Neutron capture cross sections of ^{108,110}Pd

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The thermal cross sections and resonance integrals of ^{108,110}Pd have been deduced for radiative neutron captures leading to both the ground and metastable states of ^{109,111}Pd. These determinations were based on measurements of the intensity of the γ rays emitted in the decays of ^{109}Pd^{g,m} and ^{111}Pd^{g,m} following neutron irradiation of ^{108,110}Pd. To facilitate more reliable values of the cross sections, improved values for the half-lives of these decays have also been measured.

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I. INTRODUCTION

In a previous publication [1] from our laboratory describing a measurement of the neutron capture cross sections of ^{102}Pd from irradiation of Pd samples of natural isotopic abundance, it was reported that the activity of 7.45-d ^{111}Ag (produced following the decays of ^{111}Pd^{g,m}) present in the samples was inconsistent with its expected production based on the accepted cross sections of ^{110}Pd. Because our previous experiments did not study the decays of the shorter-lived Pd isotopes, we were unable to directly determine the cross sections for the production of ^{111}Pd. The present report describes a new set of measurements of the thermal capture cross sections and resonance integrals of ^{110}Pd by neutron irradiation of Pd. This report gives the results of those measurements along with a determination of the cross sections of ^{108}Pd leading to ^{109}Pd^{g,m}.

A new determination of the Pd cross sections has been reported by Krtička *et al.* [2], based on a measurement of the intensities of the prompt γ rays that lead to states in the Pd isotopes following neutron capture. By simulating the unobserved and continuum contributions to the γ-ray intensities, these authors were able to produce a complete accounting of the total radiative intensity and thus determine the radiative capture cross section for thermal neutrons. By comparing the resulting cross sections with equally precise determinations from the activation method in the present report, it is possible to test the reliability of the simulations involved in the former and of the branching intensities and other decay parameters necessary for the analysis of the latter.

II. EXPERIMENTAL DETAILS

The experimental methods employed in this work for measuring the cross sections are identical to those of the previous work and will only be summarized here. Further details may be found in Ref. [1].

Sources of natural Pd metal of thickness 0.025 mm were irradiated at the Oregon State University TRIGA Reactor. Simultaneous irradiations of Au, Co, and Zr were done to determine the neutron flux. Two different irradiation sites in the reactor were used for this work: a thermal column (with nominal thermal and epithermal neutron fluxes, respectively, 7×10^{10} and 2×10^{8} neutrons cm^{-2} s^{-1}) and a fast pneumatic transfer facility (respectively, 1×10^{13} and 3.5×10^{11} neutrons cm^{-2} s^{-1}). Some samples irradiated in the latter facility were enclosed in Cd to isolate the epithermal component.

The γ rays were observed with a high-purity Ge detector (efficiency 35% compared with NaI at 1332 keV) coupled to a digital spectroscopy system. The resolution (full width at half maximum) was typically 1.68 keV at 1332 keV. For the cross-section determination, isolated peaks were analyzed using the Maestro software [3]. Energy and efficiency calibrations were done using sources of ^{152}Eu and ^{133}Ba.

Analysis of the activation data to determine cross sections requires knowledge of the isotopic abundances, half-lives, and γ-ray branching ratios. The currently accepted [4] values of the isotopic abundances are $26.46 \pm 0.09\%$ for ^{108}Pd and $11.72 \pm 0.09\%$ for ^{110}Pd; these values were adopted for the analysis of the present work. Values of the other parameters used in the cross-section analysis, including half-lives and γ-ray branching ratios, are compiled in Table I. Half-lives are from the present work, and γ-ray branching ratios are from a recent spectroscopic study [5], except for ^{109}Pd^{m} for which the γ-ray branching is taken from the Nuclear Data Sheets (NDS) compilation [6].

