Neutron capture cross section of ^{102}Pd

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The cross sections for radiative neutron capture by ^{102}Pd have been deduced from a measurement of the γ rays emitted by 17.0-d ^{103}Pd. The thermal cross section has been determined to be $\sigma = 1.82 \pm 0.20$ b, and the effective resonance integral is $I = 23 \pm 4$ b. We also report thermal and resonance capture cross sections for ^{108}Pd and note possible inconsistencies with the presently accepted values of the ^{110}Pd cross sections.

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I. INTRODUCTION

The radioisotope ^{103}Pd has found wide application in brachytherapy treatment of various cancers, particularly those of the prostate and the eye [1]. In such treatment, seeds of ^{103}Pd are implanted at the site of the tumor. Most of the radiation is emitted in the form of x rays (20–23 keV) and low-energy γ rays (40 keV). The short range of these low-energy radiations minimizes damage to the surrounding healthy tissue. Directionality can be achieved by partially encasing the seed in a material such as gold that absorbs the low-energy radiations.

^{103}Pd can be produced through neutron capture by stable ^{102}Pd. Only one previous measurement of this cross section has been reported [2], with a result of 4.8 b for reactor neutrons, based on a comparison with the capture cross section of ^{108}Pd (assumed in Ref. [2] to have the value 11.7 b). However, the presently accepted value [3] of the ^{108}Pd cross section, 8.5 b, differs from the value assumed by Meinke [2]. This difference requires a renormalization of Meinke's value of the ^{102}Pd cross section to 3.4 b, the presently accepted value.

The ^{102}Pd resonance integral has not been previously measured. Its value has been calculated [3] to be 10 ± 2 b, based on the measured resonance parameters of ^{102}Pd. Because of the importance of having precise and reliable cross sections for the production of ^{103}Pd, we have undertaken to measure the thermal cross section and resonance integral of ^{102}Pd.

II. EXPERIMENTAL DETAILS

The neutron irradiations were performed at the Oregon State TRIGA reactor (OSTR) [4]. Three different irradiation facilities were used: the thermal column (TC), in-core irradiation tube (ICIT), and cadmium-lined in-core irradiation tube (CLICIT). Each Pd irradiation was accompanied by a variety of primary and secondary flux monitors. Primary flux monitors were Au, in the form of a dilute (0.134%) alloy of Au in Al metal, and Co, in the form of a dilute (0.438%) alloy of Co in Al metal. Secondary flux monitors were ^{58}Fe, ^{64}Zn, ^{94}Zr, and ^{96}Zr. All secondary flux monitors were in the form of thin metal foils of natural isotopic abundances. Cross sections and decay parameters of flux monitors are listed in Table I.

Samples of ^{103}Pd were produced by irradiating Pd metal foils (mass 10–100 mg, thickness 0.025 mm) of natural isotopic abundance (1.02% of ^{102}Pd). Owing to the low abundance of ^{102}Pd and the small branching ratio of even the most intense observable γ rays emitted by ^{103}Pd, the effect of isotopic and elemental impurities in the samples requires careful attention. It is therefore necessary to study the γ spectra at the best possible detector resolution and to identify and understand the behavior of the impurities in order to have confidence that the impurities do not affect the intended ^{102}Pd measurement.

Following neutron irradiation for a time t_i and decay for a time t_d, the activity a can be written as

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

where $\lambda = \ln 2/t_{1/2}$ is the decay constant of the produced nucleus, 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 nuclei in the target (assumed to be constant).

The γ rays were observed with a high-purity Ge detector (nominal volume = 169 cm^3, efficiency = 35% compared with NaI at 1332 keV). The detector signal was processed by a digital spectroscopy system connected to a desktop computer. The counting system was optimized to produce a resolution of 1.68 keV full width at half maximum for the 1332-keV ^{60}Co γ ray. For the 357.4-keV γ ray of ^{103}Pd analyzed in this experiment, the resolution was typically 1.0 keV.

