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THERMAL NEUTRON CAPTURE CROSS SECTIONS OF THE PALLADIUM ISOTOPES

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Abstract. We have measured precise thermal neutron capture γ-ray cross sections σ_γ for all stable Palladium isotopes with the guided thermal neutron beam from the Budapest Reactor. The data were compared with other data from the literature and have been evaluated into the Evaluated Gamma-ray Activation File (EGAF)[1]. Total radiative neutron capture cross-sections σ_γ can be deduced from the sum of transition cross sections feeding the ground state of each isotope if the decay scheme is complete. The Palladium isotope decay schemes are incomplete, although transitions de-exciting low-lying levels are known for each isotope. We have performed Monte Carlo simulations of the Palladium thermal neutron capture de-excitation schemes using the computer code DICEBOX [2]. This program generates level schemes where levels below a critical energy E_{crit} are taken from experiment, and those above E_{crit} are calculated by a random discretization of an a priori known level density formula $\rho(E,J^\pi)$. Level de-excitation branching intensities are taken from experiment for levels below E_{crit} and the capture state, or calculated for levels above E_{crit} assuming an a priori photon strength function and applying allowed selection rules and a Porter-Thomas distribution of widths. The advantage of this method is that calculational uncertainties can be investigated systematically. Calculated feeding to levels below E_{crit} can then be normalized to the measured cross section deexciting those levels to determine the total radiative neutron cross-section σ_γ. In this paper we report the cross section measurements σ_γ[^{102}Pd(n,γ)]=0.9±0.3 b, σ_γ[^{104}Pd(n,γ)]=0.61±0.11 b, σ_γ[^{105}Pd(n,γ)]=21.1±1.5 b, σ_γ[^{106}Pd(n,γ)]=0.36±0.05 b, σ_γ[^{108}Pd(n,γ)(0)]=7.6±0.6 b, σ_γ[^{108}Pd(n,γ)(189)]=0.185±0.011 b, and σ_γ[^{110}Pd(n,γ)]=0.10±0.03 b. We have also determined from our statistical calculations that the neutron capture states in ^{107}Pd are best described as 2^+[59(4)%]+3^+[41(4)%]. Agreement with literature values was excellent in most cases. We found significant discrepancies between our results for ^{102}Pd and ^{110}Pd and earlier values that could be resolved by re-evaluation of the earlier results.

INTRODUCTION

Total radiative thermal neutron capture cross sections are usually determined by measuring either the neutron transmission rate through a target or the activation rate of radioactive product. Both methods require either knowledge of the neutron flux or use of a comparator material of well-known cross section. Transmission rates can be subject to significant corrections for neutron scattering and uncertainties in the target geometry. Activation measurements are usually more accurate, but they also require normalization to a comparator, typically gold, and they rely on the accuracy of the decay scheme normalization.

In this paper we report a new method for the determination of total radiative thermal neutron cross sections using prompt neutron capture γ-rays measured with guided neutron beams at the Budapest Reactor. Prompt γ-rays are measured with a high degree of sensitivity so that the cross section can be determined by both the total primary transition cross section deexciting the capture state and the total secondary transition cross section feeding the ground state. Complete decay schemes are available for very light isotopes, and complete measurements of secondary γ-ray cross sections feeding the ground state have been done for most isotopes of elements up to Fe. For heavier or low abundance isotopes, the measurements are generally incomplete.

For the more complex decay schemes of heavier elements, the contribution from unobserved continuum γ-rays feeding the ground state must be determined to obtain the total radiative cross section. In this paper we have used the Monte Carlo computer code DICEBOX [2] to calculate "complete" neutron capture decay schemes for the Palladium isotopes. These decay schemes are then normalized using measured transition cross sections deexciting low-lying levels in the Palladium isotopes to determine the total radiative cross sections.

EXPERIMENTAL

Neutron capture γ-ray cross sections for elemental targets with $Z = 1-84$, 90, 92 have been measured at the Budapest Reactor with the 2×10^6 n/s guided thermal neutron beam[3]. The target station is located far from the reactor where both primary and secondary γ-rays can be measured in low background conditions. Cross sections were measured using either stoichiometric compounds or accurately prepared mixtures containing the standard elements H, N, or Cl whose γ-ray cross sections are precisely known. The γ-ray cross sections were then accurately determined from their intensity relative to the standard γ-ray transitions of the comparators.

