Measurement of neutron induced reaction cross sections of palladium isotopes at the neutron energy of 14.54 ± 0.24 MeV with covariance analysis

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Abstract

The $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross sections have been measured with respect to the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ monitor reaction at the neutron energy of 14.54 ± 0.24 MeV by using the method of activation and off-line γ -ray spectrometry. The mono-energetic neutron beam was generated from the D–T reaction. The uncertainties in the various basic nuclear parameters in the measured reactions and their correlations were estimated by using covariance analysis. The present data were compared with the EXFOR based literature data, evaluated data of various libraries available in national nuclear data center and with the calculated values from TALYS-1.9 code.

Keywords $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$ · $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$ · $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reactions · D–T reaction neutron source · Activation and off-line γ -ray spectrometry · Covariance analysis · TALYS-1.9

Introduction

Neutron activated cross sections are the essential data that plays crucial role in structural information of atomic nuclei, safety design of nuclear reactors, accelerator-driven sub-critical system (ADSs), nuclear level densities, astrophysics, medical isotopes, nuclear waste transmutation system, nuclear science and technologies [1, 2]. The palladium catalyst is used for the transformation of long-lived radioactive

waste into a useful element and to convert the toxic into inert substances in engine exhaust [3]. The radionuclide ^{105}Rh treated as a low energy beta (β^-) emitter and thus is used in diagnosis studies. Due to its good physical nature, it is used for therapy in small tumors [4]. Palladium isotopes are important to study nucleosynthesis in stellar environment and early phases of galactic evolution [5].

The $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$ [6–16], $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$ [9, 10, 13–15, 17–21], $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ [14, 15, 17, 21] and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ [14, 15, 17, 20, 21] reactions cross sections at the neutron energy of 14 MeV are present in literature, which are compiled in EXFOR [22, 23]. However, the experiment data from literature [7–18] have disagreement, which is most probably due to various nuclear parameters like half-life, γ -ray abundances, monitor reaction and types of detectors used. Thus the neutron induced cross sections data for $^{\text{nat}}\text{Pd}$ are not very much precise for the applications and needed to re-measure. Besides this, the literature data do not have information about covariance matrix to estimate uncertainties.

In view of the above facts, we have measured the $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reactions cross sections at the neutron energy of 14.54 ± 0.24 MeV by using the method of activation and off-line γ -ray spectrometry. The measured reactions

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cross sections of palladium isotopes were compared with the EXFOR [22, 23] based literature data, evaluated data of various libraries from national nuclear data center (NNDC) and the calculated values from TALYS-1.9 [24, 25] with default parameters.

Experimental details

Neutron generator

The Purnima neutron generator at Bhabha Atomic Research Center (BARC), Mumbai was used to performed the present experiment based on the $^3\text{H}(\text{d},\text{n})^4\text{He}$ reaction neutron source. The D^+ ion beam from a RF ion source generates are collected and focused. The focused D^+ ions were accelerated and incident on a titanium–tritium (TiT) target to produce the neutron beam at zero degree angle [26]. In the current measurement, the D^+ ion were accelerated to 180 kV. The details of neutron energy calculations using GEANT4 are given in refs.[27–30]. A deuterium beam having energy of 180 ± 0.1 keV, with envelope diameter of 0.47 cm, measured using a beam profile monitor on TiT target is simulated in GEANT4. The neutrons hitting on the sample is returned and binned into histogram using root. The histogram is further normalized to the total number of counts to get the spectrum. The neutron energy spread is calculated from the standard deviation (FWHM/centroid). The obtained neutron energy from GEANT4 simulations is 14.54 ± 0.24 MeV.

Preparation of sample and irradiation

The $^{\text{nat}}\text{Pd}$ and ^{197}Au foils with purity > 99.9% from Alfa Aesar, USA were used in the present experiment. The weights of $^{\text{nat}}\text{Pd}$ and ^{197}Au metal foils are 112.8 mg and 126.0 mg, respectively. The sizes of each foils are $1.1 \text{ cm} \times 1.1 \text{ cm}$. The thicknesses of $^{\text{nat}}\text{Pd}$ and ^{197}Au foils are 0.1 mm and 0.025 mm, respectively. The foils were wrapped individually with 0.011 mm thick Al foil to prevent radioactive contamination from one another during time of activation. The stack of Pd–Au samples were mounted at zero degrees angle with respect to the neutron beam direction and irradiated for 2 h.

Gamma-ray counting

After the irradiation, the activated $^{\text{nat}}\text{Pd}$ and ^{197}Au metal samples wrapped with the Al foil were taken out from the irradiation assembly and mounted on different Perspex plates. Then the γ -ray spectrometry of the activated samples

were done by using a pre-calibrated 185-cc Baltic HPGe detector coupled to a PC-based 4096 channel analyzer. The resolution of the HPGe detector had a FWHM of 1.8 keV at the 1332.5 keV γ -ray photo-peak of ^{60}Co . A standard ^{152}Eu source was used for the energy calibration of the HPGe detector. The data acquisition was performed using CAMAC-Linux based software. The γ -ray counting of the irradiated sample was done by placing the mounted samples at a suitable distance from the end cap of the detector to keep the dead time less than 5% and to avoid the coincidence summation effect. The γ -ray counting of the standard source for calibration was also done in the same geometry.