The low emission rate of the 357.4-keV γ ray of ^{103}Pd, resulting from the low abundance of ^{102}Pd and the low 357.4-keV branching ratio, necessitated a small source-to-detector distance to obtain a statistically significant counting rate. Distances as small as 2.5 cm were used in these experiments. At this small distance, the effects of coincidence summing can distort the γ spectrum. Fortunately, summing is negligible in the ^{103}Pd decay, but it does pose difficulties for the ^{133}Ba and ^{152}Eu calibration sources used to determine the detector efficiency. Summing corrections for the calibration sources were determined by comparing each calibration source at various distances (2.5 to 20 cm) with a series of sources that decay primarily through the emission of a single γ ray and are thus not susceptible to summing. These sources included ^{141}Ce (145 keV), ^{139}Ce (165), ^{203}Hg (279), ^{51}Cr (320), ^{113}Sn (392), ^{85}Sr (514), ^{137}Cs (662), ^{95}Nb (766), ^{54}Mn (835), and ^{65}Zn (1115).

TABLE I. Cross sections [3] and decay parameters [5] of flux monitors

Flux monitor	E_γ(keV)	Branch	Half-life	Cross section (b)	Resonance integral (b)
$^{58}Fe \rightarrow {}^{59}Fe$	1099.3	56.5%	44.5 d	1.28 ± 0.05	1.7 ± 0.1
	1291.6	43.2%			
$^{59}Co \rightarrow {}^{60}Co$	1173.2	100%	5.27 y	37.18 ± 0.06	74 ± 2
	1332.5	100%			
$^{64}Zn \rightarrow {}^{65}Zn$	1115.5	50.6%	244.3 d	0.76 ± 0.02	1.45 ± 0.06
$^{94}Zr \rightarrow {}^{95}Zr$	724.2	44.2%	64.0 d	0.0499 ± 0.0024	0.23 ± 0.01
	756.7	54%			
$^{96}Zr \rightarrow {}^{97}Zr$	743.4	93%	16.9 h	0.0229 ± 0.0010	5.3 ± 0.3
$^{197}Au \rightarrow {}^{198}Au$	411.8	96%	2.70 d	98.65 ± 0.09	1550 ± 28

The partial decay scheme [5] of ^{103}Pd is illustrated in Fig. 1. The most intense γ rays are those at 357.4 (0.0221%) and 497.1 keV (0.0040%). The 40-keV isomeric transition is below the detection threshold of our counting system.

Areas of peaks in the γ spectrum were obtained using the Maestro software [6] to determine the number of counts above a linear background. Even though the γ spectrum had many peaks, those of interest in this experiment were sufficiently well isolated from their neighbors that it was not necessary to employ peak fitting to determine the areas.

III. RESULTS

The data reported in this work are derived from five irradiations of Pd samples: one in the ICIT, one in the CLICIT, and three in the TC. Of the three thermal column runs, two were done at the same location (hence at the same neutron flux) but one of these two was done using twice the irradiation time and four times the sample mass as the other. The Pd sample for this run was cut into two pieces that were stacked for the irradiation, and the activity of each piece was checked to verify that it scaled with its mass. This was done to eliminate the possibility that neutron absorption within the sample could be affecting the deduced cross sections. The third TC irradiation was done in a location with a 30% larger thermal flux.

Following each irradiation, the sample was put aside to "cool" for several days and then was counted continuously for at least three weeks; thereafter it was returned to the counting system for one day at periodic intervals of about one month to follow the decays for half-life determinations. Occasional counts were taken at larger source-to-detector distances to reduce the effect of coincidence summing in determining the activities. All counting rates were corrected for the decays during the counting intervals.

A sample γ-ray spectrum from an irradiated Pd sample is shown in Fig. 2. In addition to ^{103}Pd, other identifiable activities included those from capture by other Pd isotopes as well as those from capture by small impurities of other metals present in our sample. Impurity activities from capture by Pd were 13.7-h ^{109}Pd and 7.45-d ^{111}Ag (daughter of 23.4-min ^{111}Pd and 5.5-h ^{111m}Pd). Other capture-produced impurities with significant activities included 73.8-d ^{192}Ir, 19.3-h ^{194}Ir, 2.7-d ^{198}Au, and 3.14-d ^{199}Au (daughter of ^{199}Pt). We also observed radiations that we have identified with 4.35-d ^{101m}Rh, 35-h ^{105}Rh, and (less certainly) 207-d ^{102}Rh, all presumably formed through Pd(n,p) or Pd(n,d) reactions with fast neutrons (none of these activities were observed in the TC experiments).