An elemental target consisting of 1.6 g of $PdCl_2$ with a thickness of 0.4 g/cm^2 was irradiated in the 2×10^6 n$\cdot s^{-1}$cm^{-2} guided thermal neutron beam at Budapest Reactor for 8753 seconds. Prompt gamma-rays from the target were detected with a 25% efficient, Compton-suppressed, HPGe detector. Counting efficiency was determined over the range of 50 keV to 10 MeV with radioactive sources and (n,γ) reaction gamma rays to a precision of better that 1% from 500 keV to 6 MeV and better than 3% at all energies [4]. The γ-ray spectra were analyzed using the Hypermet PC program [5]. A total of 202 γ-rays were assigned to the six Palladium isotopes ^{103}Pd, ^{105}Pd, ^{106}Pd, ^{107}Pd, ^{109}Pd, and ^{111}Pd on the basis of energy and intensity by comparison with data from the ENSDF [6] file. The γ-ray cross section data were sufficient to determine level deexcitation cross sections, *σ(level)*, for 101 Palladium levels including at least one level from each isotope. These results are summarized in Table 1.

DICEBOX CALCULATIONS

Theoretical statistical feedings to low-lying levels have been calculated using the computer code DICEBOX [2]. This code determines the theoretical uncertainty in the level feedings due to statistical fluctuations using an algorithm based on the extreme statistical model of nuclei. Below a critical energy E_{crit} the level scheme, i.e. energies, spins and parities of all levels and all depopulating transitions, is assumed to be known from experiment. Above this energy, an unknown set of levels is determined by a random discretization of a level density formula $\rho(E,J^\pi)$. The partial radiative width $\Gamma_{a\gamma b}$ for γ-ray decay with an energy E_γ from a level a to a level b is assumed to vary with a Porter-Thomas distribution whose mean value is determined by the level density and a photon strength function $S(E_\gamma)$ given by

$$\Gamma_{a\gamma b} = \frac{S(E_\gamma)\times E_\gamma^3}{\rho(E_a,J_a^\pi)}, \tag{1}$$

where $\rho(E_a,J_a^\pi)$ is the level density at initial level a. Two choices of level density models are available with DICEBOX. The constant temperature model (CTF) [7] is given by

$$\rho(E_a,J_a^\pi) = f_J\frac{1}{T}e^{(E-E_0)/T} \tag{2}$$

where T is the nuclear temperature and E_0 is the backshift. The back-shifted Fermi gas model (BSFG) [7,8,9] is given by

$$\rho(E_a,J_a^\pi) = \frac{f_J e^{2\sqrt{a(E-E_1)}}}{12\sqrt{2}\sigma a^{\frac{1}{4}}(E-E_1)^{\frac{5}{4}}} \tag{3}$$

where a is the conventional shell-model level-density parameter and E_1 is another back shift. The factor f_J represents the probability that a randomly chosen level has spin J [7] and is given by

$$f_J = \frac{2J+1}{2\sigma^2}e^{-(J+\frac{1}{2})^2/2\sigma^2} \tag{4}$$

where $\sigma = 0.98A^{0.29}$ for the CTF model and $\sigma = 0.298A^{1/3}a^{1/4}(E-E_1)^{1/4}$ for the BSFG model. The parameters for both the CTF and BSFG models are determined from the known levels below E_{crit} and resonance data for levels above the neutron capture state. The CTF and BSFG models gave very similar results in these calculations.

The photon strength function $S^{XL}(E_\gamma)$ for multipolarities XL is assumed to be independent of spin and parity and is defined as

$$S^{XL}(E_\gamma) = \frac{1}{E_\gamma^{2L+1}}\frac{\langle\Gamma_{ab}^{(J)}\rangle_a}{\langle D_J\rangle} \tag{5}$$

Here $\langle\Gamma_{ab}^{(J)}\rangle_a$ represents the value of the partial radiation width $\Gamma_{ab}^{(J)}$ for a transition $a\rightarrow b$, averaged over initial levels a with a fixed parity π and spin J. D_J is the average spacing between initial levels. Only the lowest-order multipolarities E1, M1, and E2 are included in these calculations. Numerous options for calculating $S^{XL}(E_\gamma)$ including single-particle, Axel and Brink [10], Kopecky and Chrien [11], Kadmenskij *et al* [12], and various other semi-empirical models are available in DICEBOX. The choice of model did not significantly affect our results.

