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Cross section measurements of a few threshold reactions induced by fast neutrons from an Am/Be source: integral tests of differential neutron reaction cross section data

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Summary. Cross sections of the reactions ^{24}Mg(n, p)^{24}Na, ^{27}Al(n, p)^{27}Mg, ^{27}Al(n, α)^{24}Na and ^{95}Mo(n, p)^{95}Nb were measured for the first time with fast neutrons ($E_n > 1.5$ MeV) from an Am/Be source. Furthermore, upper cross section limits for the reactions ^{98}Mo(n, α)^{95}Zr and ^{103}Rh(n, p)^{103}Ru were deduced. The results were compared with data calculated using the neutron spectral distribution and the excitation function of each reaction given in the data files and libraries ENDF/B-VII, JEFF-3.1.2, JENDL-4.0, IRDF-2004, TENDL-2010, ROSFOND-2010 and CENDL-3.1. In general, the integral measurement and the integrated value agreed within ±10%. The fast neutron field (with $E_n > 1.5$ MeV) of the Am/Be source is thus well suited for integral tests of excitation functions of neutron threshold reactions.

1. Introduction

The Rajshahi University (Bangladesh) recently procured an Am/Be neutron source for teaching and research activities in its Department of Applied Chemistry and Chemical Engineering. The radioactivity of the α-emitting radionuclide ^{241}Am ($T_{1/2} = 433$ a) was estimated on 1 October 2008 to be 105 GBq and the neutron yield to be 7.5×10^6 ns^{-1}. The detailed characteristics of the source have been described in a recent publication [1] which also contains an analysis of the neutron energy spectrum and the neutron flux available at various irradiation positions around the source. Furthermore, it was shown that the shape of the neutron spectrum is well defined over the energy range of 1.5 to 11.0 MeV, so that such a source could be conveniently used for integral tests of excitation functions of some known neutron threshold reactions. This was indeed done in the case of a few (n, p) reactions on medium mass target nuclei [1].

In the present study a few more neutron threshold reactions were chosen and their cross sections, averaged over the Am/Be neutron spectrum, were experimentally determined. The integral data obtained were then used to test the excitation functions of those reactions, as given in several data files.

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2. Experimental

2.1 Sample preparation for irradiations

The materials used in irradiations, their purities, and the reaction products studied are given in Table 1. The decay data used in nuclear reaction cross section measurements were taken from literature [2]; they are also given in Table 1. The elements Al, Ni and Mo were available in the form of sheets of thicknesses 550, 125 and 250 μm, respectively. Samples for irradiations were prepared by cutting discs of 2 cm diameter. The respective weights of the Al, Ni and Mo discs amounted to 460, 347 and 835 mg, respectively. The Rh sample was prepared by spreading 270 mg of $RhCl_3$ on a 2 cm diameter paper to form a circle of 1.7 cm diameter; it was then covered by cellulose tape. The Mg sample was prepared by packing 835 mg of the sponge in a polyethylene bag of dimensions 2 cm × 2 cm.

2.2 Irradiations

Irradiations with neutrons were carried out in three different ways, depending on the reaction to be studied. All irradiations were done in Position II of the source schematically given in Ref. [1], *i.e.* at the position where the fast neutron flux is the highest.

2.2.1 Study of the short-lived product ^{27}Mg

Due to the short half-life of ^{27}Mg ($T_{1/2} = 9.5$ min) an Al disc was irradiated alone for 24 h and the counting was done immediately at Rajshahi. The ratio of the count rates of the two products, namely ^{27}Mg and ^{24}Na ($T_{1/2} = 15.0$ h), was then determined through a simple decay curve analysis. A total of three such measurements were performed.

Table 1. Materials irradiated, activation products studied and decay data used.

Sample	Purity (supplier)	Nuclear reaction	Q-value (MeV)	Threshold energy (MeV)	Reaction threshold (MeV)	$T_{1/2}$	Decay data of the activation product used			
							E_β (keV)	I_β (%)	E_γ (keV)	I_γ (%)
Mg powder	> 99.0% (Aldrich)	^{24}Mg(n, p)^{24}Na	−4.732	4.92	5.00	15.0 h			1368.0	100.0
Al foil	99.9% (Goodfellow)	^{27}Al(n, p)^{27}Mg	−1.827	1.89	2.40	9.5 min	1670	100		
		^{27}Al(n, α)^{24}Na	−3.131	3.24	5.20	15.0 h	1390	100	1368.0	100.0
Mo foil	99.9% (Goodfellow)	^{92}Mo(n, p)^{92m}Nb [a]	+0.288	0.0	2.25	10.15 d			934.5	99.0
		^{95}Mo(n, p)^{95}Nb	−0.144	0.15	5.80	34.97 d			765.8	99.8
		^{98}Mo(n, α)^{95}Zr	+3.194	0.0		64.02 d			756.8	54.5
$RhCl_3$ powder	99.98% (Aldrich)	^{103}Rh(n, p)^{103}Ru	+0.019	0.0		39.25 d			497.0	88.7
Ni foil	99.98% (Goodfellow)	^{58}Ni(n, p)^{58}Co [b]	+0.402	0.0	1.50	70.86 d			811.0	99.0

a: This reaction served as an internal monitor in measurement of cross sections of other reactions induced on isotopes of Mo.
b: This was the monitor reaction for determining the neutron flux density.

