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Thermal neutron capture cross sections and resonance integrals of ruthenium isotopes- ^{96}Ru, ^{102}Ru and ^{104}Ru

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HIGHLIGHTS

- Thermal neutron capture cross sections and resonance integrals of ^{96,102,104}Ru.
- The cross sections are measured with reference to a ^{55}Mn monitor.
- Neutrons from an Am–Be neutron source kept inside a concrete bunker is used.
- ^{104}Ru data is measured from ^{105}Ru and ^{105}Rh gamma emissions.

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ABSTRACT

Thermal neutron capture cross sections and resonance integrals of ^{96}Ru, ^{102}Ru and ^{104}Ru are measured with reference to a ^{55}Mn monitor. The experiments are carried out using the neutrons from an Am–Be neutron source kept inside a concrete bunker. The thermal neutron capture cross sections measured are in good agreement with the evaluations as well as with one of the recent measurements. The resonance integral of ^{102}Ru measured in the present study is in good agreement with older measurements.

1. Introduction

The thermal neutron capture cross sections and resonance integrals are two important low energy neutron reaction data. There are continuous efforts in the form of experiments and evaluations in updating the data with improved accuracy and compiling the results. Ruthenium is one such element that has discrepancies between various measurements and evaluations due to the change in various nuclear data including abundance and half-life (Krane, 2010). The isotopes of Ru have relatively larger cumulative yields for thermal neutron fission of ^{235}U (0.061 for ^{99}Ru, 0.052 for ^{101}Ru, and 0.043 for ^{102}Ru). Production cross sections of long lived nuclides ^{93}Mo, ^{97}Tc, ^{99}Tc *etc.* from Ru isotopes are important in light water reactors. Determination of Ru concentration is important since it is one of the possible comparator for neutron activationanalysis of geological samples. Therefore, the accurate knowledge of neutron interaction cross sections of Ru isotope is important (Krane, 2010; Shibata, 2013)

The stable isotopes of Ru addressed in this study are ^{96}Ru,^{102}Ru and ^{104}Ru forming ^{97}Ru, ^{103}Ru and ^{105}Ru on neutron capture respectively. There are experimental data and the evaluated or recommended data of thermal neutron capture cross-section and resonance integrals of ^{96}Ru, ^{102}Ru and ^{104}Ru available in literature (Arboccò et al., 2014; Heft, 1978; Ishikawa, 1969; Halperin and Druschel, 1965; Lantz, 1965; Katcoff and Williams, 1958; Bereznai et al., 1977; Van der Linden et al., 1972; Ricabarra et al., 1969; Mughabghab, 2006; Sublet et al., 2010; Shibata et al., 2011; Koning et al., 2011; Brown et al., 2018; Pritychenko and Mughabghab, 2012) and are summarised in Table 1 and Table 2.

Out of the experimental studies, the recent measurements (Krane, 2010; Arboccò et al., 2014) are not in a good agreement with each other in the thermal neutron capture cross section measurements. The measured resonance integrals have a large spread in the case of ^{96}Ru and ^{104}Ru. There exists discrepancy among the various evaluations in the case of resonance integral of ^{96}Ru. The careful analysis of the literature values suggests that additional measurements are of importance in the case of Ru isotopes.

All these measurements utilised the neutrons from a reactor whereas the present study focuses on the thermal neutron capture cross section and resonance integral measurements of ^{96}Ru, ^{102}Ru and ^{104}Ru irradiated in an Am–Be neutron source facility. The most commonly used

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Table 1
The thermal neutron cross sections ($\sigma_{0,S}$)of ^{96}Ru, ^{102}Ru and ^{104}Ru including measurements and the evaluations.

Reference	Thermal neutron capture cross section, $\sigma_{0,S}$ (b)		
	^{96}Ru	^{102}Ru	^{104}Ru
F.F.Arbocco et al., 2014 (Arboccò et al., 2014)	0.248 ± 0.002	1.241 ± 0.001	0.505 ± 0.005
K.S.Krane,2010 (Krane, 2010)	0.207 ± 0.01	1.14 ± 0.05	0.468 ± 0.019
R.E.Heft, 1978 (Heft, 1978)	0.218 ± 0.004	1.31 ± 0.03	0.466 ± 0.015
H.Ishikawa, 1969 (Ishikawa, 1969)		1.37 ± 0.13	
J.Halperin et al., 1965 (Halperin and Druschel, 1965)	0.271 ± 0.027		
P.M.Lantz et al., 1965 (Lantz, 1965)		1.23 ± 0.12	0.47
S.Katcoff et al., 1958 (Katcoff and Williams, 1958)	0.21	1.5	
S. F. Mughabghab,2003 (Mughabghab, 2006)	0.29 ± 0.02	1.27 ± 0.04	0.49 ± 0.01
EAF-2010 (Sublet et al., 2010)	0.249	1.27	0.472
JENDL 4.0 (Shibata et al., 2011)	0.271	1.48	0.469
JEFF 3.3 (Koning et al., 2011)	0.289	1.27	0.472
ENDF-B.VIII (Brown et al., 2018)	0.290	1.27	0.472

Table 2
The resonance integrals ($I_{0,S}$) of ^{96}Ru, ^{102}Ru and ^{104}Ru including measurements and the evaluations.