III. HALF-LIFE MEASUREMENTS

There is considerable variation in the reported measurements of the half-life of ^{109}Pd^{g}. The value previously recommended by the NDS, 13.7012 ± 0.0024 h, was reported by Abzouzi *et al.* [7], but that value is in substantial disagreement with previously measured smaller values: 13.427 ± 0.014 h by Gindler and Glendenin [8], 13.45 ± 0.01 h by Starner [9], and 13.47 ± 0.01 h by Brandhorst and Cobble [10]. Other previous less precise values, including those of Gueben and Govaerts [11] (13.99 ± 0.16 h), Bormann *et al.* [12] (13.67 ± 0.07 h), and Chatterjee and Baliga [13] (13.85 ± 0.17 h), favor the larger value of the half-life although they would agree with the smaller value within about 3 standard

TABLE I. Properties of Pd isotopes for cross-section determination.

Pd Isotope	$t_{1/2}$	γ ray (keV)	Branch (%)
109g	13.45(1) h	88.0	3.67(10)
		309.2+311.4	0.0380(10)
		636.3	0.0111(3)
		647.3	0.0260(7)
		781.4	0.0121(3)
109m	4.694(2) min	189.0	56.0(3)
111g	23.6(1) min	547.0	0.332(20)
		580.0	0.730(43)
		650.5	0.481(29)
		835.8	0.233(14)
		1388.4	0.425(25)
111m	5.563(13) h	172.2	35.4(8)
		391.3	4.65(35)
		575.1	2.79(21)
		632.6	2.92(22)
		694.1	1.71(13)
		762.0	0.99(8)

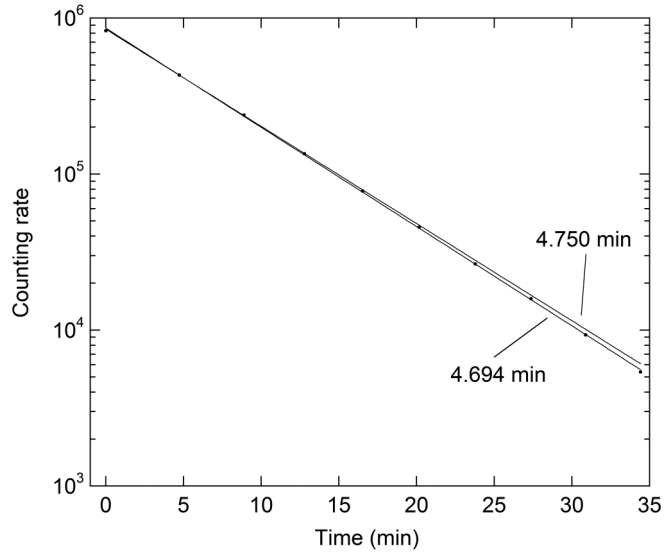

FIG. 2. Counting rate vs. time of 189.0-keV γ ray in the decay of $^{109}Pd^{m}$. The previous half-life value of 4.750 min is a poor fit to the data.

deviations. NDS presently recommends an intermediate value of 13.59 ± 0.12 h.

To resolve this discrepancy, the decay of $^{109}Pd^{g}$ has been followed in the present work by observing the 88.0-keV γ ray. Figure 1 shows this decay, which yields a half-life of 13.45 ± 0.01 h, in agreement with the smaller set of values and in substantial disagreement with the result of Abzouzi *et al.* The uncertainty quoted for the present result is at the level of one standard deviation and includes statistical uncertainties only. Two different types of systematic uncertainties were investigated. The first is the effect of the background under the 88-keV peak, which is due to higher-energy γ rays from decays with different half-lives. By using background regions of differing widths to evaluate the peak area, and by measuring the half-life with and without the presence of 5.5-h $^{111}Pd^{m}$ (a main contributor to the background in the 88-keV region) in the sample, it can be concluded that there is no effect on the deduced half-life at the level of 1 part in 10^4. A second possible contribution to systematic uncertainty is associated with variations in the dead-time corrections of the counting system as the source activity decreases. This effect was assessed by measuring the $^{109}Pd^{g}$ half-life with a ^{60}Co source present. From the analysis of the ^{60}Co γ rays, it was determined that the dead-time compensation of the counting apparatus had no effect on the half-life determination at the level of 1 part in 10^4. It can thus be concluded that the two major sources of systematic uncertainty could contribute to the present result only at a level that is below the statistical uncertainty. Similar tests for systematic effects were made for the remaining half-life measurements discussed below, with similar results and conclusions.