Data analysis and results

Efficiency calibration of HPGe detector with uncertainty

The efficiency calibration of the HPGe detector was performed by using the characteristic γ -ray energies of standard ^{152}Eu and ^{133}Ba sources with the half-life ($T_{1/2}$) of 13.517 ± 0.014 y and 10.551 ± 0.011 y, respectively. The elapsed time (t) between the dates of manufacture of ^{152}Eu and ^{133}Ba to the time of detector calibration are 19.701 y and 4.515 y, respectively. The activity (A_0) of ^{152}Eu was 5036.89 ± 71 Bq as on 1 October 1999 and for ^{133}Ba was $60,000 \pm 244$ Bq as on 15 November 2014. The efficiency of HPGe detector was calculated by using the following expression,

$$\varepsilon(E_\gamma) = \frac{CK_C}{I_\gamma A_0 e^{-\ln(2)t/T_{1/2}}} \quad (1)$$

where $\varepsilon(E_\gamma)$ is the efficiency of the respective γ -ray energies (E_γ) of ^{152}Eu and ^{133}Ba standard sources. The counting time (t) of the detected γ -ray counts (C) of ^{152}Eu and ^{133}Ba are 2691 and 98 s, respectively. The basic nuclear data like half-life and γ -ray branching ratio (I_γ) for each γ -ray energies of ^{152}Eu and ^{133}Ba were retrieved from NuDat 2.7 [31–33]. The coincidence summing correction factor, K_C was simulated using EFFTRAN code [34]. The data given in Table 1 are then substituted in Eq. (1) to determine the efficiency $\varepsilon(E_\gamma)$ at each identified γ -ray energy of the ^{152}Eu and ^{133}Ba , which are presented in column 5 of Table 1.

The related covariance uncertainty matrix V_ε was estimated considering uncertainties in the four attributes C , I_γ , A_0 and λ as well as their correlation. The details of calculations to find covariance matrix is given in refs.[35, 36]. The γ -ray energies of ^{152}Eu and ^{133}Ba sources are different

Table 1 Efficiency calibration of HPGe detector using the γ -ray energies of ^{152}Eu and ^{133}Ba standard sources with uncertainties

E_γ (keV)	I_γ (%)	C	K_c	$\epsilon(E_\gamma)$
80.99	32.9 ± 0.3	$50,045 \pm 2635$	1.32	$4.62\text{E}-02$
121.8	28.53 ± 0.16	$137,393 \pm 4279$	1.24	$1.20\text{E}-01$
244.7	7.55 ± 0.04	$24,329 \pm 590$	1.35	$8.80\text{E}-02$
411.1	2.827 ± 0.014	4273 ± 155	1.41	$5.43\text{E}-02$
778.9	12.93 ± 0.08	$15,996 \pm 326$	1.23	$3.08\text{E}-02$
867.4	4.23 ± 0.03	4123 ± 121	1.42	$2.81\text{E}-02$
964.1	14.51 ± 0.07	$15,494 \pm 302$	1.16	$2.51\text{E}-02$
1085.8	10.11 ± 0.05	$10,940 \pm 334$	0.90	$1.97\text{E}-02$
1112.1	13.67 ± 0.08	$12,315 \pm 179$	1.09	$1.98\text{E}-02$
1212.9	1.415 ± 0.008	916 ± 96	1.41	$1.85\text{E}-02$
1299.1	1.633 ± 0.011	1240 ± 68	1.23	$1.89\text{E}-02$
1408.0	20.87 ± 0.09	$16,497 \pm 366$	1.12	$1.79\text{E}-02$

Fig. 1 Plot of fitted efficiency using standard ^{152}Eu and ^{133}Ba sources

from the characteristic γ -ray energies of the reaction products ^{109}Pd , ^{101}Pd , ^{105}Rh , $^{106\text{m}}\text{Rh}$ and ^{196}Au . To estimate the efficiency of the γ -ray energies corresponding to the radionuclide's ^{109}Pd , ^{101}Pd , ^{105}Rh , $^{106\text{m}}\text{Rh}$ and ^{196}Au , the following linear parametric function has been considered.

$$\ln(\epsilon_i) = \sum_{k=1}^m p_k (\ln[E_i])^{k-1} \quad 1 \leq i \leq 12 \quad (2)$$

The best quality of fit was achieved for $m=8$, with $\frac{\chi^2}{12-8} = 1.76 \approx 1$ and was used for corresponding linear parametric model.

