^{101} to a 312-kev level of Ru^{101} is also consistent with these assignments. The energy of this K-capture transition is estimated as approximately 0.5 Mev from β-decay systematics[10]; therefore the $\log(ft)$ value is about 4.5 and the transition is allowed. A reasonable assignment for this 312-kev level is $g_{7/2}$ with an $M1$ γ transition to the known[11] $d_{5/2}$ ground state of Ru^{101}.

TABLE II. Isotopic constitutions of Ru targets.

Ru isotope	Abundance in atom percent		
	Natural Ru [a]	Enriched Ru^{96}	Enriched Ru^{99}
96	5.50	74.1±0.3	0.2±0.05
98	1.91	0.6±0.1	0.5±0.1
99	12.70	2.7±0.1	91.2±0.3
100	12.69	1.6±0.1	4.3±0.1
101	17.01	2.4±0.1	1.6±0.1
102	31.52	16.5±0.2	1.3±0.1
104	18.67	2.1±0.05	1.0±0.1

[a] L. Friedman and A. P. Irsa, J. Am. Chem. Soc. **75**, 5741 (1953).

Level assignments of Pd^{99} and Rh^{99} can be made in a similar way. The 740-kev positrons, $\log(ft)$ 5.0, of 4.7-hr Rh^{99} probably represent a transition from a $g_{9/2}$ level to a $g_{7/2}$ level at 335 kev in Ru^{99}. The ground state of Ru^{99} is known[11] to be $d_{5/2}$. For 21.6-min Pd^{99}, the observed positrons have a $\log(ft)$ value of 4.7 but the transition is not to the $g_{9/2}$ ground state of Rh^{99}. Therefore the initial state (Pd^{99}) is probably $d_{5/2}$ rather than $g_{7/2}$ because the latter assignment would permit an allowed ground-state transition.

[10] K. Way and M. Wood, Phys. Rev. **94**, 119 (1954).
[11] J. H. E. Griffiths and J. Owen, Proc. Roy. Soc. (London) **A65**, 951 (1952).

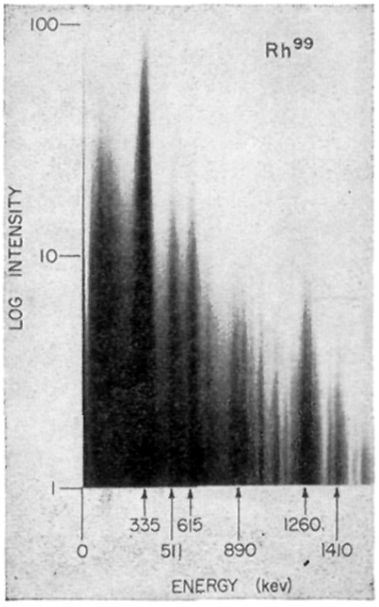

FIG. 4. Gray-wedge photograph of γ-ray spectrum of 4.7-hr Rh^{99}.

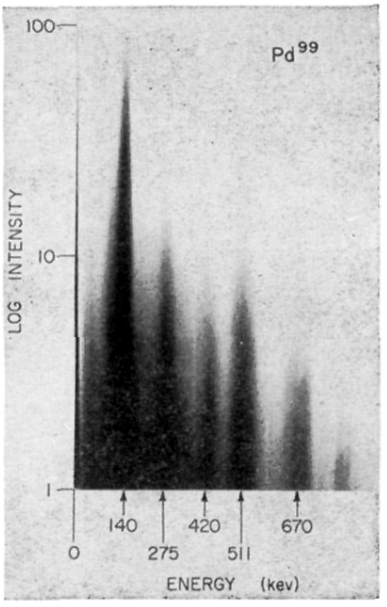

FIG. 6. Gray-wedge photograph of the γ-ray spectrum of 21.6-min Pd^{99}. The low-intensity peak near 800 kev is very likely from addition of the 670- and 140-kev γ rays and of the 420+420 kev γ rays.

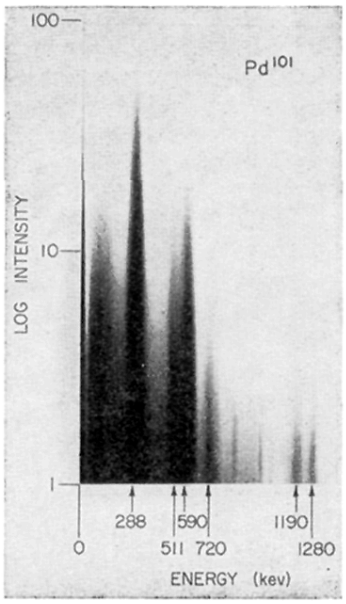

FIG. 7. Gray-wedge photograph of the γ-ray spectrum of 8.5-hr Pd^{101}.