Reference	Resonance Integrals, $I_{0,S}$ (b)		
	^{96}Ru	^{102}Ru	^{104}Ru
K.S.Krane,2010 (Krane, 2010)	7.21 ± 0.36	4.85 ± 0.24	7.03 ± 0.35
R.E.Heft, 1978 (Heft, 1978)	7.0 ± 0.3	4.68 ± 0.75	7.70 ± 0.65
T. Bereznai et al., 1977 (Bereznai et al., 1977)	6.5 ± 0.8	4.7 ± 3.6	5.9 ± 2.5
R.Van der Linden et al., 1972 (Van der Linden et al., 1972)	4.8 ± 0.2	4.3 ± 0.4	6.5 ± 0.3
M.D.Ricabarra,1969 (Ricabarra et al., 1969)	6.67 ± 0.11	4.80 ± 0.52	4.36
J.Halperin et al., 1965 (Halperin and Druschel, 1965)	5.51 ± 0.39		
P.M.Lantz et al., 1965 (Lantz, 1965)		4.14 ± 41	4.6
S.F.Mughabghab,2003 (Mughabghab, 2006)	6.36 ± 0.23	4.9 ± 0.3	6.3 ± 0.2
EAF-2010 (Sublet et al., 2010)	6.79	5.24	6.45
JENDL 4.0 (Shibata et al., 2011)	5.99	4.19	6.45
JEFF 3.3 (Koning et al., 2011)	8.73	5.24	6.45
ENDF-B.VIII (Brown et al., 2018)	6.87	5.24	6.45

monitors for cross section measurements in the activation technique are ^{197}Au and ^{55}Mn. Our previous studies showed that the cross sections measured with reference to the monitor reactions ^{55}Mn (n, γ) ^{56}Mn and ^{197}Au (n, γ) ^{198}Au are in good agreement (Panikkath and Mohanakrishnan, 2016, 2017; Panikkath et al., 2019). In the present work the cross sections are measured with reference to the monitor reaction ^{55}Mn (n, γ) ^{56}Mn reaction. The details of the uncertainty analysis are also explained in this work.

2. Experimental

The irradiation experiments were performed in the neutron beam at the Am–Be neutron source facility available at Manipal Centre for Natural Sciences, Manipal Academy of Higher Education. The doubly encapsulated neutron source having a yield of 4×10^7 n/s is kept inside a concrete bunker having an irradiation channel in one of the side. The details of the experimental facility are available in the previous works (Panikkath and Mohanakrishnan, 2016, 2017; Panikkath et al., 2019). The fast neutron spectrum of Am–Be source is modified significantly due to the scattering of neutrons with concrete surrounding. The modified spectrum contains thermal, epithermal and fast neutrons. The epithermal neutrons are having a $1/E^{(1+\alpha)}$dependency where α is known as the epithermal spectrum shaping factor that accounts for the deviation of the epithermal neutron spectrum from $1/E$behaviour. The parameter α is estimated as -0.148 ± 0.007 from a multi-foil activation and subsequent spectrum unfolding method. The neutron spectrum obtained from the unfolding method is compared with that obtained from a Monte Carlo simulation in a previous work (Panikkath and Mohanakrishnan, 2016). Due to the different bin structure followed in the unfolding code and the Monte Carlo simulation, one to one comparison is not possible. However, the total neutron fluxes obtained from both these methods are comparable.

Analytical grade RuO_2 powder prepared in small packets was used as the sample material. Mn (Mn(83 wt%)-Cu) foil procured from Shieldwerx with purity 99.9% was used as the monitor foil. The length and breadth of the powder samples prepared were measured and the uniformity of the thickness is assured by firmly pressing the sample. The details of the dimensions of irradiated samples are summarised in Table 3. The thicknesses of the powder samples were estimated from the known values i.e., density (6.97 g/cm^3), mass and the cross sectional area. Two sets of sample and monitor were irradiated together in the irradiation channel where one set of each were kept inside a cadmium foil of 1 mm thick. The samples were irradiated for a duration of 14 days.