$$\ln \epsilon = -3.75 - 1.46 \ln E - 0.89(\ln E)^2 + 3.66(\ln E)^3 + 10.7(\ln E)^4 + 9.97(\ln E)^5 + 3.94(\ln E)^6 + 0.57(\ln E)^7 \quad (3)$$

From Eq. (3), we estimated the efficiencies corresponding to the γ -ray energies of the reaction products ^{109}Pd , ^{101}Pd , ^{105}Rh and $^{106\text{m}}\text{Rh}$ and ^{196}Au . The fitted efficiencies are shown in the Fig. 1. In Table 2, the estimated detector efficiencies of the γ -ray energies of the corresponding reaction products ^{109}Pd , ^{101}Pd , ^{105}Rh and $^{106\text{m}}\text{Rh}$ and ^{196}Au with correlations among them are presented. The efficiencies obtained are presented in Table 2, which were used to estimate the cross sections. The details of calculations are given in refs.[37, 38].

Estimation of $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross-sections with uncertainty

We calculated the $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross sections at the neutron energy of 14.54 ± 0.24 MeV. The reaction cross sections were determined using standard neutron activation ratio formula

$$\sigma_{\text{Pd}} = \sigma_{\text{Au}} \frac{C_{\text{Pd}} \lambda_{\text{Pd}} W_{\text{tAu}} a_{\text{Au}} A_{\text{VAu}} I_{\gamma\text{Au}} \epsilon(E_{\gamma})_{\text{Au}} (1 - e^{-\lambda_{\text{tAu}}}) (e^{-\lambda_{\text{tAu}}}) (1 - e^{-\lambda_{\text{tAu}}})}{C_{\text{Au}} \lambda_{\text{Au}} W_{\text{tPd}} a_{\text{Pd}} A_{\text{VPd}} I_{\gamma\text{Pd}} \epsilon(E_{\gamma})_{\text{Pd}} (1 - e^{-\lambda_{\text{tPd}}}) (e^{-\lambda_{\text{tPd}}}) (1 - e^{-\lambda_{\text{tPd}}})} \prod_k \frac{(C_k)_{\text{Au}}}{(C_k)_{\text{Pd}}} \quad (4)$$

Table 2 Interpolated efficiencies with uncertainty and correlation matrix

Nuclide	γ -ray Energy	ϵ_c	$C_{\epsilon c}$				
^{109}Pd	88.03	0.0912 ± 0.0189	1				
^{101}Pd	296.3	0.0782 ± 0.0044	0.861	1			
^{105}Rh	318.9	0.0731 ± 0.0043	0.862	0.993	1		
$^{106\text{m}}\text{Rh}$	450.8	0.0456 ± 0.0020	0.314	0.004	0.075	1	
^{196}Au	355.7	0.0646 ± 0.0048	0.776	0.945	0.974	0.285	1

Table 4 Fractional uncertainties (%) with correlations in various attributes of measured reactions

Attributes	¹⁰⁹ Pd	¹⁰¹ Pd	¹⁰⁵ Rh	^{106m} Rh	Correlation
σ_{Au}	0.965	0.965	0.965	0.965	Fully correlated
C_{Pd}	3.516	5.315	10.54	8.422	Uncorrelated
C_{Au}	0.949	0.949	0.949	0.949	Fully correlated
λ_{Pd}	6.59E–02	5.79E–02	1.39E–02	5.29E–02	Uncorrelated
λ_{Au}	0.654	0.654	0.654	0.654	Fully correlated
Wt_{Pd}	2.56E–02	2.56E–02	2.56E–02	2.56E–02	Fully correlated
Wt_{Au}	2.28E–02	2.28E–02	2.28E–02	2.28E–02	Fully correlated
a_{Pd}	0.768	0.980	0.358	0.109	Uncorrelated
A_{VPd}	6.36E–06	5.89E–06	1.14E–05	1.13E–05	Uncorrelated
A_{VAu}	3.05E–07	3.05E–07	3.05E–07	3.05E–07	Fully correlated
$I_{\gamma Pd}$	3.542	4.167	3.141	5.372	Uncorrelated
$I_{\gamma Au}$	0.345	0.345	0.345	0.345	Fully correlated
$\varepsilon(E_{\gamma})_{Pd}$	20.79	5.647	6.542	4.434	Uncorrelated
$\varepsilon(E_{\gamma})_{Au}$	7.444	7.444	7.444	7.444	Fully correlated
$(\Gamma_{attn})_{Pd}$	2.861	2.286	2.088	1.934	Uncorrelated
$(\Gamma_{attn})_{Au}$	0.654	0.654	0.654	0.654	Fully correlated

