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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 σγ 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 σγ 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 Ecrit are taken from experiment, and those above Ecrit are calculated by a random discretization of an a priori known level density formula ρ(E,Jk). Level de-excitation branching intensities are taken from experiment for levels below Ecrit and the capture state, or calculated for levels above Ecrit 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 Ecrit can then be normalized to the measured cross section deexciting those levels to determine the total radiative neutron cross-section σγ. In this paper we report the cross section measurements σγ[102]Pd(n,γ)=0.9±0.3 b, σγ[104]Pd(n,γ)=0.61±0.11 b, σγ[105]Pd(n,γ)=21.1±1.5 b, σγ[106]Pd(n,γ)=0.36±0.05 b, σγ[108]Pd(n,γ)(0)=7.6±0.6 b, σγ[108]Pd(n,γ)(189)=0.185±0.011 b, and σγ[110]Pd(n,γ)=0.10±0.03 b. We have also determined from our statistical calculations that the neutron capture states in 107Pd 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 102Pd and 110Pd 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 de-
exciting low-lying levels in the Palladium isotopes to de-
terminate the total radiative cross sections.

**EXPERIMENTAL**

Neutron capture γ-ray cross sections for elemental tar-
gets with $Z = 1 - 84$, 90, 92 have been measured at the
Budapest Reactor with the $2 \times 10^6$ 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 sec-
tions were measured using either stoichiometric com-
ounds 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 PdCl₂ with
a thickness of 0.4 g/cm² was irradiated in the $2 \times 10^6$
n·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 de-
served over the range of 50 keV to 10 MeV with ra-
dioactive sources and (n,γ) reaction gamma rays to a pre-
cision of better than 1% from 500 keV to 6 MeV and bet-
ter 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 ba-
sis 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,
$\sigma$(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 al-
gorithm based on the extreme statistical model of nuclei.
Below a critical energy $E_{\text{crit}}$ the level scheme, i.e. en-
ergies, spins and parities of all levels and all depopu-
lating transitions, is assumed to be known from exper-
iment. 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 $\Gamma_{\alpha\beta}$ for γ-
ray decay with an energy $E_\gamma$ 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

$$\Gamma_{\alpha\beta} = \frac{S(E_\gamma) \times E_\gamma^3}{\rho(E_\gamma,J^\pi_\beta)}, \quad (1)$$

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

$$\rho(E_\alpha,J^\pi_\alpha) = \frac{f_J}{T} e^{(E-E_0)/T} \quad (2)$$

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

$$\rho(E_\alpha,J^\pi_\alpha) = \frac{f_J e^{2\sqrt{aE-E_\gamma}}}{12\sqrt{2}a\pi^2 (E-E_\gamma)^{5/2}} \quad (3)$$

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

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

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 pa-
rameters for both the CTF and BSFG models are deter-
mined from the known levels below $E_{\text{crit}}$ and resonance
data for levels above the neutron capture state. The CTF
and BSFG models gave very similar results in these cal-
culations.