TABLE 1. Experimental neutron capture cross sections feeding excited states in Palladium isotopes. Ground state feedings for ^{106,109}Pd were determined from experimental and theoretical side feedings to all levels below E_{crit} as described in the text.

Isotope	E(Level) (keV)	J$^{\pi}$	σ(level) (barns)	E$_{crit}$ (keV)	Feeding (%)	σ_{γ}(This work) (barns)	σ_{γ}(Literature)* (barns)
^{103}Pd	118.736(17)	3/2+	0.51(14)	500	58(8)	0.9(3)	1.82(20)†
^{105}Pd	280.51(22)	3/2+	0.145(13)	680	30(8)	0.48(14)	
	306.25(3)	7/2+	0.040(8)		3.9(14)	1.02(42)	
	344.512(18)	1/2+	0.099(18)		15(5)	0.66(31)	
	560.75(3)	3/2+	0.050(10)		6.5(15)	0.77(24)	
	644.53(4)	7/2-	0.063(6)		3.8(20)	1.7(9)	
	Average					0.61(11)	0.6(3)
^{106}Pd	0	0+	20.0(3)	2505	94.8(15)	21.1(15)	21.0(15)
^{107}Pd	115.74(12)	1/2+	0.095(9)	480	28(7)	0.34(9)	
	302.78(15)	5/2+	0.046(4)		8.5(30)	0.54(20)	
	312.20(10)	7/2+	0.024(4)		3.2(22)	0.8(5)	
	381.80(13)	3/2+	0.043(6)		11(2)	0.39(9)	
	471.21(24)	(3/2)+	0.024(5)		8(2)	0.30(10)	
	Average					0.36(5)	0.29(3)
^{109}Pd	0	5/2+	5.93(8)	350	78(6)	7.6(6)	7.6(4)
	188.990(10)	11/2-	0.185(11)		100	0.185(11)	0.18(3)
^{111}Pd	191.3(3)	[3/2]+	0.016(4)	200	18(5)	0.09(3)	
	195.1(2)	[3/2]+	0.019(8)		18(4)	0.11(5)	
	Average					0.10(3)	0.19(3)

* from reference [13] except where noted
† from reference [14]

DICEBOX constructs a complete, artificial decay scheme and then randomly simulates γ-ray cascades. Selection rules for different types of transitions are taken into account. One unique choice of a level scheme and all partial radiative widths is termed a nuclear realization. As there are an infinite number of possible nuclear realizations allowed by these assumptions, the simulated population of low-lying levels is subject to fluctuations. If the intensities of primary transitions to low-lying levels are known, as in the case of ^{106}Pd and ^{109}Pd, they are taken from experiment. Otherwise they are simulated and their values will differ from realization to realization.

The fraction of the total cross section feeding the observed levels in the Palladium isotopes assuming the indicated values of E$_{crit}$ are given in Table 1. The uncertainties in these values were determined from the variations in the multiple realizations of each decay scheme. Complete γ-ray data were available for all levels below E_{crit} in ^{106}Pd and ^{109}Pd. For these cases we normalized the calculation by performing a least-squares fit of the calculated side feeding to each level from levels above E_{crit} to the experimental side feeding to each level determined by the intensity balance, $\Sigma\sigma_{\gamma}(in) - \Sigma\sigma_{\gamma}(out)$, from levels below E_{crit}. The total feedings to the ground states of ^{106}Pd and ^{109}Pd, calculated from this normalization, are given in Table 1. The total radiative neutron capture cross sections σ_{γ} in Table 1 are determined simply by the ratio 100 × σ(level)/%Feeding.

RESULTS

The total radiative neutron capture cross sections for the Palladium isotopes derived from the experimental cross sections, $I_{\gamma+e}$, feeding the levels and the relative branching intensities calculated with DICEBOX feeding these levels is shown in Table 1. For ^{102}Pd(n,γ) we measured σ_{γ}=0.9±0.3 b based on the de-excitation cross section of the first excited state in ^{103}Pd. This value is considerably lower than σ_{γ}=1.82±0.20 b from Duncan and Krane [14]. That value was measured assuming P$_{357.4\gamma}$=0.0221±0.0007% for ^{103}Pd decay from reference [15]. A previous measurement by Zoller *et al* [16] gave P$_{357.4\gamma}$=0.0324±0.0016% leading to σ_{γ}=1.24±0.15 b which is in better agreement with our value.