The subscripts Pd and Au appearing in Eq. (4) represent the ^{nat}Pd and ¹⁹⁷Au samples. $\sigma_{Pd}(E_n)$ and $\sigma_{Au}(E_n)$ at the neutron energy E_n represent the reaction cross sections. C_{Pd} and C_{Au} are the detected photo-peak counts of the γ -rays of radionuclide's. λ_{Pd} and λ_{Au} represent the decay constants, Wt_{Pd} and Wt_{Au} represent the weights. a_{Pd} and a_{Au} represent isotopic abundances, A_{VPd} and A_{VAu} represent average atomic masses, $I_{\gamma Pd}$ and $I_{\gamma Au}$ represent the γ -ray branching ratios, $\varepsilon(E_{\gamma})_{Pd}$ and $\varepsilon(E_{\gamma})_{Au}$ represent the efficiencies. t_i , t_d and t_C represent irradiation, decay and counting time, $(C_k)_{Pd}$ and $(C_k)_{Au}$ represent the correction factors of dead time, where k represent the dead time of the detector and γ -ray self-attenuation factor (Γ_{attn}). The retrieve values of mass attenuation coefficient μ from XMuDat ver. 1.0.1 [39, 40] were further use to find self-attenuation factor (Γ_{attn}). The ¹⁹⁷Au (n,2n) ¹⁹⁶Au monitor reaction cross section at the neutron energy of 14.54 ± 0.24 MeV was estimated by using a linear interpolation technique, which is 2.1236 ± 0.0205 barns [41, 42].

From NuDat 2.7 [33, 43–47] database, the nuclear spectroscopic information's were retrieved and are presented in

Table 3. The parameters present in the neutron activation Eq. (4) with uncertainties are σ_{Au} , C_{Pd} , C_{Au} , λ_{Pd} , λ_{Au} , Wt_{Pd} , Wt_{Au} , a_{Pd} , A_{VPd} , A_{VAu} , $I_{\gamma Pd}$, $I_{\gamma Au}$, $\varepsilon(E_{\gamma})_{Pd}$, $\varepsilon(E_{\gamma})_{Au}$, $(\Gamma_{attn})_{Pd}$ and $(\Gamma_{attn})_{Au}$, whereas other parameters with negligible uncertainties are a_{Au} , t_i , t_d and t_C , respectively.

Covariance's analysis for the uncertainties of reaction cross sections

The covariance matrix $V_{\sigma S}$ [48] for uncertainty corresponding to the radio nuclides was estimated as follows.

$$(V_{\sigma S})_{ij} = \sum_{kl} (e_k)_i (s_{kl})_{ij} (e_l)_j \quad 1 \leq i, j \leq 4, \quad 1 \leq k, l \leq 16 \quad (5)$$

where $(s_{kl})_{ij}$ is the mirco-correlation matrix between the k th attribute in the i th reaction and l th attribute in the j th reaction. The partial uncertainties, $(e_k)_i = \frac{\partial \sigma_{Si}}{\partial (x_k)_i} \Delta(x_k)_i$, $(e_l)_j = \frac{\partial \sigma_{Sj}}{\partial (x_l)_j} \Delta(x_l)_j$ in σ_{Si} , σ_{Sj} due to the k th and l th attributes, respectively. The fractional uncertainties in percentage of different parameters in obtaining the

Table 3 Nuclear spectroscopic data of reaction products from Pd-isotopes and for ¹⁹⁷Au

Nuclear reaction	Threshold energy (MeV)	Product nuclide	Isotopic abundance	Half-life	γ -ray Energy (keV)	γ -ray branching ratio (%)	Decay mode (%)
¹¹⁰ Pd(n,2n)	8.878	¹⁰⁹ Pd	11.72 ± 0.09	13.59 ± 0.12 h	88.0336 ± 0.010	3.67 ± 0.13	$\beta^-(100)$
¹⁰² Pd(n,2n)	10.65	¹⁰¹ Pd	1.02 ± 0.01	8.47 ± 0.06 h	296.29 ± 0.03	19.2 ± 0.8	ε (100)
¹⁰⁵ Pd(n,p)	0.0	¹⁰⁵ Rh	22.33 ± 0.08	35.36 ± 0.06 h	318.9 ± 0.1	19.1 ± 0.0	$\beta^-(100)$
¹⁰⁶ Pd(n,p)	2.762	^{106m} Rh	27.33 ± 0.03	131 ± 0.2 min	450.8 ± 0.1	24.2 ± 0.13	$\beta^-(100)$
¹⁹⁷ Au(n,2n)	8.114	¹⁹⁶ Au	100	6.1669 ± 0.0006 day	355.7	87 ± 3	ε (93) + $\beta^-(7)$

Table 5 The measured reaction cross sections with uncertainty and correlations

Reaction	Cross section (barns)	Correlation
$^{110}\text{Pd}(n,2n)^{109}\text{Pd}$	1.9054 ± 0.4361	1
$^{102}\text{Pd}(n,2n)^{101}\text{Pd}$	0.9163 ± 0.1069	0.216 1
$^{105}\text{Pd}(n,p)^{105}\text{Rh}$	0.0433 ± 0.0064	0.172 0.337 1
$^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$	0.0079 ± 0.00096	0.207 0.406 0.322 1

$^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross sections with respect to the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ monitor reaction are presented in Table 4. The observed correlations among different observations are summarized in the sixth column of Table 4. For more details, we suggest to refer the following refs. [28, 35, 48, 49]. The measured reactions cross sections results at the 14.54 ± 0.24 MeV neutron energy with uncertainties and correlations are presented in Table 5.