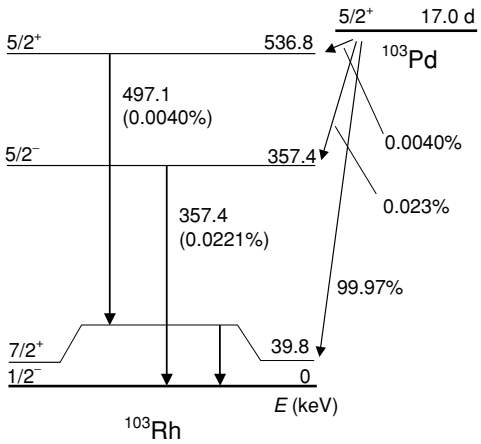

FIG. 1. Partial decay scheme of ^{103}Pd to ^{103}Rh. Level and γ-ray energies are given in keV.

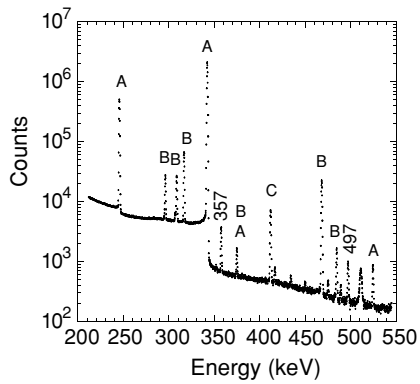

FIG. 2. Ge γ-ray spectrum of irradiated Pd sample. Impurity peaks are marked from ^{111}Ag (A), ^{192}Ir (B), and ^{198}Au (C).

Impurity identifications were checked to confirm consistency with accepted isotopic ratios, γ branching ratios, and decay half-lives. From this analysis, impurity levels in our samples were determined to be 0.5 ppm of Au, 5 ppm of Ir, and 200 ppm of Pt.

A. Flux monitors

To avoid difficulties with coincidence summing, flux monitors were counted at distances greater than 10 cm from the Ge detector. The results using various combinations of the flux monitors listed in Table I were cross checked for consistency in order to arrive at the best possible representation for the flux at each location.

For the CLICIT, which has an energy cutoff of about 0.5 eV, we assume that the thermal flux is negligible. The Au, Co, Zn, and ^{96}Zr flux monitors give a consistent epithermal flux of $(1.33 \pm 0.10) \times 10^{12}$ neutrons/cm^2/s, where the uncertainty represents the range of the four values (which is far larger than the statistical uncertainty). The Fe sample gives a smaller flux of about 1.0×10^{12} neutrons/cm^2/s, while the ^{94}Zr gives a larger value of 2.0×10^{12} neutrons/cm^2/s. The adopted resonance integral for ^{58}Fe given in Table I (1.7 ± 0.1 b) is from a measurement of Van der Linden *et al*. [7] and is the value currently listed on the NuDat website [3]. However, three other measurements give smaller values of 1.21 ± 0.06 b [8], 1.27 ± 0.07 b [9], and 1.17 b (no error given) [10]. If we choose instead a value of 1.2 b for the resonance integral of ^{58}Fe, the corresponding value of the epithermal flux would be 1.43×10^{12} neutrons/cm^2/s, which is in much better agreement with the other flux monitors.

For the ^{94}Zr, there is wide disagreement among the various measurements of the resonance integral, with values ranging [3] from the adopted 0.23 to 0.38 b. To obtain consistency with the other flux monitors, we would favor a value of 0.35 b for the ^{94}Zr resonance integral.

For the ICIT irradiations, after correcting for the epithermal contributions, five of the flux monitors give a consistent set of results corresponding to a thermal flux of $(7.8 \pm 0.7) \times 10^{12}$ neutrons/cm^2/s, where again the uncertainty represents the spread in the range of values. Only ^{96}Zr is unreliable for this purpose; because the resonance integral is so much larger than the thermal cross section, the uncertainty in the thermal flux due to the uncertainty in the resonance integral is larger than the thermal flux itself.

For the two locations used in the TC runs, we obtain thermal fluxes of 6.1×10^{10} (with epithermals of about 2×10^8) and 7.5×10^{10} neutrons/cm^2/s (epithermals of about 4×10^8). The epithermal contribution adds only a small correction to the deduced thermal cross sections. We estimate the uncertainty of these thermal flux values to be no more than 10%.