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

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

Here $\langle \Gamma_{ab} \rangle_a$ represents the value of the partial radiation
width $\Gamma_{ab}$ for a transition $a \rightarrow b$, averaged over initial
levels $a$ with a fixed parity $\pi$ and spin $J$. $D_J$ is the aver-
age spacing between initial levels. Only the lowest-order
multipolarities E1, M1, and E2 are included in these cal-
culations. Numerous options for calculating $S^{XL}(E_\gamma)$ in-
cluding 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 $E_{\text{crit}}$ as described in the text.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$E$(Level) (keV)</th>
<th>J$^\pi$</th>
<th>$\sigma$(level) (barns)</th>
<th>$E_{\text{crit}}$ (keV)</th>
<th>Feeding (%)</th>
<th>$\sigma_\gamma$(This work) (barns)</th>
<th>$\sigma_\gamma$(Literature)$^*$ (barns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{103}$Pd</td>
<td>118.736(17)</td>
<td>3/2+</td>
<td>0.51(14)</td>
<td>500</td>
<td>58(8)</td>
<td>0.9(3)</td>
<td>1.82(20)†</td>
</tr>
<tr>
<td>$^{105}$Pd</td>
<td>280.51(22)</td>
<td>3/2+</td>
<td>0.145(13)</td>
<td>680</td>
<td>30(8)</td>
<td>0.48(14)</td>
<td>1.02(42)</td>
</tr>
<tr>
<td></td>
<td>306.25(3)</td>
<td>7/2+</td>
<td>0.040(8)</td>
<td>3.9(14)</td>
<td>15(5)</td>
<td>0.66(31)</td>
<td>3.9(14)</td>
</tr>
<tr>
<td></td>
<td>344.512(18)</td>
<td>1/2+</td>
<td>0.099(18)</td>
<td>6.5(15)</td>
<td>7/2+</td>
<td>0.040(8)</td>
<td>3.9(14)</td>
</tr>
<tr>
<td></td>
<td>560.75(3)</td>
<td>3/2+</td>
<td>0.050(10)</td>
<td>3.8(20)</td>
<td>7/2-</td>
<td>0.063(6)</td>
<td>0.6(3)</td>
</tr>
<tr>
<td></td>
<td>644.53(4)</td>
<td>7/2-</td>
<td>0.099(18)</td>
<td>6.5(15)</td>
<td>7/2-</td>
<td>0.063(6)</td>
<td>0.6(3)</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td></td>
<td>0.61(11)</td>
<td></td>
<td></td>
<td>0.36(5)</td>
<td>0.29(3)</td>
</tr>
<tr>
<td>$^{106}$Pd</td>
<td>0</td>
<td>0+</td>
<td>20.0(3)</td>
<td>2505</td>
<td>94.8(15)</td>
<td>21.1(15)</td>
<td>21.0(15)</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>115.74(12)</td>
<td>1/2+</td>
<td>0.095(9)</td>
<td>480</td>
<td>28(7)</td>
<td>0.34(9)</td>
<td>0.34(9)</td>
</tr>
<tr>
<td></td>
<td>302.78(15)</td>
<td>5/2+</td>
<td>0.046(4)</td>
<td>8.5(30)</td>
<td>5/2+</td>
<td>0.046(4)</td>
<td>8.5(30)</td>
</tr>
<tr>
<td></td>
<td>312.20(10)</td>
<td>7/2+</td>
<td>0.024(4)</td>
<td>3.2(22)</td>
<td>7/2+</td>
<td>0.024(4)</td>
<td>3.2(22)</td>
</tr>
<tr>
<td></td>
<td>381.80(13)</td>
<td>3/2+</td>
<td>0.043(6)</td>
<td>11.2</td>
<td>7/2+</td>
<td>0.043(6)</td>
<td>11.2</td>
</tr>
<tr>
<td></td>
<td>471.21(24)</td>
<td>(3/2)+</td>
<td>0.024(5)</td>
<td>8(2)</td>
<td>7/2-</td>
<td>0.063(6)</td>
<td>0.6(3)</td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td></td>
<td>0.36(5)</td>
<td></td>
<td></td>
<td>0.29(3)</td>
<td></td>
</tr>
<tr>
<td>$^{109}$Pd</td>
<td>0</td>
<td>5/2+</td>
<td>5.93(8)</td>
<td>350</td>
<td>78(6)</td>
<td>7.6(6)</td>
<td>7.6(4)</td>
</tr>
<tr>
<td></td>
<td>188.990(10)</td>
<td>11/2-</td>
<td>0.185(11)</td>
<td>100</td>
<td>11/2-</td>
<td>0.185(11)</td>
<td>0.18(3)</td>
</tr>
<tr>
<td>$^{111}$Pd</td>
<td>191.3(3)</td>
<td>[3/2]+</td>
<td>0.016(4)</td>
<td>200</td>
<td>18(5)</td>
<td>0.09(3)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>195.1(2)</td>
<td>[3/2]+</td>
<td>0.019(8)</td>
<td>18(4)</td>
<td>18(5)</td>
<td>0.11(5)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Average</td>
<td></td>
<td>0.10(3)</td>
<td></td>
<td></td>
<td>0.19(3)</td>
<td></td>
</tr>
</tbody>
</table>

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

DICEBOX constructs a complete, artificial decay scheme and then randomly simulates $\gamma$-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 $^{106,109}$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 $E_{\text{crit}}$ are given in Table 1. The uncertainties in these values were determined from the variations in the multiple realizations of each decay scheme. Complete $\gamma$-ray data were available for all levels below $E_{\text{crit}}$ in $^{108,109}$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 $E_{\text{crit}}$ to the experimental side feeding to each level determined by the intensity balance, $\Sigma \sigma_\gamma$(in) $- \Sigma \sigma_\gamma$(out), from levels below $E_{\text{crit}}$. The total feedings to the ground states of $^{106,109}$Pd and $^{109}$Pd, calculated from this normalization, are given in Table 1. The total radiative neutron capture cross sections $\sigma_\gamma$ in Table 1 are determined simply by the ratio $100 \times \sigma$(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,$\gamma$) we measured $\sigma_\gamma=0.9\pm0.3$ b based on the de-excitation cross section of the first excited state in $^{103}$Pd. This value is considerably lower than $\sigma_\gamma=1.82\pm0.20$ b from Duncan and Krane [14]. That value was measured assuming $P_{357.4\gamma}=0.0221\pm0.0007\%$ for $^{103}$Pd decay from reference [15]. A previous measurement by Zoller et al [16] gave $P_{357.4\gamma}=0.0324\pm0.0016\%$ leading to $\sigma_\gamma=1.24\pm0.15$ b which is in better agreement with our value.

For $^{104}$Pd(n,$\gamma$) we determined that $\sigma_\gamma=0.61\pm0.11$ b based on the average population of five levels in $^{105}$Pd. This value is in good agreement with $\sigma_\gamma=0.6\pm0.3$ 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]. Comparision 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 $^{109}$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 $σ_γ^{109}$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 $J^\pi$ 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 $\chi^2/ν$ 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.
ACKNOWLEDGEMENTS

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REFERENCES

6. Evaluated Nuclear Structure Data File, a computer file of evaluated experimental nuclear structure data maintained by the National Nuclear Data Center, Brookhaven National Laboratory.