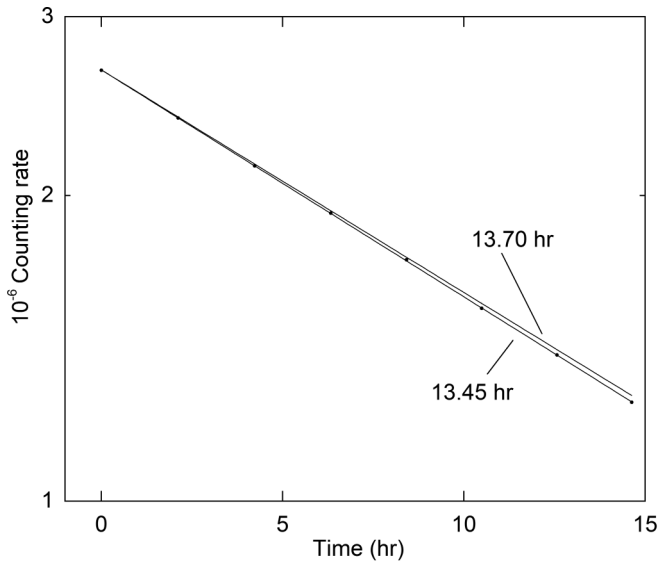

FIG. 1. Counting rate vs. time of 88.0-keV γ ray in the decay of $^{109}Pd^{g}$. The previous half-life value of 13.70 h is a poor fit to the data.

The two most precise previous values of the half-life of $^{109}Pd^{m}$ come from the same research group but are in substantial disagreement with one another: 4.696 ± 0.003 min by Abzouzi *et al.* [7] and 4.750 ± 0.004 min by Antony *et al.* [14]; the two values disagree with one another by more than 10 standard deviations. The present experiments have followed this decay by observing the 189.0-keV isomeric transition over more than 3 half-lives from each of 6 different samples (see Fig. 2), yielding a half-life of 4.694 ± 0.002 min, in agreement with the earlier result of Abzouzi *et al.* [7] and disagreeing with the later result of Antony *et al.* [14].

The currently accepted value of the half-life of $^{111}Pd^{g}$ is 23.4 ± 0.2 min, based on a measurement reported by Kracíková *et al.* [15]. From measurements with four different samples in which the 547- and 580-keV γ rays were counted over 3 half-lives (see Fig. 3), the present result for this half-life was determined to be 23.6 ± 0.1 min, in excellent

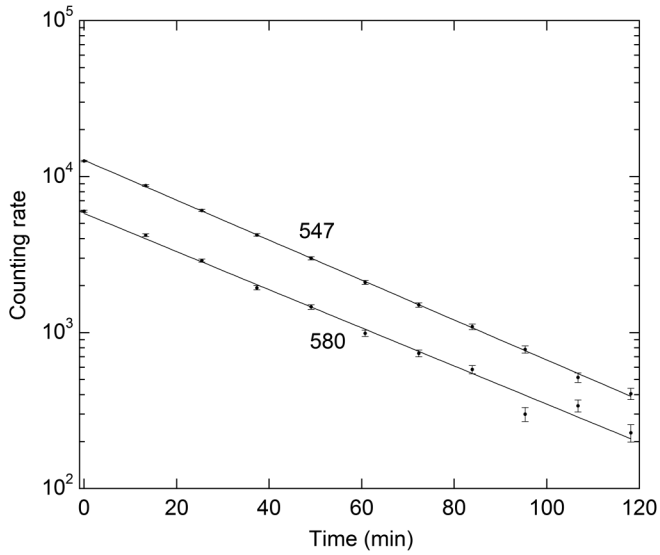

FIG. 3. Counting rates vs. time of 547- and 580-keV γ rays in the decay of ^{111}Pd^{g}.