Discussion

The nuclear reaction cross section of palladium isotopes have been measured with respect to the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ monitor reaction at the neutron energy of 14.54 ± 0.24 MeV by using the method of activation and off-line γ -ray spectrometry. The Purnima neutron generator was used to produced mono-energetic neutron from the $^3\text{H}(d,n)^4\text{He}$ reaction. The efficiency calibration of HPGe detector was performed by using ^{152}Eu and ^{133}Ba standard sources. The ratio method was used to find the cross sections of four different nuclear reactions. The uncertainties in the various basic nuclear parameters used in the measured reactions and their correlations were estimated by using covariance analysis. The present data were compared with literature data based on EXFOR [22, 23] compilation, the evaluated data from different libraries [50–57] and the calculated values from TALYS-1.9 [24, 25].

$^{110}\text{Pd}(n,2n)^{109}\text{Pd}$ reaction cross section

It can be seen from Fig. 2 that the measured $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$ reaction cross section at the neutron energy of 14.54 ± 0.24 MeV is in good agreement with the values of Augustynaik et al. [10], Reggoug et al. [12] and Paul et al. [6] as well as with the value of EAF-2010 [54], which are higher than the values of Minetti et al. [8], Bormann et al. [9], Sothras [11], Kong et al. [14], Filatenkov et al. [15] and Luo et al. [16]. The data of Bonazzola and Zhou et al. [13] are higher than the present value. However, the evaluated data from ENDF/B-VIII.0 [50], JEFF-3.3 [51], JENDL-4.0 [52] and BROND-3.1 [53] libraries and the calculated values

Fig. 2 Plot of $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$ reaction cross section from the present work with the literature data, evaluated data of ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0, BROND-3.1 and EAF-2010 libraries as well as with the calculated value from TALYS-1.9 as a function of neutron energy

from TALYS-1.9 [24] are significantly lower than the present data. Figure 2 also shows that the literature data [7, 9, 10] are in agreement with the calculated value from TALYS-1.9 [24] and with the evaluated data of ENDF/B-VIII.0 [50], JEFF-3.3 [51] and BROND-3.1 [53] libraries. The cross section values of ENDF/B-VIII.0 [50] and BROND-3.1 [53] libraries are the same.

$^{102}\text{Pd}(n,2n)^{101}\text{Pd}$ reaction cross section

For the $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$ reaction, Fig. 3 shows that the cross section value from the present work is lower than the literature data [9, 10, 14, 15, 17–21] and the evaluated values of ENDF/B-VIII.0 [50], JEFF-3.3 [51], JENDL-4.0 [52] and EAF-2010 [54] libraries as well as the calculated value from TALYS-1.9 [24]. Figure 3 also shows that the present and literature data [13] within the neutron energies of 12–20 MeV are in close agreement with the calculated data of ROSFOND-2010 [55] library. The cross section values of ENDF/B-VIII.0 [50] and ROSFOND-2010 [55] libraries are the same.

$^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross section

For the $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ reaction, Fig. 4 shows that the cross section value from the present work is slightly higher than the literature data [14, 15, 21] and evaluated data of ENDF/B-VIII.0 [50], JEFF-3.3 [51], JENDL-4.0 [52] and ROSFOND-2010 [55] libraries as well as the calculated values from TALYS-1.9 [24]. Figure 4 also shows that the present and literature data [17] within the neutron energies

Fig. 3 Plot of $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$ reaction cross section from the present work with the literature data, evaluated data of ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0, ROSFOND-2010 and EAF-2010 libraries as well as with the calculated value from TALYS-1.9 as a function of neutron energy

of 12–20 MeV are in close agreement. The cross section values of ENDF/B-VIII.0 [50] and JEFF-3.3 [51] libraries are the same.

$^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross section

For the $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction, Fig. 5 shows that the cross section value from the present work at the neutron energy of 14.54 ± 0.24 MeV is in very good agreement with the

Fig. 4 Plot of $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross section from the present work with the literature data, evaluated data of ENDF/B-VIII.0, JEFF-3.3, JENDL-4.0 and ROSFOND-2010 libraries as well as with the calculated value from TALYS-1.9 as a function of neutron energy

Fig. 5 Plot of $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross section from the present work with the literature data, evaluated data of JEFF-3.1/A, JENDL/AD-2017 and EAF-2010 libraries as well as with the calculated value from TALYS-1.9 as a function of neutron energy

literature data [15, 17, 20, 21]. Figure 5 also shows that the data from the present work and literature [15, 17, 20, 21] within the neutron energies of 12–20 MeV are in close agreement with the calculated values from TALYS-1.9 code [24] but are lower than the evaluated data of Kong et al. [14], JEFF-3.1/A [56], JENDL/AD-2017 [57] and EAF-2010 [54]. The cross section values of JEFF-3.1/A [56] and EAF-2010 [54] libraries are the same.