B. ^{102}Pd cross sections

In a sample with several activities present, confidence in the deduced cross sections would be enhanced if several γ rays from the decay of interest could be analyzed, in order to reduce the possibility that impurities might be contributing. The half-life of each transition can be determined to verify the radioisotope from which it originates. In the case of ^{103}Pd, only the transitions at 357.4 and 497.1 keV have sufficient intensity to be observable. The half-life of the 357.4-keV transition was consistently determined to be 17.0 d (within a range of ± 0.5 d), in excellent agreement with the accepted half-life of ^{103}Pd. However, the 497.1-keV transition consistently showed a longer half-life, typically about 30 d. Moreover, the intensity ratio of the 497.1 and 357.4 keV lines was larger in all samples than the value 0.179 expected from the known properties of the ^{103}Pd decay [5]. Values for this ratio were observed to vary between 0.3 and 1.0 depending on the age of the sample.

We therefore suspected an impurity contribution in the vicinity of 497 keV. A direct search for an impurity line near 497.1 keV was unsuccessful. None of the identified impurities has an emitted γ at that energy. The peak shape at 497 keV was consistent with a pure single line, suggesting that if an impurity were present its energy must agree with that of the ^{103}Pd decay energy to within 0.1 keV. However, the variation in the decay rate of the 497.1-keV line suggested two contributions to the activity. By subtracting the expected ^{103}Pd contribution from the total 497.1-keV line intensity, we were able to determine the half-life of the second component to be 39 ± 2 d. We conclude that the impurity line originates with the decay of 39.3-d ^{103}Ru, which populates the same excited states in ^{103}Rh as ^{103}Pd. Thus the energy of the impurity line from the ^{103}Ru decay is identical to that of the ^{103}Pd decay. The presence of this ^{103}Ru activity would lead us to conclude that a Ru impurity of 10 ppm was present in our Pd, but no other Ru activities could be observed in our samples to confirm the presence of this impurity.

Our determination of the ^{102}Pd cross sections is therefore based only on the analysis of the 357.4-keV γ transition emitted following the decay of ^{103}Pd. Because of the good agreement of the half-life of this line with that expected for ^{103}Pd (17.0 d), we are confident that this line represents only the decay of ^{103}Pd. To check the possibility of an impurity line contributing near 357 keV, we considered all radioactive isotopes with half-lives greater than 1 d and γ's within ± 1 keV of 357.4 keV. The on-line Table of Isotopes [5] lists 20 γ rays meeting these criteria. By analyzing our spectra to set upper limits on the intensities of other γ's emitted in these decays, we deduced that the total intensity of all such γ's can contribute no more than 0.5% to the intensity of our observed 357.4-keV peak. We are therefore confident that no impurity line can interfere with our measurement.

The deduced values of the ^{102}Pd cross sections from the various reactor irradiations are

$$\begin{aligned} \text{CLICIT}: &\quad I = 23 \pm 4\,\text{b}, \\ \text{ICIT}: &\quad \sigma = 2.06 \pm 0.20\,\text{b}, \\ \text{TC1}: &\quad \sigma = 1.49 \pm 0.20\,\text{b}, \\ \text{TC2}: &\quad \sigma = 1.96 \pm 0.20\,\text{b}, \\ \text{TC3}: &\quad \sigma = 1.77 \pm 0.20\,\text{b}. \end{aligned}$$

The listed uncertainties represent our best estimates based on possible systematic uncertainties in the flux determinations, detector efficiency calibrations, and detector summing

corrections. The statistical uncertainties of the counting process are a negligibly small component of these experimental uncertainties. The average value of the thermal cross section is

$$\sigma = 1.82 \pm 0.20\text{b}.$$

C. ^{108}Pd cross sections

Our samples also included γ rays from the decays of ^{109}Pd (13.7 h). Owing to the longer decay times between the end of neutron irradiation and the start of counting, this activity was too weak in samples TC1 and TC2 to produce analyzable data. For the analysis of the remaining samples, we used the 311-keV line (0.032%). The stronger 88-keV line cannot be used because it suffers from competition with Pb x rays; moreover, the detector efficiency calibration is rather uncertain in that energy region.