agreement with the result of Kracíková *et al.* The current value of the ^{111}Pd^{m} half-life, 5.5 ± 0.1 h, is based on an early measurement reported by McGinnis [16]. In the present experiments, the 172.2-keV γ ray has been counted from 4 samples, each decaying over more than 3 half-lives (Fig. 4), yielding a value of 5.563 ± 0.013 h, in excellent agreement with but an order of magnitude more precise than the previous result of McGinnis.

IV. CROSS-SECTION MEASUREMENTS

For all four of the neutron capture processes considered in the present work (^{108}Pd $\rightarrow$ ^{109}Pd^{g,m}, ^{110}Pd $\rightarrow$ ^{111}Pd^{g,m}), the resonance integral is at least an order of magnitude larger

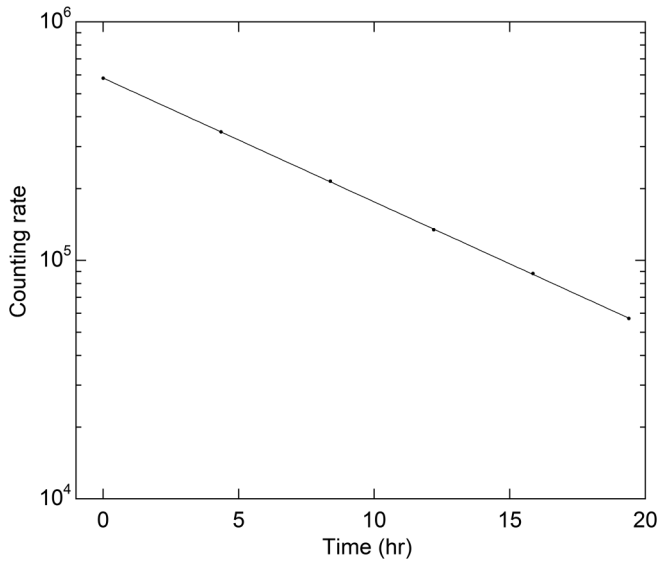

FIG. 4. Counting rate vs. time of 172.2-keV γ ray in the decay of ^{111}Pd^{m}.

TABLE II. Activation cross sections for neutron capture by ^{108,110}Pd.

Pd capture Reaction	I (b)	σ (b)
108 $\rightarrow$ 109g	138(8)	7.7(5)
108 $\rightarrow$ 109m	3.07(15)	0.172(10)
110 $\rightarrow$ 111g	3.33(23)	0.289(21)
110 $\rightarrow$ 111m	0.152(12)	0.0116(6)

than the thermal cross section. Thus data for determination of the thermal cross section must be corrected for the effect of the epithermal neutrons. Considering both contributions, the activity a produced following neutron irradiation for a time t_i and subsequent decay for a time t_d can be written as

$$a = e^{-\lambda t_d}(\phi_{\text{th}}\sigma + \phi_{\text{epi}}I)N(1 - e^{-\lambda t_i}), \quad (1)$$

where ϕ_{th} and ϕ_{epi} are, respectively, the thermal and epithermal neutron fluxes, σ is the thermal cross section, I is the resonance integral, and N is the number of target nuclei (assumed constant).

The resonance integrals were deduced from a series of runs in which the Pd samples and the flux monitors were enclosed in a Cd box of 1-mm wall thickness and irradiated in the pneumatic transfer facility for periods of 2 to 5 min. Data accumulation typically began 5 to 20 min following the irradiations. The Pd samples were counted at distances of 10 to 20 cm from the Ge detector to minimize summing corrections. Typical activities following bombardment were: ^{109}Pd^{m} – 33 MBq; ^{109}Pd^{g} – 7 MBq; ^{111}Pd^{m} – 0.008 MBq; ^{111}Pd^{g} – 2 MBq.