2.2.2 Measurements on longer lived products

For measurements on two nuclear reactions leading to the same product ^{24}Na ($T_{1/2} = 15.0$ h), two stacks of samples, each consisting of Ni-Mg-Al-Ni, were irradiated for 47 h. They were then transported to the Institute of Nuclear Science and Technology (INST), Savar, Dhaka, where γ-ray spectrometric analysis was carried out. In each stack the Ni foils served as neutron flux monitors by virtue of the well-known excitation function of the ^{58}Ni(n, p)^{58}Co reaction [*cf*. 3].

2.2.3 Investigations on long-lived products

Many of the investigated products are long-lived. To study them, two stacks, each consisting of Ni-$RhCl_3$-Mo-Ni, were irradiated for about 400 h. Thereafter they were transferred to Forschungszentrum Jülich, Germany, where low-level γ-ray spectrometric measurements were carried out.

2.3 Measurement of radioactivity

The radioactivity of the short-lived ^{27}Mg (formed by the ^{27}Al(n, p)-reaction) was determined relative to that of ^{24}Na (formed through the ^{27}Al(n, α)-reaction) in the same target. The irradiated Al disc was subjected to gross β^- counting using a Geiger counter, Type Alpha ix 3000, INNOVA (Germany), counting tube G (open window). Counting was done periodically for two days and the decay curve was analysed in two components, one with a half-life of 9.5 min (^{27}Mg) and the other with a half-life of 15.0 h (^{24}Na). The count rates were extrapolated to the end of bombardment (EOB) and therefrom the ratio of the count rate of ^{27}Mg to that of ^{24}Na was obtained. Since the intensity of the β^- particle emission from both the nuclides is the same and since the endpoint energies of the β^- particles in both the cases are approximately equal (see Table 1), it was assumed that, except for a small correction for the difference in self-absorption, the experimentally determined ratio of the count rates should represent the ratio of the decay rates of the two radionuclides as well. The difference in the efficiency of the detector for the two β^- particles was negligible because the detector efficiency for $E_{\beta^-} > 1$ MeV is almost constant. A correction of 8% was applied for the difference in the self-absorption of the two β^- particles in the Al disc; the uncertainty in this correction was assumed to be < 3%.

The radioactivity of the radionuclide ^{24}Na formed in both the Al and the Mg samples through the ^{27}Al(n, α)^{24}Na and ^{24}Mg(n, p)^{24}Na reactions, respectively, as well as of the radionuclide ^{58}Co produced in the Ni monitor foils was measured using HPGe detector (Canberra, 20% relative efficiency, 1.9 keV resolution at 1332.5 keV of ^{60}Co, coupled with ORTEC DSPEC jr™) γ-ray spectrometry at the Institute of Nuclear Science and Technology (INST), Savar, Dhaka, Bangladesh. The samples were counted at 10 cm from the detector surface to minimize the sample-size effect on the efficiency, and the random coincidence effect is negligible at this distance. Counting was carried out a few times over a period of several days to check the half-lives of the activation products. The γ-ray spectra were analysed by the software GammaVision. The details on γ-ray counting and detector efficiency measurement have been described earlier [1].

Measurement of the radioactivity of the long-lived products formed in the Mo and Rh samples, as well as in the relevant Ni-monitor foils, was carried out at the Institut für Nuklearchemie, Forschungszentrum Jülich, Germany, using a low-level HPGe detector as described earlier [4]. Due to weak activities, the samples were placed directly on the surface of the detector, where an efficiency loss of 12.6% for the extended sample had to be corrected. The analysis of the γ-ray spectra was done at Jülich also using GammaVision software (Version 6.01).

2.4 Neutron flux measurement

The ^{58}Ni(n, p)^{58}Co reaction induced in the Ni-foil was used to monitor the neutron flux effective at that foil. The two Ni-foils (one placed in front and the other at the back of the sample) were counted together to obtain the average ^{58}Co activity. By using the well-characterised part of the neutron spectrum [*cf*. 1, 5] with $E_n > 1.5$ MeV and adopting the excitation function of the ^{58}Ni(n, p)^{58}Co reaction given in the IRDF (2004) [3], the spectrum averaged cross section of this reaction was calculated as 391 mb. By using this value and the experimentally determined decay rate of the activation product ^{58}Co, the flux of neutrons above 1.5 MeV was calculated.