The induced activity in each irradiated material was estimated from the corresponding gamma spectra using a 30% relative efficiency HPGe detector pre-calibrated using a^{152}Eu source. The gamma energies 121.78 keV, 244.69 keV, 344.28 keV, 411.12 keV, 778.90 keV, 867.38 keV, 964.06 keV, 1112.08 keV and 1408.01 keV of ^{152}Eu are used for estimating the detector efficiency. The distance between the irradiated material as well as the calibration source and the detector was 2 cm. The coincidence summing effects of the ^{152}Eu lines are

Table 3
The properties of samples used in the experimental study.

Sample	Type of irradiation	Mass (mg)	dimension	Thickness (mm)
RuO_2	Bare	465	2.4 cm × 2 cm	0.14
	Cd-covered	470	2.1 cm × 1.9 cm	0.17
Mn(83%)-Cu	Bare	49.7	12 mm (dia)	0.05
	Cd-covered	49.4	12 mm (dia)	0.05

estimated and corrected using the Monte Carlo simulation code EFFTRAN (Vidmar, 2005). However, in the case of irradiated samples, the co-incidence effects were neglected due to the lower activity that will be produced. The cadmium covered samples were counted after a delay of 15600 s. The delay time in the case of bare samples except ^{55}Mn(n,γ)^{56}Mn and ^{104}Ru(n,γ)^{105}Ru reactions were 76000 s. The ^{55}Mn (n,γ)^{56}Mn and ^{104}Ru(n,γ)^{105}Ru were measured with a delay time of 6600 s and counting time of 6000 s due to their shorter half-lives. Other foils were counted for 20000 s–60000 s depending upon the half-life and the counting statistics.

The irradiated samples in the present study, especially the Ru samples can be considered as an extended source. The efficiencies of the disc source are obtained from the efficiencies of the point source at the corresponding energies using the code EFFTRAN. It was found that the change in the efficiencies can be neglected since the ratios of the efficiencies are utilised in the estimation. The induced activities in the ^{56}Mn, ^{97}Ru, ^{103}Ru and ^{105}Ru isotopes were determined by measuring the gamma peak area under 846.7 keV, 215.7 keV, 497.1 keV and 724.3 keV gamma lines respectively. The ^{105}Ru activity produced was determined from the ^{105}Rh activity measurement also where the ^{105}Rh is the beta decay product of ^{105}Ru. The net areas of the gamma peaks were obtained using the WINSPEC gamma spectrum analysis software after the continuum subtraction.

3. Data analysis

3.1. Estimation of the reaction rate

The reaction rate (R) per target nuclei is estimated from the measured gamma peak counts (C) using eq. 1

$$R = \frac{CMf}{TN_A\theta I_\gamma \varepsilon m} \tag{1}$$

where M is the molar mass, f is the gamma attenuation factor, N_A is the Avogadro's number, θ is the isotopic abundance, ε is the efficiency of the detector, m is the sample mass, I_γ is the gamma yield and T is the time factor. The time factor is defined as in eq. (2) for the case of ^{56}Mn, ^{97}Ru, ^{103}Ru and ^{105}Ru. The time factor T is defined as shown in eq. (3) to estimate the reaction rate of ^{105}Ru from the ^{105}Rh gamma photo peak.

$$T = \frac{(1-e^{-\lambda t_i})(e^{-\lambda t_d})(1-e^{-\lambda t_c})}{\lambda} \tag{2}$$

$$T = \frac{(1-e^{-\lambda_1 t_i})}{\lambda_1}\left(\frac{\lambda_2}{\lambda_2-\lambda_1}\right)\left(e^{-\lambda_1 t_d}\right)\left(1-e^{-\lambda_1 t_c}\right)+$$

$$\left[\frac{(1-e^{-\lambda_1 t_i})\lambda_2-(1-e^{-\lambda_2 t_i})\lambda_1}{\lambda_2(\lambda_2-\lambda_1)}-\frac{(1-e^{-\lambda_1 t_i})}{(\lambda_2-\lambda_1)}\right]\left(e^{-\lambda_2 t_d}\right)\left(1-e^{-\lambda_2 t_c}\right) \tag{3}$$

where λ is the decay constant, t_i, $t_d$and $t_c$are the irradiation time, delay time (cooling time) and counting time respectively. The subscripts 1 and 2 in eq. (3) stands for ^{105}Ru and ^{105}Rh respectively. Nuclear data used in the measurement and further estimation of the cross section are given in Table 4. The reaction rates obtained from the present measurement are also tabulated Table 4.