The results from the present determination of the Pd resonance integrals are shown in Table II. Results for capture to the ground state have been corrected for the effects of captures proceeding through the isomer. These data represent the averages of the results from several samples. Uncertainties are mainly due to the intercomparison of the flux monitors (±3%) and to the branching ratios (±3–8%); statistical (counting) uncertainties and uncertainties in detector efficiencies are generally below 1%.

Using these data to correct for the epithermal captures permits the contribution to the activation due to thermal captures to be isolated. For the irradiations in the thermal column, where the thermal-to-epithermal ratio is approximately 400, the effect of a resonance integral that is an order of magnitude greater than the thermal cross section amounts to a correction of a few percent. For irradiations in the pneumatic transfer facility, where the thermal-to-epithermal ratio is about 30, the correction is about 30%. Even though this is a large effect, the correction can be made with great precision and does not significantly increase the uncertainty of the deduced thermal cross sections.

Thermal column samples were irradiated for 1 hr, producing activities as follows: ^{109}Pd^{g} – 0.9 MBq; ^{111}Pd^{m} – 0.001 MBq; ^{111}Pd^{g} – 0.2 MBq. Samples irradiated in the pneumatic transfer facility for 5−10 min produced activities of: ^{109}Pd^{m} – 90 MBq; ^{109}Pd^{g} – 46 MBq; ^{111}Pd^{m} – 0.07 MBq; ^{111}Pd^{g} – 17 MBq. Sample γ-ray spectra are

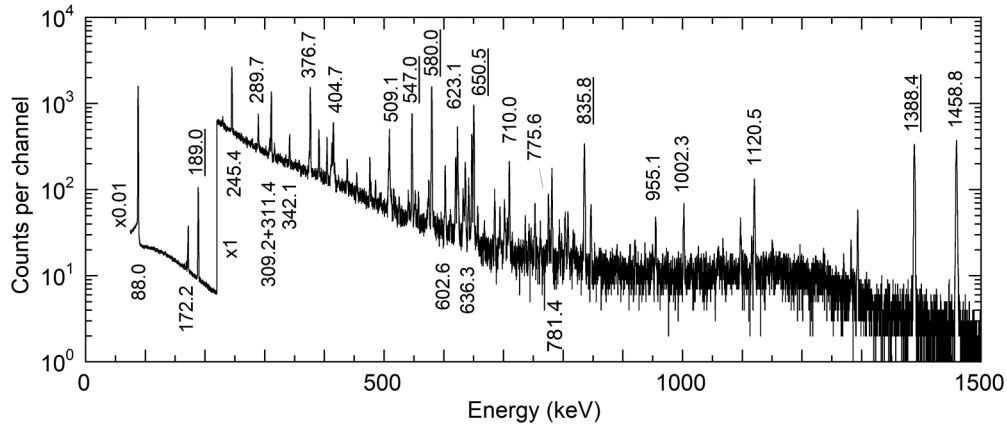

FIG. 5. Partial γ-ray spectrum of Pd sample taken immediately after irradiation. Lines from short-lived decays (^{109}Pd^{m} and ^{111}Pd^{g}) are labeled above the spectrum; underlined labels show lines used in the cross-section determination. Lines from longer-lived decays (^{109}Pd^{g}, ^{111}Pd^{m}, and ^{111}Ag) are labeled below the spectrum.

shown in Figs. 5 and 6. The results for the thermal cross sections are shown in Table II. Except for the case of captures leading to ^{109}Pd^{m} (for which thermal column data could not be obtained because of the short half-life), values for the thermal cross sections from irradiations in the thermal column and the fast pneumatic transfer facility agreed with one another; this good agreement, despite the differing thermal fluxes and epithermal-to-thermal ratios, lends confidence to the present results. The known neutron resonances [17,18] in ^{108}Pd and ^{110}Pd are narrow and occur sufficiently far from the thermal region that the cross section shows the usual $1/v$ behavior in the thermal region.