Summary

The $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reaction cross sections have been measured with respect to the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ monitor reaction at the neutron energy of 14.54 ± 0.24 MeV. The method of an activation and off-line γ -ray spectrometry have been used. The efficiency calibration of HPGe detector was performed using the γ -ray energies of standard ^{152}Eu and ^{133}Ba sources. The uncertainties in various attributes were performed using covariance analysis with their correlations coefficients. The measured reaction cross sections data of $^{110}\text{Pd}(n,2n)^{109}\text{Pd}$, $^{102}\text{Pd}(n,2n)^{101}\text{Pd}$, $^{105}\text{Pd}(n,p)^{105}\text{Rh}$ and $^{106}\text{Pd}(n,p)^{106\text{m}}\text{Rh}$ reactions were compared with the literature data of EXFOR compilation, evaluated data of various libraries as well as with the calculated values from TALYS-1.9. In general, the experimental values are found to be in good agreement with the calculated data from TALYS-1.9 and evaluated data of different libraries.

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References

- Moinul Haque Meaze AKM, Kim GN (2011) Determination of niobium and palladium resonance cross sections by time-of-flight transmission experiment. *Indian J Phys* 85(2):329–338
- Uwamino Y, Sugita H, Kondo Y, Nakamura T (1992) Measurement of neutron activation cross sections of energy up to 40 MeV using semi-monoenergetic p-Be neutrons. *Nucl Sci Eng* 111(4):391–403
- Iwamura Y, Sakano M, Itho T (2002) Elemental analysis of Pd complexes: effects of D₂ gas permeation. *Jpn J Appl Phys* 41:4642–4650
- Khandaker MU, Kim K, Kim G, Otuka N (2010) Cyclotron production of the ^{105,106m}Ag, ^{100,101}Pd, ^{100,101m,105}Rh radionuclides by ^{nat}Pd(p,x) nuclear processes. *Nucl Instrum Methods Phys Res B* 268:2303–2311
- Wang T, Kim G, Oh YD, Cho MH, Ko IS, Namkung W (2016) Neutron total cross sections and resonance parameters of palladium. *Nucl Sci Eng* 183:286–297
- Paul EB, Clarke RL (1953) Cross section measurements of reactions induced by neutrons of 14.5 MeV energy. *Can J Phys* 32:267–277
- Bonazzola GC, Brovetto P, Chiavassa E, Spinoglio R (1964) The measurement by activation of cross sections for 14.7 MeV neutrons. *Nucl Phys* 51:337–344
- Minetti B, Pasquarelli A (1967) Isomeric cross section ratio for ¹¹⁰Pd(n,2n)¹⁰⁹Pd reaction at 14.7 MeV. *Zeitschrift Physik* 207:132–137
- Bormann M, Bissem HH, Magiera E, Warnemunde R (1970) Total cross sections and isomeric cross section ratios for (n,2n) reactions in energy region 12–18 MeV. *Nucl Phys A* 157:481–496
- Augustyniak W, Herman M, Marcinkowski A, Zwięglinski B (1977) Cross sections for the (n,2n) reaction on ¹⁰²Pd, ¹¹⁰Pd, ¹¹²Cd and ¹⁹²Os. *Nucl Phys A* 285:145–155
- Sothras SL (1977) A study of the systematics for (n,2n) reactions. Thesis: Sothras
- Reggoug A, Paic G, Berrada M (1982) Measurement of (n,2n) reaction cross sections by X-ray spectroscopy. Progress, Univ. Mohammed V, Rabat, Annual report, 5:14
- Zhou M, Zhang Y, Wang C, Zhang L, Chen Y, Zhou S, Zhang S, Xie K, Zhou S, Chen X, Zhang Y, Yan Q (1987) Shell effect from the cross section of the (n,2n) reaction produced by 14.6 MeV neutron. *Chin J Nucl Phys* 9:34
- Kong X, Wang R, Wang Y, Yang J (1999) Cross sections for 13.5–14.7 MeV neutron induced reactions on palladium isotopes. *Appl Radiat Isot* 50:361–364
- Filatenkov AA (2016) Neutron activation cross sections measured at KRI in neutron energy region 13.4–14.9 MeV. INDC (CCP)-0460 Rev
- Luo J, Jiang L, He L (2018) Activation cross sections for the ¹¹⁰Pd(n,2n) reaction and the isomeric cross sections ratio of ^{109m,g}Pd in the neutron energy range of 13–15 MeV. *J Radioanal Nucl Chem* 316:733–739