3.2. Determination of thermal neutron capture cross section

The thermal neutron capture cross section of the reaction of interest ($\sigma_{0,S}$) is estimated with reference to that of ^{55}Mn(n,γ)^{56}Mn reaction ($\sigma_{0,M}$ = 13.36 b) (Mughabghab, 2006) reaction using the eq. 4

$$\sigma_{0,S} = \frac{[R - R_{Cd}/F_{Cd}]_S}{[R - R_{Cd}/F_{Cd}]_M}\frac{[G_{th}\, g]_M}{[G_{th}\, g]_S}\sigma_{0,M} \tag{4}$$

where R and R_{Cd} reaction rates of bare and cadmium covered samples, F_{Cd} is the cadmium transmission factor, $G_{th}$is the thermal self shielding correction factor, g is the Westcott's factor for correcting the deviation of cross sections from $1/v$ behaviour. The subscripts S and Mindicate the sample and monitor respectively.

3.3. Determination of resonance integral

The resonance integral for the real spectrum having a $1/E^{(1+\alpha)}$ dependency $I(\alpha)$) and the resonance integral for ideal spectrum with $\alpha = 0$ (I_0) are related as shown in eq. (5).

$$I(\alpha) = (1eV)^\alpha\left[\frac{I_0 - 0.45\sigma_0}{(\overline{E_r}^\alpha)} + \frac{0.45\sigma_0}{(2\alpha+1)(E_{Cd})^\alpha}\right] \tag{5}$$

where $\overline{E_r}$ is the effective resonance energy and E_{Cd} is the Cadmium cut off energy. The value of E_{Cd} depends on the thickness of cadmium foil ($E_{Cd} \approx 0.5\,eV$ for 1 mm thickness). The value 0.45 is obtained from the ratio of thermal neutron energy (E_o = 0.025 eV) and $E_{Cd}(2\sqrt{E_o/E_{Cd}} \approx 0.45)$. The $I(\alpha)$can be measured from the reaction rates as follows and then the I_0 can be estimated using eq. (5).

$$I(\alpha)_S = I(\alpha)_M\frac{\sigma_{0,S}(CR-1)_M}{\sigma_{0,M}(CR-1)_S}\frac{G_{epi,M}}{G_{epi,S}}\frac{G_{th},\, S}{G_{th,M}} \tag{6}$$

With

$$CR = \frac{R}{R_{Cd}/F_{Cd}} \tag{7}$$

where G_{epi} is the epithermal self-shielding factor. The $I(\alpha)$ of the ^{55}Mn (n,γ)^{56}Mn reaction can be estimated using eq. (5) with $I_{0,M}$ = 13.4 b (Mughabghab, 2006)

The thermal and epithermal self-shielding factors (G_{th} and G_{epi})were estimated analytically using the formula described below (Panikkath and Mohanakrishnan, 2017; Blaauw, 1995; Martinho et al., 2003).

$$G_{th} = \frac{(1-e^{-\xi})}{\xi};\ \xi = \frac{2}{\sqrt{\pi}}\frac{\rho N_A}{M}\sigma_0 t \tag{8}$$

where t is the thickness of the target along the beam direction and ρ is the density of the target.

$$G_{epi} = \frac{0.94}{1+(z/2.70)^{0.82}} + 0.06;\ z = \frac{\rho N_A}{M}\sigma(E_{res}) \times 1.5t \times \frac{\Gamma_\gamma}{\Gamma}6 \tag{9}$$

where $\sigma(E_{res})$ is the cross section at the resonance peak corresponding to energy E_{res}, Γ_γ is the resonance width corresponding to the neutron

Table 4
Abundance θ (Krane, 2010), Westcott factor g (Brown et al., 2018), Effective resonance energy $\overline{E_r}$ (Jaćimović et al., 2014). Decay data (gamma energy$E\gamma$, gamma yield $I\gamma$, half life $T_{1/2}$) (Junde et al., 2011; Nica, 2010; De Frenne, 2009; De Frenne and Jacobs, 2005), gamma attenuation factor f (Berger et al., 2010) adopted in the present study.

Target nuclei	θ(%)	g	$\overline{E_r}(eV)$	Product nuclei	$E\gamma$ (keV)	$I\gamma$ (%)	$T_{1/2}$	f	R	R_{Cd}
^{55}Mn	100	1.0006	468 ± 51	^{56}Mn	846.76	98.85 ± 0.03	2.5789 ± 0.0001 h	1.005	3.31E-24	2.81E-24
^{96}Ru	5.54 ± 0.14	1.0006	776 ± 16	^{97}Ru	215.76	85.62	2.83 ± 0.23 d	1.012	4.73E-24	2.36E-24
^{102}Ru	31.55 ± 0.14	1.0005	181 ± 4	^{103}Ru	497.09	91.0 ± 1.2	39.247 ± 0.013 d	1.005	3.84E-24	2.99E-24
^{104}Ru	18.62 ± 0.27	1.0006	495 ± 10	^{105}Ru	724.3	47.3 ± 0.5	4.44 ± 0.02 h	1.004	3.90E-24	3.04E-24
				^{105}Rh	318.9	19.1 ± 0.6	35.36 ± 0.06 h	1.007	2.97E-23	5.88E-24

Table 5
The thermal and epithermal self shielding factors estimated and the parameters (total cross section σ_{res}at the resonance peak E_{res}, gamma width Γ_γ and total width Γ (Brown et al., 2018)) used in the estimation.