Table 2. Sources of uncertainties in the measured integral cross sections.

Source of uncertainty	Uncertainty (%)
Measurement of radioactivity	
Peak area analysis	2
Counting statistics	1–14
Self-absorption correction in beta counting	< 3
Coincidence loss	< 1
Efficiency of detector	5
Extended source correction	< 2
Half-life of product	< 0.1
γ-ray intensity	< 0.3
Calculation of cross section	
Sample mass	0.01
Monitor cross section	6
Total	8–16

During the measurement on the ^{27}Al$(n, p)^{27}$Mg reaction, on the other hand, no neutron flux determination was performed. The activity of ^{27}Mg was simply compared with that of ^{24}Na, and since the cross section of the ^{27}Al$(n, \alpha)^{24}$Na reaction was independently determined, the cross section of the ^{27}Al$(n, p)^{27}$Mg reaction could be calculated using ^{24}Na as an internal monitor reaction product.

2.5 Calculation of integral reaction cross sections and their uncertainties

The measured count rate of each product radionuclide was extrapolated to the end of bombardment and it was converted to decay rate by applying the usual corrections, like the intensity of the emitted γ-ray and the efficiency of the detector (except for ^{27}Mg which was treated differently as described above). From the decay rates and the flux of neutrons of energies above 1.5 MeV, the neutron spectrum averaged cross section $\langle\sigma\rangle$ of each investigated reaction, *e.g.* ^{24}Mg$(n, p)^{24}$Na, ^{27}Al$(n, \alpha)^{24}$Na and ^{95}Mo$(n, p)^{95}$Nb, was deduced by using the usual activation formula. The combined uncertainty in the experimentally determined cross section was estimated by taking the square root of the quadratic sum of the individual uncertainties as given in Table 2. The overall uncertainty in the cross section is around 8–16% (1σ). The large uncertainty contributing to the overall value originates from the neutron flux inferred from the monitor measurements and from counting statistics due to low activity of the activated product.

3. Results and discussion

3.1 Spectrum averaged cross sections

The experimentally determined integral cross sections, averaged over the fast neutron spectrum ($E_n > 1.5$ MeV), are given in Table 3. Each value is based on at least two independent measurements. The total uncertainty of each value amounts to between 8 and 16%.

For the ^{24}Mg$(n, p)^{24}$Na, ^{27}Al$(n, p)^{27}$Mg, ^{27}Al$(n, \alpha)^{24}$Na and ^{95}Mo$(n, p)^{95}$Nb reactions, cross sections could be measured with reasonable accuracies. As far as we know, these measurements have been done for the first time. With regard to the reactions ^{98}Mo$(n, \alpha)^{95}$Zr and ^{103}Rh$(n, p)^{103}$Ru, however, only an upper limit of the reaction cross section could be given, since even intensive checks, involving a sample counting time of more than one week each on the low-level detector, did not show any sign of the searched γ-ray peak. The cross section for the ^{92}Mo$(n, p)^{92m}$Nb reaction was measured previously [1]. While working on the Mo sample we re-measured the cross section of this reaction to check the validity of the present set of measurements and techniques. The present value is in good agreement with the previously determined cross section value.

Table 3. Measured neutron spectrum averaged cross sections.

Nuclear reaction	Cross section (mb)
^{24}Mg$(n, p)^{24}$Na	26 ± 3
^{27}Al$(n, p)^{27}$Mg	30 ± 5
^{27}Al$(n, \alpha)^{24}$Na	9.7 ± 1.2
^{92}Mo$(n, p)^{92m}$Nb	43 ± 4 [a]
^{95}Mo$(n, p)^{95}$Nb	1.9 ± 0.3
^{98}Mo$(n, \alpha)^{95}$Zr	< 0.5
^{103}Rh$(n, p)^{103}$Ru	< 0.3

a: This reaction cross section was re-measured to check the validity of the present techniques.

3.2 Integral tests of excitation functions

The integrated cross section for the standard Am/Be neutron spectrum [*cf.* 1, 5] was obtained by using the excitation function of each reaction reported in several evaluated data files and libraries, *e.g.* ENDF/B-VII, JEFF-3.1.2, JENDL-4.0, IRDF-2004, TENDL-2010, ROSFOND-2010 and CENDL-3.1 [3, 6–10]. Based on the uncertainties in the neutron spectrum and the excitation function, the overall uncertainty in the integrated reaction cross section was estimated to be about 6%. The results are given in Table 4.