- Lu WD, Ranakumar N, Fink RW (1970) Activation cross sections for (n,p), [(n,np)+(n,np)+(n,d)], and (n,a) reactions in the region of Z=40 to 58 at 14.4 MeV. *Phys Rev C* 1:350–357
- Temperley JK, Barnes DE (1970) Neutron activation cross sections for some isotopes of ruthenium, palladium, indium, tin and iridium. Ballistic research labs reports, No. 1491
- White RL, Gray TJ (1972) Neutron cross sections in Pd isotopes over an energy range of 15.1–16.3 MeV. *J Bull Am Phys Soc* 17:687
- Goncalves IF, Schram ZS, Papp Z, Daroczy S (1987) (n,p), (n,α) and (n,2n) reaction cross-sections for some isotopes of Zr, Pd and Cd at 14.8 MeV. *Appl Radiat Isot* 11:989–991
- Konno C, Ikeda Y, Oishi K, Kawade K, Yamamoto H, Maekawa H (1993) Activation cross section measurements at neutron energy from 13.3 to 14.9 MeV. R, JAERI-1329
- IAEA-EXFOR Database available at <https://www-nds.iaea.org/exfor>
- Otuka N, Dupont E, Semkova V, Pritychenko B, Blokhin AI, Aikawa M, Babykina S, Bossant M, Chen G, Dunaeva S, Forrest RA, Fukahori T, Furutachi N, Ganesan S, Ge Z, Gritzay OO, Herman M, Hlavac S, Zhuang Y (2014) Towards a more complete and accurate experimental nuclear reaction data library (EXFOR): international collaboration between nuclear reaction data centers (NRDC). *Nucl Data Sheets* 120:272–276
- Koning AJ, Hilaire S, Goriely S (2017) TALYS-1.9, A Nuclear Reaction Program (NRG-1755 ZG Petten, The Netherlands). <https://www.talys.eu/download-talys/>
- Koning AJ, Rochman D (2012) Modern nuclear data evaluation with the TALYS code system. *Nucl Data Sheets* 113:2841–2934
- Sinha A, Roy T, Yogesh K, Ray N, Shukla M, Patel T, Bajpai S, Sarkar PS, Bishnoi S (2015) Experimental subcritical facility driven by D-D/D-T neutron generator at BARC, India. *Nucl Instrum Methods Phys Res B* 350:66–70
- Patel T, Sinha A (2013) Development of low energy deuteron accelerator based DC and pulsed neutron generators, BARC newsletter 146
- Pasha I, Rudraswamy B, Yerranguntla SS, Suryanarayana SV, Naik H, Meghna K, Radha E, Pandikumar G, Sunitha AM, Sachhidananda HB (2019) Measurement of ⁶⁷Zn(n,p)⁶⁷Cu, ⁶⁴Zn(n,2n)⁶³Zn, ⁸⁹Y(n,g)^{90m}Y and ⁸⁹Y(n,2n)⁸⁸Y reaction cross sections at the neutron energy of 14.54 MeV with covariance analysis. *J Radioanal Nucl Chem* 322(3):2057–2064
- Pasha I, Rudraswamy B, Yerranguntla SS, Suryanarayana SV, Naik H, Midhun CV, Tarun P (2020) Measurement of 14.54 MeV neutron induced reaction cross sections of Cr and Mn with covariance analysis. *Radiochim. Acta*. <https://doi.org/10.1515/ract-2019-3189>
- Agostinelli S et al (2002) GEANT4—a simulation toolkit. *Nucl Instrum Methods Phys Res A* 506:250–303
- Martin MJ (2013) Nuclear data sheets for A = 152*. *Nucl Data Sheets* 114:1497–1847
- Khazov Yu, Rodionov A (2011) Nuclear data sheets for A = 133*. *Nucl Data Sheets* 112:855–1113
- NuDat 2.7 (2016) National Nuclear Data Center, Brookhaven National Laboratory, <https://www.nndc.bnl.gov/nudat2>
- Vidmar T (2005) EFFTRAN—a monte carlo efficiency transfer code for gamma-ray spectrometry. *Nucl Instrum Methods Phys Res A* 550:603–608
- Sheela SY, Naik H, Prasad KM, Ganesan S, Suryanarayana SV (2017) Detailed data sets related to covariance analysis of the measurement of cross section of ⁵⁹Co(n,γ)⁶⁰Co reaction relative to the cross section of ¹¹⁵In(n,γ)^{116m}In. Internal report no. Mu/Statics/DAE-BRNS/2017, 10.13140/RG.2.2.26729.49764
- Geraldo LP, Smith DL (1990) Covariance analysis and fitting of germanium gamma-ray detector efficiency calibration data. *Nucl Instrum Methods Phys Res A* 290:499–508
- Pasha I, Rudraswamy B, Radha E, Sathiamoorthy V (2018) Efficiency of high-purity germanium detector at characteristic gamma energies of ¹⁹⁸Au and ⁵⁸Co and covariance analysis. *Radiat Prot Environ* 41:110–114