Isotope	E_{res} (eV)	σ_{res} (b)	$\Gamma\gamma$(eV)	Γ(eV)	G_{th}	G_{epi}
^{55}Mn	340.79	3239	0.41	24.09	0.999	0.891
^{96}Ru	–	–	–	–	0.999	1
^{102}Ru	9.802	12.41	0.128	0.128		0.994
^{104}Ru	65.66	86.16	0.103	0.103		0.980

capture (n, γ) reaction and Γ is the total resonance width. It can be seen that the self shielding correction factors estimated analytically are universal and independent of the irradiation channel surroundings. The factors depend on the material properties (density, thickness and molar mass) and the nuclear data (cross section, gamma width, neutron width and total width) (Brown et al., 2018). These parameters and the estimated G_{th} and G_{epi} are given in Table 5. Since no resonance parameters are available for ^{96}Ru, the epithermal self-shielding factor is considered as unity in the present study.

4. Uncertainty analysis

The thermal neutron capture cross section and the resonance integrals measured depends on various nuclear data as well as experimental quantities. Hence it is important to report the result with a detailed uncertainty analysis. However, most of the earlier works report total uncertainties as the quadratic sum of the individual uncertainties or just report the total uncertainties without any details. The total uncertainty on σ_0 cannot be calculated as the quadratic sum of the fractional sensitivities $\left(\frac{\Delta k_i}{k_i}\right)$ of the individual parameters (Panikkath et al., 2019), where $k_i$are different parameters that are used in the estimation of σ_0. Following the methodology explained in reference (Panikkath et al., 2019), the total uncertainty $\frac{\Delta X}{X}$ where $X = \sigma_0$or I_o is defined as

$$\frac{\Delta X}{X} = \sqrt{\sum_i \left(S_i \frac{\Delta k_i}{k_i}\right)^2} \tag{10}$$

with

$$S_i = \frac{k_i}{X}\frac{\partial X}{\partial k_i} \tag{11}$$

The fractional sensitivity coefficient S_i is defined as the sensitivity of the parameter k_i on X. The sensitivity coefficient is obtained by taking the partial derivative of the X with respect to k_i analytically. The same can be obtained using a computer code as follows (Panikkath and Mohanakrishnan, 2016, 2017). Each parameter (k_i) is varied by its corresponding uncertainty $(k_i + \Delta k_i)$ one at a time. The X (σ_0 or I_o) is estimated using this altered parameter as X_i' (σ_0 'or I_o'). The fractional change between X_i' and Xcorresponding is found to be equal to $S_i\frac{\Delta k_i}{k_i}$. Thus the quadrature sum of $X_i' - X$ gives the total uncertainty. The sensitivity coefficient S_i can be obtained from the ratios of $X_i' - X$ and $\frac{\Delta k_i}{k_i}$ for each parameter, k_i. The entire analysis is performed using a program written in MATLAB.

The Δk_i 's are available readily from nuclear data or measurement except for the time constant. The time factor is not proportional to the decay constant (eq. (2) and eq. (3)) and hence to the reaction rate, but their relation is more complicated (exponential). So, the uncertainties in the time factor is propagated from the uncertainties in the decay constant as below:

$$\Delta T = \frac{dT}{d\lambda}\Delta\lambda \tag{12}$$

The uncertainties in the irradiation time, delay time and the counting time are assumed to be negligible in the present experiment. The detail explanation on the uncertainty analysis can be seen in the references (Panikkath et al., 2019; Otuka et al., 2017).

5. Results and discussions

The fractional uncertainties $\frac{\Delta k_i}{k_i}$ and the corresponding sensitivity coefficient $S_i$that contribute to the total uncertainties of the thermal neutron capture cross sections and the resonance integrals are tabulated in Table 6 and Table 7 respectively. The parameters indicated in these tables are having the same meaning as explained in eq (1) – eq (7), where the subscript b and Cd are used to indicate the measurement without and with cadmium cover respectively whereas the subscripts M and S are used to indicate the monitor and the sample respectively. The total uncertainties for each measurements are tabulated in the last row.