The deduced ^{108}Pd cross sections are

$$\begin{aligned} \text{CLICIT}: &\quad I = 122 \pm 18\ \text{b}, \\ \text{ICIT}: &\quad \sigma = 9.3 \pm 0.9\ \text{b}, \\ \text{TC3}: &\quad \sigma = 7.6 \pm 0.8\ \text{b}, \end{aligned}$$

with an average value for the thermal cross section of

$$\sigma = 8.5 \pm 0.9\ \text{b}.$$

These values actually represent the combined effect of captures leading directly to the ^{109}Pd ground state plus the effects of captures to the ^{109}Pd isomeric state followed by γ decay to the ground state. The necessary corrections to isolate the contributions from direct capture are very small: using the NuDat [3] recommended value of the isomeric thermal cross section ($\sigma = 0.183$ b) and the value of Van den Linden *et al.* [7] for the resonance integral leading to the isomeric state (2.26 b), we obtain finally

$$\sigma(\text{g}) = 8.3 \pm 0.9\ \text{b}, \qquad I(\text{g}) = 120 \pm 18\ \text{b}.$$

D. ^{110}Pd cross sections

Neutron capture by ^{110}Pd leads to ^{111}Pd (23.4 min) and $^{111\text{m}}$Pd (5.5 h). Both of these activities decay to ^{111}Ag (7.45 d). We were not able to observe the short-lived ^{111}Pd or $^{111\text{m}}$Pd decays, but all of our samples showed a strong presence of the 342 (6.68%) and 245 keV (1.33%) γ rays from the ^{111}Ag decay. Based on the currently accepted [3] cross sections of ^{110}Pd ($\sigma = 0.19$ b, $I = 2.4$ b) and $^{111\text{m}}$Pd ($\sigma = 0.037$ b, $I = 0.7$ b), our calculated values for the ^{111}Ag activities in all five experiments were too low by factors of 0.67–0.80. We suspect that the cause of this discrepancy lies in the ^{111}Pd or $^{111\text{m}}$Pd thermal cross sections or resonance integrals; but without further direct measurements, we are unable to identify which of the four values might be at fault. Moreover, we note that these currently accepted values are inconsistent with the measured value of the ratio of the ground-state and isomeric cross sections. Namboodiri *et al.* [11] measured $\sigma(\text{m})/\sigma(\text{g}) = 0.047 \pm 0.001$, while the presently accepted cross sections give a value of 0.19 ± 0.04 for this ratio.

IV. DISCUSSION

The difference between the present result for the ^{102}Pd thermal cross section (1.82 b) and the renormalized value (3.4 b) based on Meinke's work [2] may be explained by the failure of the latter to correct for the influence of resonance neutrons or perhaps, as pointed out by Meinke, uncertainties in the counting efficiency of the GM tube used to observe the emitted β's. Our results give overlapping values for the effective thermal cross section in measurements in the thermal column, where the effect of resonance neutrons is negligible, and in the reactor core, where the resonance neutrons contribute two-thirds of the activation leading to ^{103}Pd. Our accompanying measurements in the Cd-lined facility allow us to correct for the effects of these resonant captures.

Our value for the ^{108}Pd thermal cross section (8.3 b) agrees precisely with the recommended NuDat value. The individual reported experimental values (11.2 ± 2.2 [12], 9.3 ± 0.7 [13], 14 ± 2 [14], and 13.5 ± 1.4 b [15]) are all larger than our value, perhaps reflecting the need to correct for the significant effects of resonant capture. Our deduced value for the ^{108}Pd resonance integral (120 b) is half the recommended NuDat value (244 b), but it agrees with a previously measured [7] value (131 ± 73 b) if we renormalize the reported value using the present value of the thermal cross section.

Bishop *et al.* [16] measured the ratio of the isomeric and ground-state cross sections and resonance integrals. We can express their result as $\sigma(\text{m})/\sigma(\text{g}) = 0.0225 \pm 0.0042$, and also $I(\text{m})/I(\text{g}) = 0.0225 \pm 0.0042$. With the accepted NuDat value for the thermal cross section of the isomer (0.183 b) and the present value of the ground-state cross section, the ratio evaluates to

$$\frac{\sigma(\text{m})}{\sigma(\text{g})} = \frac{0.183\ \text{b}}{8.3\ \text{b}} = 0.0220,$$

with an uncertainty of about $\pm 20\%$. This is in excellent agreement with Bishop's value for the ratio. For the resonance integrals, using the measured value of $I(\text{m}) = 2.26$ b by Van den Linden *et al.* [7] and our value for the ground-state resonance integral, we have

$$\frac{I(\text{m})}{I(\text{g})} = \frac{2.26\ \text{b}}{120\ \text{b}} = 0.0188,$$

with an uncertainty of about $\pm 20\%$. This is likewise in good agreement with Bishop's value for the ratio.

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