38. Geraldo LP, Smith DL (1989) least square methods and covariance matrix applied to the relative efficiency calibration of a Ge(Li) detector. *Inst de Pesquisas Energeticas Nucleares* 243:1–16
39. Millsap DW, Landsberger S (2015) Self-attenuation as a function of gamma ray energy in naturally occurring radioactive material in the oil and gas industry. *Appl Radiat Isot* 97:21–433
40. Nowotny R (1998) XMuDat photon attenuation data on PC. IAEA Report IAEA-NDS 195. <https://www-nds.iaea.org/publications/iaea-nds>
41. Capote R, Zolotarev KI, Pronyaev VG, Trkov A (2012) Updating and extending the IRDF-2002 dosimetry library. <https://www-nds.iaea.org/IRDF/>
42. Trkov A, Griffin PJ, Simakov SP, Greenwood LR, Zolotarev KI, Capote R, Aldama DL, Chechev V, Destouches C, Kahler AC, Konno C, Kostal M, Majerle M, Malambu E, Ohta M, Pronyaev VG, Radulovic V, Sato S, Schulc M, Simeckova E, Vavtar I, Wagemans J, White M, Yashima H (2020) IRDFF-II: a new neutron metrology library. *Nucl Data Sheets* 163:1–108
43. Kumar S, Chen J, Kondev FG (2016) Nuclear data sheets for A = 109*. *Nucl Data Sheets* 137:1–286
44. Jean B (1985) Nuclear data sheets for A = 101*. *Nucl Data Sheets* 45:701
45. De Frenne D, Jacobs E (2005) Nuclear data sheets for A = 105*. *Nucl Data Sheets* 105:775–958
46. De Frenne D, Negret A (2008) Nuclear data sheets for A = 106. *Nucl Data Sheets* 109:943–1102
47. Huang X (2007) Nuclear data sheets for A = 196*. *Nucl Data Sheets* 108:1093–1286
48. Yerraguntla SS, Naik H, Karantha M P, Ganesan S, Suryanarayana SV, Badwar S (2017) Measurement of $^{59}\text{Co}(n, \gamma)^{60}\text{Co}$ reaction cross sections at the effective neutron energies of 11.98 and 15.75 MeV. *J Radioanal Nucl Chem* 314(1):457–465
49. Pasha I, Basavanna R, Yerraguntla SS, Suryanarayana SS, Meghna K, Naik H, Prasad MK, Danu LS, Saroj B, Petel T, Rajeev K (2019) $^{93}\text{Nb}(n, 2n)^{92\text{m}}\text{Nb}$, $^{93}\text{Nb}(n, \alpha)^{90\text{m}}\text{Y}$ and $^{92}\text{Mo}(n, p)^{92\text{m}}\text{Nb}$ reactions at 14.78 MeV and covariance analysis. *J Radioanal Nucl Chem* 320:561–568
50. Herman M, Trkov A, Capote GR, Nonre GPA, Brown DA, Arcilla R, Danon Y, Plompen A, Mughabghab SF, Jing Q, Zhigang G, Tingjin L, Hanlin L, Xichao R, Leal L, Carlson BV, Kawano T, Sin M, Simakov Stanislav P, Guber K (2018) Evaluation of neutron reactions on iron isotopes for CIELO and EDNF/B-VIII.0. *Nucl data sheets* 148:214–253
51. Koning AJ, Bauge E, Dean CJ, Dupont E, Fisher U, Forrest RA, Jacqmin R, Leeb H, Kellet MA, Mills RW, Nordborg C, Pescarini M, Rungma Y, Rullhusen P (2011) Status of the JEFF nuclear data library. *J Kor Phys Soc* 59:1057–1062
52. Shibata K, Iwamoto O, Nakagawa T, Iwamoto N, Ichihara A, Kunieda S, Chiba S, Furutaka K, Otuka N, Ohasawa T, Murata T, Matsunobu H, Zukeran A, Kamada S (2011) JENDL-4.0, a new library for nuclear science and engineering. *Nucl Sci Technol* 48:1–30
53. Blokhin AI, Gai EV, Ignatyuk AV, Koba II, Manokin VN (2016) New version of neutron evaluation data library BROND-3.1, problems of atomic science and technology. *Ser. Nucl. React. Constants* 2:2–5
54. Sublet J-C, Packer LW, Kopecky J, Forrest RA, Koning AJ, Rochman DA (2010) The European activation file: EAF-2010 neutron-induced cross section. *CCFE-R* 10:05
55. Zabrodskaya SV, Ignatyuk AV, Koscheev VN (2007) Nuclear constants. *ROSFOND-2010*, pp 1–2
56. Sublet J-Ch, Koning AJ, Forrest RA, Kopecky J (2004) The JEFF-3.1/A. In: *Proceedings of international conference on nuclear data for science technology*
57. Shibata K, Iwamoto N, Kunieda S, Minato F, Iwamoto O (2016) Activation cross-section file for decommissioning of LWRs. *IAEA-Conference* 47

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