Table 6
The uncertainties in various parameters of the thermal neutron capture cross section measurements and the total uncertainties.

Parameter	Fractional uncertainty (%)				Sensitivity coefficient			
	^{96}Ru	^{102}Ru	^{104}Ru	^{105}Rh	^{96}Ru	^{102}Ru	^{104}Ru	^{105}Rh
$C_{b,S}$	4.85	4.31	10.48	6.21	6.67	1.99	4.51	4.56
$C_{b,M}$		2.97				1.20		
$C_{Cd,S}$	2.83	3.47	7.14	3.63	5.67	0.99	3.51	3.56
$C_{Cd,M}$		5.67				0.25		
$I_{\gamma,S}$	–	1.3	0.17	3.16		1		
$I_{\gamma,M}$		0.03				1		
$T_{b,S}$	1.05	0.04	0.19	0.04	6.67	1.99	4.51	4.56
$T_{b,M}$		0.0027				1.20		
$T_{Cd,S}$	0.10	0.04	0.67	0.08	5.67	0.99	3.51	3.56
$T_{Cd,M}$		0.01				0.25		
θ_S	2.53	0.44	1.45	1.45		1		
$m_{b,S}$		0.13			6.67	1.99	4.51	4.56
$m_{b,M}$		0.4				1.20		
$m_{Cd,S}$		0.13			5.67	0.99	3.51	3.56
$m_{Cd,M}$		0.4				0.25		
ε_S	2.69	2.38	2.78	2.33		1		
ε_M		3.02				1		
$\sigma_{0,M}$		0.37				1		
Total Uncertainty $\frac{\Delta X}{X} = \sqrt{\sum_i \left(S_i \frac{\Delta k_i}{k_i}\right)^2}$					37.22	10.84	53.85	31.82

Table 7
The sensitivity co-efficient and the uncertainties after multiplication with the sensitivity coefficients in each of the parameters used to estimate the resonance integral. The total uncertainty is also tabulated in the last line.

Parameter	Fractional uncertainty (%)				Sensitivity coefficient			
	^{96}Ru	^{102}Ru	^{104}Ru	^{105}Rh	^{96}Ru	^{102}Ru	^{104}Ru	^{105}Rh
$C_{b,S}$	4.85	4.31	10.48	6.21	0.10	0.11	0.10	0.10
$C_{b,M}$	2.97				0.02	0.06	0.03	0.03
$C_{Cd,S}$	2.83	3.47	7.14	3.63	0.90	0.89	0.90	0.90
$C_{Cd,M}$	5.67				0.93	0.88	0.92	0.92
$m_{b,S}$	0.13				0.10	0.11	0.10	0.10
$m_{b,M}$	0.4				0. 02	0.07	0. 03	0.03
$m_{Cd,S}$	0.13				0.90	0.89	0.90	0.90
$m_{Cd,M}$	0.4				0. 98	0.93	0. 97	0.97
θ_S	2.53	0.44	1.45	1.45	1			
ε_S	2.69	2.38	2.78	2.33	1			
ε_M	3.02				1			
$I_{\gamma,S}$	–	1.3	0.17	3.16	1			
$I_{\gamma,M}$	0.03				1			
$\sigma_{0,M}$	0.37				0.26	0.21	0.25	0.25
$I_{0,M}$	3.73				1.26	1.21	1.25	1.25
$\bar{E}_{r,M}$	11				0.10	0.09	0.10	0.10
$\bar{E}_{r,S}$	2.06	2.15	2.04	2.04	0.14	0.13	0.14	0.14
α	4.72				−0.24	−1.8e-5	−0.17	−0.17
$T_{b,s}$	1.05	0.04	0.19	0.04	0.10	0.11	0.10	0.10
$T_{b,M}$	0.0027				0.02	0.07	0.03	0.03
$T_{Cd,S}$	0.10	0.04	0.67	0.08	0.90	0.89	0.91	0.90
$T_{Cd,M}$	0.01				0.98	0.93	0.97	0.97
Total Uncertainty					9.01	8.54	10.61	9.37
$\frac{\Delta X}{X} = \sqrt{\sum_i \left(S_i \frac{\Delta k_i}{k_i}\right)^2}$								

Table 8
The thermal neutron capture cross section and the resonance integral measured in the present study.