For ^{104}Pd(n,γ) we determined that σ_{γ}=0.61±0.11 b based on the average population of five levels in ^{105}Pd. This value is in good agreement with σ_{γ}=0.6±0.3 b from Mughabghab *et al* [13]. Similarly, For ^{106}Pd(n,γ) we determined that σ_γ=0.36±0.05 b from the average population of five levels in ^{107}Pd in agreement with σ_γ=0.29±0.03 b from Ref. 13. The experimental value σ_γ=0.10±0.03 b for ^{110}Pd(n,γ), based on the observation of two levels in ^{111}Pd, is lower than the compiled value of σ_γ=0.19±0.03 b which is from Sehgal *et al* [17]. Comparison of other cross sections measured in Ref. 17 indicates that they were typically about twice as large as the values compiled in Ref. 13. Presumably this discrepancy resulted from an imprecise determination of the neutron flux in Ref. 17. The Sehgal *et al* value can be renormalized by a factor of 0.54, necessary to correct their measurement of 14±2 b for the ^{108}Pd(n,γ) cross section, yielding σ_γ=0.10±0.02 b, which agrees with our result.

The values σ_γ=21.1±1.5 b for ^{105}Pd(n,γ) and σ_γ=7.6±0.6 b for ^{108}Pd(n,γ) were determined using data for many levels as described above. They are in excellent agreement with the compiled values from Ref. 13. In addition, a new value has been determined for the cross section feeding the 4.7 min isomer in ^{109}Pd σ_γ[^{108}Pd(n,γ)(189)]=0.185±0.011 b, determined by measuring the 188.99-keV γ-ray cross section in equilibrium with the prompt transitions. Calculation of ^{106}Pd statistical feeding was complicated by the unknown relative contributions of capture states with spins 2^+, and 3^+. Figures 1 and 2 show comparisons of the experimental and statistical side feedings for each J^π assignment. Agreement between experiment and theory is poor in both cases. We then solved for the mixing fraction of capture state spins that minimizes the χ^2/f comparison between experiment and theory and determined that 2^+[59(4)%]+3^+[41(4)%] gave an excellent fit to the experimental and theoretical side feedings, as is shown in Figure 3. The good agreement between experimental and statistical side feedings for ^{109}Pd, where the capture state is uniquely $1/2^+$, is shown in Figure 4.

CONCLUSIONS

New measurements of the total thermal neutron radiative cross sections for ^{102,104,105,106,108,110}Pd(n,γ) have been performed using prompt γ-ray cross sections to renormalize statistical model calculations using DICEBOX. These results had comparable precision to previous values. Our measurements provide independent confirmation of previous results and can be used to identify and resolve problems with earlier experiments. A large library of prompt neutron capture γ-ray data has been measured for all other stable elements and will be analyzed in the future to determine additional total radiative cross sections.

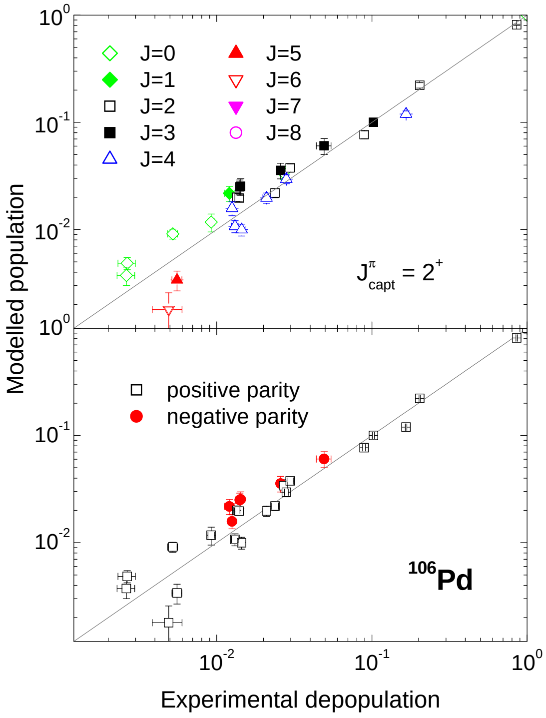

FIGURE 1. Experimental and statistical side feedings for ^{105}Pd(n,γ) assuming the capture state $J^\pi=2^+$.