The cross sections of ^{108}Pd leading to ^{109}Pd^{g} listed in Table II are consistent with the values reported in our previous work [1] ($\sigma = 8.3 \pm 0.9$ b, $I = 120 \pm 18$ b) as well as with the ratio $I/\sigma = 15.5$ obtained by Van der Linden *et al.* [19]. Previous values for the thermal capture cross section leading to ^{109}Pd^{m} measured by the activation technique include 0.26 ± 0.04 b by Sehgal *et al.* [20] and 0.17 ± 0.02 b by Tilbury and Kramer [21].

The ^{110}Pd $\rightarrow$ ^{111}Pd^{g} cross sections are in acceptable agreement with previously measured values from activation ($\sigma = 0.19 \pm 0.03$ b by Sehgal *et al.* [20] and $I/\sigma = 12.7 \pm 1.5$ by Van der Linden *et al.* [19]). The latter value is independent of the branching ratios, but the former would be revised upward by about 25% if the present branching ratios had been used, bringing it into better agreement with the presently deduced cross section. The ^{110}Pd $\rightarrow$ ^{111}Pd^{m} cross sections are not in good agreement with the results of previous work ($\sigma = 0.037 \pm 0.006$ b by Mangal and Gill [22]; $\sigma = 0.033 \pm 0.003$ b and $I = 0.66 \pm 0.07$ b by Heft [23]), but they are in agreement with a measurement reported by De Corte *et al.* [24], $\sigma = 0.012$ b. The relatively small differences between the present and previous branching ratios are not able to account for these large discrepancies between the reported cross sections. A previous measurement [25] of the thermal cross-section ratio $\sigma(m)/[\sigma(m) + \sigma(g)]$ gave the value 0.045 ± 0.001, which compares well with the value deduced from the present results, 0.038 ± 0.003. If the isomeric cross section were as large as 0.033 b, the expected value of this ratio would

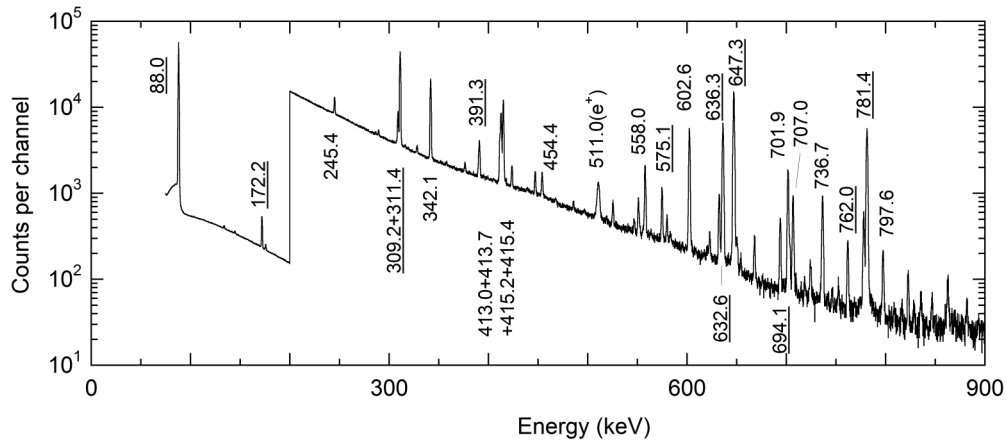

FIG. 6. Partial γ-ray spectrum of longer-lived Pd activities (^{109}Pd^{g} and ^{111}Pd^{g}). Underlined labels show lines used in the cross-section determination.

be in the range of 0.09-0.10, in serious disagreement with the measured ratio.

In a more recently reported study of the cross sections of many nuclides of interest for neutron activation analysis, Farina Arboccò *et al.* [26] reported values of $\sigma(m)+\sigma(g)$ of 8.57 ± 0.09 b for ^{109}Pd and 0.291 ± 0.004 b for ^{111}Pd, along with the separate value of 0.0130 ± 0.0002 b for ^{111}Pd^m. These results, obtained in part by observing the decays of the Ag daughter radioisotopes, are in good agreement with the present ones.