	^{96}Ru	^{102}Ru	^{104}Ru
Thermal neutron capture cross section$\sigma_{0,S}$ (b)	0.28 ± 0.10	1.33 ± 0.14	0.48 ± 0.13
Resonance Integral $I_{0,S}$(b)	4.21 ± 0.38	4.58 ± 0.39	4.96 ± 0.35

It can be seen that the total uncertainty in the $\sigma_0$is under estimated if the sensitivity coefficients are not considered in the calculation. All parameters have $S_i = 1$ except for the counts, time factor and the mass of the sample. The sensitivity coefficient for the measurement of ^{96}Ru(n,γ) ^{97}Ru, ^{104}Ru(n,γ) ^{105}Ru and ^{104}Ru(n,γ) ^{105}Rh reactions are very high and thus contributes highly to the total uncertainty. It is interesting to note that the reaction rates R and R_{Cd} of these reactions are comparatively closer leading to the smaller values of $R - R_{Cd}$.However, the situation is different in the resonance integral estimation, where the ratio of R and R_{Cd} are utilised (eq. (6)). In R and R_{Cd} the parameters that differs are the C, T and m (C_b, T_b and m_b) and thus the smaller values of $R - R_{Cd}$ is contributing to higher sensitivity coefficient to these parameters. The higher sensitivity of counts, time factor and the mass can be reduced by the selection of proper irradiation time, decay time and counting time such that the reaction rate obtained from the bare and the cadmium covered samples are sufficiently different. However, in a low flux facility like in the present study, irradiation time, counting time etc. cannot be reduced drastically when the abundance and the gamma yield of the target and the product are small (^{96}Ru and ^{104}Ru). Similarly, it cannot be further increased when the half-life of the product is small (example ^{104}Ru (n,γ) ^{105}Ru). One possibility is to reduce the fractional uncertainty even if the sensitivity coefficient is higher. The fractional uncertainty in the counts can be reduced in the present experiment by increasing the amount of irradiated sample, but the self-shielding effects and gamma attenuation will also be increased. Due to all the above reasons, the total uncertainty is higher in the present measurements compared to the reactor based older measurements.

In Table 7, most of the sensitivity coefficients are deviated from unity. The sensitivity coefficient for the $I_{0,M}$ is greater than unity and thus contribute significantly to the total uncertainty. It can be seen that the major contributions towards total uncertainty is the counting statistics of the cadmium covered sample as well as the $I_{0,M}$. Similarly, the sensitivity coefficient of epithermal shaping factor depends on the value of $\bar{E}_r$ i.e, lower contribution to total uncertainty when $\bar{E}_r$ is smaller.

The thermal neutron capture cross sections and the resonance integrals estimated in the present measurements are tabulated in Table 8. The following observations are made by comparing the cross section measured in the present study and the previous literature values tabulated in Table 1. The $\sigma_{0,S}$estimated of ^{96}Ru in the present study is 0.28 ± 0.10 b and is in good agreement with the evaluated data. Similarly, the present value is in accordance with the measurement by Halperin et al. (Halperin and Druschel, 1965). It is greater than the recent measurements by Krane (2010) and Arbocco (Arboccò et al., 2014) however in agreement if the large error bar is considered. The neutron capture cross section of ^{102}Ru measured in the present study is 1.33 ± 0.14 b and it is well within the range of other measurements; however more close to the evaluations than the measurements. The $\sigma_{0,S}$estimated is smaller than JENDL data (Shibata et al., 2011); but higher than all other evaluations by ≈ 5% and in good comparison with the measurement by Heft (1978). However it is higher than the recent measurements which are not in agreement with each other either (Krane, 2010; Arboccò et al., 2014). The thermal neutron capture cross section and resonance integrals of ^{104}Ru are measured from the ^{105}Ru activity produced as well as the ^{105}Rh activity produced. The σ_0 estimated from the ^{105}Ru measurement is 0.477 ± 0.257 b and estimated from the beta decay product of ^{105}Ru; i.e., ^{105}Rh are 0.479 ± 0.152b with a mean value of 0.48 ± 0.13 b. The present results are in agreement with each other as well as comparable with previous measurements and evaluations.

The resonance integrals measured in the present study are compared with the literature values listed in Table 2 and the following

observations are obtained. The uncertainty in the counting statistics in the present measurements are higher; however, the total uncertainties in the measured $I_{0,S}$ are comparable with reactor beam measurements. This is due to the fact that many of the parameters (especially the counting statistics, effective resonance energy etc) in the resonance integral estimation are having sensitivity coefficient smaller than unity and thus contribute less to the final uncertainty. The $I_{0,S}$ of ^{102}Ru measured in the present study is 4.58 ± 0.39 b. The measured resonance integral is in agreement with all other previous measurements, but smaller than evaluated data. However, the agreement is not good between the present measurements and various measurements/evaluations in the case of ^{96}Ru and ^{104}Ru. This is particularly true for ^{96}Ru (4.21 ± 0.38 b). The $I_{0,S}$ estimated from the ^{105}Ru measurement is 4.92 ± 0.52 b and estimated from the ^{105}Rh is5.00 ± 0.47 b with a mean value of 4.96 ± 0.35. It is smaller than the evaluations, but is in agreement within the uncertainty with older measurements (Lantz, 1965; Ricabarra et al., 1969).