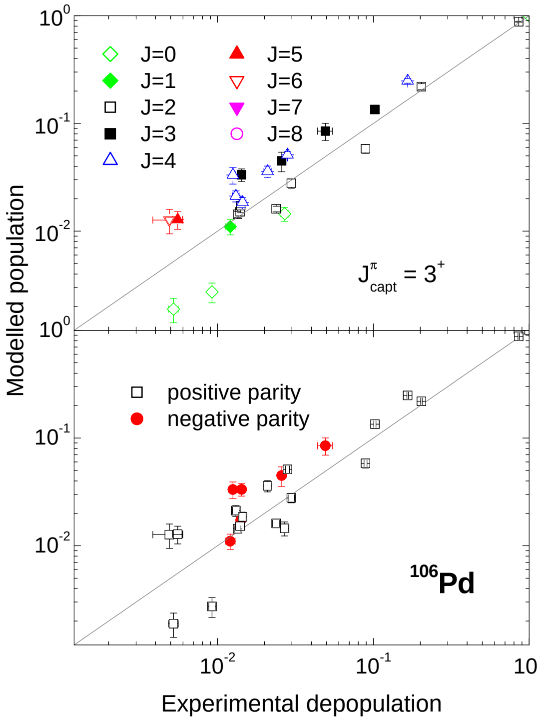

FIGURE 2. Experimental and statistical side feedings for ^{105}Pd(n,γ) assuming the capture state $J^\pi=3^+$.

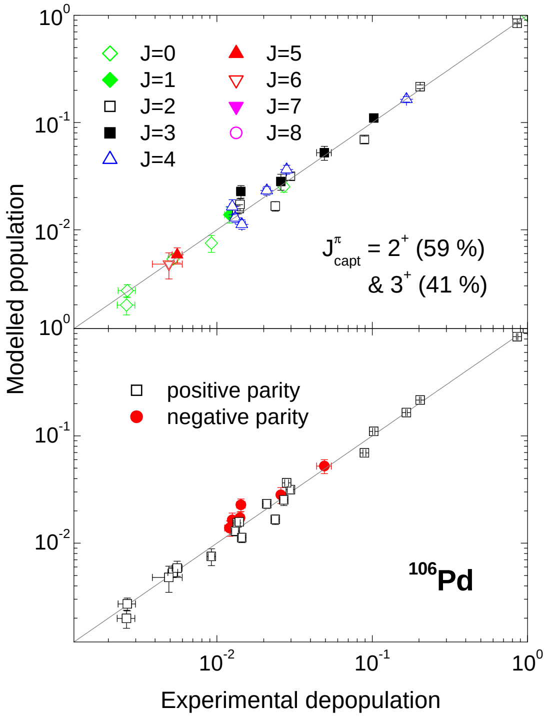

FIGURE 3. Experimental and statistical side feedings for ^{105}Pd(n,γ) assuming the capture state is 2^+[59(4)%]+3^+[41(4)%]

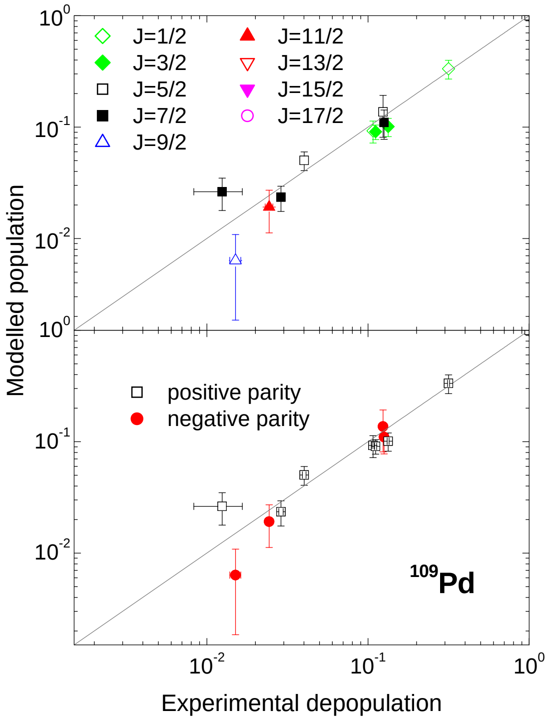

FIGURE 4. Experimental and statistical side feedings for ^{108}Pd(n,γ).

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