The integrated reaction cross section from each evaluated library/file was compared with the experimentally measured integral cross section. The result $\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$ for the reaction ^{92}Mo$(n, p)^{92m}$Nb was discussed earlier [1]. For other reactions the results are given in Table 4. For the first three reactions, namely ^{24}Mg$(n, p)^{24}$Na, ^{27}Al$(n, p)^{27}$Mg and ^{27}Al$(n, \alpha)^{24}$Na, the $\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$ ratio for all the files lies between 0.90 and 1.10, showing that the agreement between the integrated data and the integrally measured data is within ±8%, except for two cases where it is within 10%. This good agreement reflects the status of the excitation functions of the three reactions considered. Since they have been experimentally well investigated (cf. EXFOR [12]), the evaluations appear to be more reliable. The present integral measurements thus validate the experimental excitation functions of those three reactions very well.

With regard to the reaction ^{95}Mo$(n, p)^{95m+g}$Nb, the $\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$ for the ENDF/B-VII, JEFF-3.1.2 and CENDL-3.1 libraries is very far from 1.00, but it is 1.10, 1.01 and 0.89 for TENDL-2010, ROSFOND-2010 and JENDL-4.0, respectively. The latter three libraries thus give better re-

Table 4. Measured and calculated neutron spectrum averaged cross sections.

Reaction	Spectrum averaged cross section $\langle\sigma\rangle$ (mb)																			
	$\langle\sigma\rangle_{meas}$	ENDF/B-VII		JEFF-3.1.2			JENDL-4.0			IRDF-2002			TENDL-2010		ROSFOND-2010			CENDL-3.1		
		$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$	$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$		$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$		$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$		$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$	$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$		$\langle\sigma\rangle_{cal}$	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$
^{24}Mg(n, p)^{24}Na	26 ± 3	24.6 ± 1.5	0.94 ± 0.12	24.6 ± 1.5	0.94 ± 0.12		24.6 ± 1.5	0.94 ± 0.12		25.5 ± 1.5	0.98 ± 0.13		23.4 ± 1.4	0.9 ± 0.11	24.6 ± 1.5	0.94 ± 0.12		24.5 ± 1.5	0.94 ± 0.12	
^{27}Al(n, p)^{27}Mg	30 ± 5	28.9 ± 1.7	0.96 ± 0.17	29.0 ± 1.7	0.97 ± 0.15		33.1 ± 1.7	1.10 ± 0.17		32.5 ± 1.9	1.08 ± 0.18		32.1 ± 1.9	1.07 ± 0.18	28.9 ± 1.7	0.96 ± 0.17		32.9 ± 1.9	1.10 ± 0.1	
^{27}Al(n, α)^{24}Na	9.7 ± 1.2	10.2 ± 0.6	1.05 ± 0.14	10.2 ± 0.6	1.02 ± 0.14		9.7 ± 0.6	1.00 ± 0.13		10.3 ± 0.6	1.06 ± 0.14		10.2 ± 0.6	1.05 ± 0.14	10.2 ± 0.6	1.05 ± 0.14		9.9 ± 0.6	1.02 ± 0.14	
^{95}Mo(n, p)^{95}Nb	1.9 ± 0.3	0.4 ± 0.02	0.21 ± 0.03	0.8 ± 0.05	0.42 ± 0.07		1.7 ± 0.1	0.89 ± 0.13					2.1 ± 0.12	1.1 ± 0.18	1.9 ± 0.11	1.01 ± 0.17		2.6 ± 0.15	1.38 ± 0.23	

sults. It should be pointed out that this reaction has not been so extensively investigated as the first three reactions discussed above. In the critical energy range of 3 to 10 MeV, for example, only measurements by Rahman and Qaim [13] are available. Thus, in evaluations, reliance is more on nuclear model calculations rather than on experimental data. The disagreement between the calculated results from the ENDF/B-VII and CENDL-3.1 and the integral measurement is possibly due to this reason. On the basis of this work, the TENDL-2010 and ROSFOND-2010 evaluations for this reaction appear to be more reliable.

Regarding the remaining two reactions given in Table 3, namely ^{98}Mo(n, α)^{95}Zr and ^{103}Rh(n, p)^{103}Ru, integral tests of excitation functions are not meaningful because the measured values depict only upper limits and the evaluated excitation functions are based on theoretical calculations rather than on experimental data.

4. Conclusion

Using the recently installed Am/Be neutron source at the Rajshahi University, neutron spectrum averaged cross sections were measured for four nuclear reactions for the first time. The fast neutron field (with $E_n > 1.5$ MeV) was found to be well suited for integral tests of excitation functions of the four investigated neutron threshold reactions.

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