The variations in the resonance integral between various measurements can be attributed to the differences in the epithermal spectra of various reactors (Krane, 2010). As explained in eq. (5) and eq. (6), the resonance integral I_0 is derived based on the assumption that the epithermal neutron spectra is an ideal spectrum (varies as $1/E$). However, the real spectrum deviates from this $1/E$ behaviour by a factor α known as epithermal spectrum shaping factor (varies as $1/E^{(1+\alpha)}$). The parameter α depends on the irradiation facility and the accuracy of α is very crucial in the resonance integral estimation. In the present study, the epithermal shaping factor is estimated by fitting the unfolded neutron spectrum after multiple foil activation. Due to the comparatively lower flux of the neutron source, only 10 foils were used to neutron spectrum unfolding with satisfactory counting statistics (Panikkath and Mohanakrishnan, 2016, 2017). It can be seen from Table 7 that the sensitivity of the shaping parameter α is negligible in the case of ^{102}Ru whereas it is highly sensitive in the case of ^{96}Ru and ^{104}Ru. This is due to their higher Er values. Thus the smaller values of present estimates of resonance integrals ^{96}Ru and ^{104}Ru can be due to the discrepancy in the α estimation due to the uncertainty in unfolding and fitting.

The variation in the thermal neutron flux will not affect the thermal neutron capture cross section since the inclusion of Westcott's factor makes the cross section $1/v$ dependent; thus any change in the spectrum from Maxwellian shape will be cancelled out by taking the reference method. But, due the variation of distribution of resonances energies in the epithermal region, the resonance integral measurement depends on the energy spectrum even though the use of reference helps to reduce the error of measurement.

6. Conclusion

The present study demonstrates the thermal neutron capture cross section and resonance integrals of ^{96}Ru, ^{102}Ru and ^{104}Ru with reference to ^{55}Mn using the neutron flux from an Am–Be neutron source kept inside a thick concrete bunker.

The presently estimated σ_0 of ^{96}Ru, ^{102}Ru and ^{104}Ru with reference to ^{55}Mn are 0.28 ± 0.10 b, 1.33 ± 0.14 b and 0.48 ± 0.14 b respectively. These results are more comparable with evaluations than the measurements. The various σ_0 of ^{104}Ru including the one from the present study are already in good agreement with each other as well as with the evaluations. The uncertainty is high for ^{96}Ru and ^{104}Ru due to the large sensitivity coefficient as well as the fractional uncertainty of gamma counts. This can be reduced to some extend by increasing the weight of the sample under measurement. Even then it is expected be an inherent issue in a low flux neutron irradiation facility when the abundance, half-life and the gamma yield are small.

The presently estimated I_0 of ^{96}Ru, ^{102}Ru and ^{104}Ru with reference to ^{55}Mn are 4.21 ± 38 b, 4.58 ± 0.39 b and 4.96 ± 0.35 b respectively. The Io estimated for ^{102}Ru is in good agreement with various measurements and but smaller than the evaluations. However the present results are not in good agreement with various literature values for ^{96}Ru and ^{104}Ru, which requires further measurements since the existing measurements are having discrepancy among them as well as with the evaluations. One possible reason for the variations in the resonance integral between various measurements can be attributed to the differences in the epithermal spectra.

In particular, the $\sigma_{0,S}$, and $I_{0,S}$ of ^{104}Ru are measured from the activity produced due to the capture product ^{105}Ru as well as from its beta decay product ^{105}Rh separately. The σ_0 estimated from the ^{105}Ru measurement is 0.477 ± 0.257b and estimated from the ^{105}Rh is 0.479 ± 0.152 b. Similarly, the I_0 estimated from the ^{105}Ru measurement is 4.92 ± 0.52 b and estimated from the ^{105}Rh is 5.00 ± 0.47b. Thus, the estimates from two different activity measurements are in good agreement. This indicates the use of beta decay product of the neutron capture product in estimating the cross sections is satisfactory especially while the latter is a short lived nuclei.

The uncertainty analysis that followed in this study reveals that the error propagation using a quadratic sum formula is not suitable in the case of thermal neutron cross section and resonance integral analysis. Many parameters are having sensitivity coefficient different from unity and which will affect the final uncertainty. Thus a detailed sensitivity coefficient approach for all parameters is preferred here.

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