V. DISCUSSION

A. Production of ^{111}Ag

The newly determined values of the ^{110}Pd cross sections nicely resolve the discrepancy in the production of ^{111}Ag^g reported in the previous work [1], in which the use of the values of the cross sections recommended at that time [$\sigma(m) = 0.037$ b, $I(m) = 0.7$ b, $\sigma(g) = 0.19$ b, $I(m) = 2.4$ b] led to predicted values of the ^{111}Ag^g activity that were more than 20% too small. There are altogether six different paths that lead from neutron capture by ^{110}Pd to 7.45-d ^{111}Ag^g: capture to ^{111}Pd^m or ^{111}Pd^g followed by β decay to 70-s ^{111}Ag^m or to ^{111}Ag^g (4 paths) and capture to ^{111}Pd^m followed by the isomeric decay to ^{111}Pd^g and then by β decay to ^{111}Ag^m or ^{111}Ag^g (2 paths). The 70-s isomer in ^{111}Ag decays entirely by an isomeric transition to the ground state. Owing to the relative cross sections (which favor production of ^{111}Pd^g over ^{111}Pd^m) and the relative decay probabilities (which favor the isomeric decay of ^{111}Pd^m over its β decay and also favor the β decays of both ^{111}Pd^m and ^{111}Pd^g to ^{111}Ag^m over their decays to ^{111}Ag^g), the path ^{110}Pd $\rightarrow$ ^{111}Pd^g $\rightarrow$ ^{111}Ag^m $\rightarrow$ ^{111}Ag^g typically accounts for about 96% of the production of ^{111}Ag^g observed about one day after the irradiation, with most of the remainder (3%) following the path ^{110}Pd $\rightarrow$ ^{111}Pd^m $\rightarrow$ ^{111}Pd^g $\rightarrow$ ^{111}Ag^m $\rightarrow$ ^{111}Ag^g. The production of ^{111}Ag^g is therefore very insensitive to the cross sections to form ^{111}Pd^m.

As an example of the resolution of the previously observed discrepancy in the production of ^{111}Ag^g, one experiment in the rabbit facility yielded a ^{111}Ag^g activity of 9.11 kBq measured 18 h after the irradiation based on the observation of the intensity of the 342-keV γ ray (6.68%). The calculated activity is 7.01 kBq using the previous values of the cross sections and 8.95 kBq using the present cross sections. A similar measurement in the thermal column resulted in an observed ^{111}Ag^g activity of 1.05 kBq, compared with calculated values of 0.80 kBq (previous cross sections) and 1.08 kBq (present values). An additional irradiation using only epithermal neutrons in a Cd-shielded facility yielded a measured activity of 212 kBq, compared with calculations of 181 kBq (previous values) and 202 kBq (present values). Thus, within the 7% uncertainty of the present ^{111}Pd^g cross sections, the production of ^{111}Ag^g is systematically inconsistent with the previous values of the cross sections and consistent with the present values.

B. Isomer ratios in ^{109,111}Pd

The formation of radioactive ground states or isomers following neutron capture depends primarily on two factors: the distribution of resonance states produced in the capture process and the multiplicity of the γ-ray cascade de-exciting the resonance states to reach either the radioactive decaying isomer or ground state. Assuming s-wave capture by ^{108}Pd and ^{110}Pd, the resonances would be spin 1/2. If the γ-ray cascade were identical irrespective of whether the resonances are produced by thermal or epithermal neutrons, then the isomer ratios, defined for thermal and epithermal neutrons as

$$R_{\rm th} = \frac{\sigma(m)}{\sigma(g)+\sigma(m)} \qquad R_{\rm epi} = \frac{I(m)}{I(g)+I(m)} \tag{2}$$

should be in agreement for each nuclide. The isomer ratios deduced from the presently measured cross sections are

$$108 \rightarrow 109 : R_{\rm th} = 0.0219 \pm 0.0019, R_{\rm epi} = 0.0218 \pm 0.0017,$$

$$110 \rightarrow 111 : R_{\rm th} = 0.0385 \pm 0.0034, R_{\rm epi} = 0.0437 \pm 0.0046.$$

The good agreement of the thermal and epithermal isomer ratios for each nuclide supports the validity of the model. If, for example, *p*-wave capture were occurring, then production of the high-spin isomer would be enhanced because one fewer dipole transition would be required to reach the high-spin isomer from the 3/2 capture state. Since p-wave capture would be more likely for the epithermal neutrons, the agreement of $R_{\rm th}$ and $R_{\rm epi}$ suggests that p-wave capture does not have a significant effect.

A more formal model of this sort was developed by Huizenga and Vandenbosch [27]. In their model the isomer ratios depend on two parameters: the width σ of the statistical distribution (in the form of a skewed Gaussian [28]) describing the density of nuclear spin states and the number N_γ of γ rays in the cascade between the capture states and the isomeric or ground state. Assuming that only dipole transitions contribute to the cascade, one can use the statistical distribution of spin states for various values of the width parameter to calculate the populations of different spin states after each particular step in the cascade. At the final step, states with spins 1/2 to 7/2 will populate the spin-5/2 ground state, while states with spins of 9/2 to 13/2 will populate the 11/2 isomer.

The path from a spin-1/2 capture state to the 11/2 isomer must comprise a minimum of five dipole transitions. For a width parameter of $\sigma = 3$, the resulting isomer ratio for $N_\gamma = 5$ is 0.022, in excellent agreement with the values for ^{109}Pd. For a broader distribution with $\sigma = 4$ and $N_\gamma = 5$, the isomer ratio is 0.036, which agrees with the values for ^{111}Pd. The thermal cross sections for capture by ^{106}Pd to ^{107}Pd^{g,m} have been tabulated by Mughabghab [18]. Based on those values, $R_{\rm th}$ for capture by ^{106}Pd is calculated to be 0.0426 ± 0.0077, which would agree with the broader distribution similar to ^{111}Pd (and in fact the thermal cross sections for ^{106}Pd, 0.292 ± 0.029 b and 0.013 ± 0.002 b, are identical with those of ^{110}Pd).

VI. CONCLUSIONS

Enabled by a redetermination of the decay parameters (half-life and γ-ray branching ratios), the present work has

been able to produce a precise and self-consistent set of resonance integrals and thermal cross sections using the activation technique for neutron capture by the naturally occurring isotopes of Pd leading to the ground and isomeric states of the radioisotopes ^{109}Pd and ^{111}Pd. Consistent values were obtained from irradiations at two different reactor sites with very different relative fluxes of thermal and epithermal neutrons. The ^{110}Pd cross sections directly measured by observing the shorter-lived ^{111}Pd decays are consistent with those necessary to account for the production of the longer-lived daughter ^{111}Ag.

The effectiveness of the activation method, with its need for accurate decay scheme information, can be tested by comparing the deduced cross sections with those determined from a different method that does not rely on such additional parameters. The Pd thermal cross sections have been determined by Krtička *et al.* [2] using a method based on the observation of the primary and secondary γ rays following neutron capture. Their results for the thermal cross sections are 7.2 ± 0.5 b for ^{109}Pd^{g}, 0.185 ± 0.011 b for ^{109}Pd^{m}, and 0.34 ± 0.10 b for ^{111}Pd^{g}. These results are in excellent agreement with the present results, such agreement providing a strong verification of the assumptions underlying both methods.

Assuming a model based on a statistical distribution of spin states in the nuclear level density, there is evidence to suggest that the distribution is broader for capture by ^{106}Pd and ^{110}Pd than it is for ^{108}Pd.

ACKNOWLEDGMENTS

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