Integral measurement of spectrum-averaged cross sections of a few threshold reactions induced by fast neutrons of a TRIGA reactor: comparison with integrated data from excitation functions given in various data libraries

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Summary. Integral cross sections of the reactions ^{24}Mg(n,p)^{24}Na, ^{27}Al(n,p)^{27}Mg, ^{27}Al(n,α)^{24}Na, ^{58}Ni(n,d+np)^{57}Co and ^{60}Ni(n,p)^{60}Co were measured for the first time using the fast neutron spectrum of a TRIGA reactor extending from 0.5 to 20 MeV. The values obtained in this work were comparable with the recommended values for a pure ^{235}U prompt fission spectrum. The measured integral value was utilized for integral test of excitation function of each reaction given in some data libraries, namely ENDF/B-VIII.0, TENDL-2017, IRDFF-1.05 and ROSFOND-2010. The integral measurements are generally consistent with the integrated values within 5 %, except for a few cases, e.g. the reaction ^{60}Ni(n,p)^{60}Co, where the data libraries appear to need improvement.

1. Introduction

A 3 MW TRIGA Mark II reactor has been in operation at the campus of Atomic Energy Research Establishment (AERE) of Bangladesh Atomic Energy Commission (BAEC) in Savar since 1986. A brief description of the basic characteristics of the reactor has been given [cf.1]. The shape and intensity of the neutron spectrum at a well-defined irradiation position, known as Dry Central Thimble (DCT), was characterized from the energy 0.5 to 20 MeV. This part of the neuron spectrum is interesting for practical applications, such as production of special radionuclides using (n,n′γ), (n,p) and (n,α) reactions, integral tests of excitation functions of some neutron threshold reactions, etc. [1,2]. Several general purpose data libraries have been established [3-8]. Data testing and validation by integral measurements thus appears to be timely and useful. The TRIGA neutron spectrum is somewhat higher in the energy region between 0.5 and 1.5 MeV than the pure ^{235}U prompt fission spectrum [cf.1]. There could be several reasons, e.g. difference in fuel cladding, presence of $ZrH_{1.6}$, lower enrichment of ^{235}U in TRIGA (i.e. higher concentration of ^{238}U), presence of a thick graphite plate in the vicinity of the core, which thermalizes and reflects neutrons, etc.

The major aim of the present work was to measure cross sections of a few neutron threshold reactions averaged over the fast neutron spectrum of the TRIGA reactor and to compare the results with data reported for a pure ^{235}U prompt fission neutron spectrum. Testing of data files utilizing the TRIGA reactor neutron spectrum was also aimed at. As far as we know, besides our studies, spectrum-averaged cross section measurements and data testing using a TRIGA reactor have hitherto not been reported.

2. Experimental techniques

2.1 Samples and irradiations

High purity foils of the metals Al and Ni were cut in circular discs with a diameter of 1 cm and were placed in an Al-container, and finally in an irradiation vial. The thicknesses of Al and Ni discs were 550 and 125 μm, respectively. Furthermore, a few Mg samples were prepared by pressing 97 mg of MgO (purity: 99.5 %; Aldrich) in the form of a disc with a size similar to that of the above foils. Two stacks were prepared for irradiations: one for short irradiation and the second for longer irradiation. All samples in a stack were irradiated together with neutrons at the dry central thimble (DCT) of the TRIGA reactor. The Ni and Al foils were irradiated for 40 min at the reactor power of 1 MW. A short irradiation of Al, Ni and MgO was performed for 10 min at DCT, with the reactor power again set to 1 MW. The reaction $^{58}Ni(n,p)^{58}Co$ induced in the Ni-foil was used to monitor the fast neutron flux (0.5-20 MeV) effective in the investigated samples. The details of the irradiation samples are given in Table 1.

2.2. Gamma-ray spectrometric measurements

The radioactivity induced in each irradiated sample was measured non-destructively using a high-purity germanium (HPGe) gamma-ray detector (Canberra, 20% relative efficiency, 1.9 keV resolution at 1332.6 keV ^{60}Co) associated with a digital gamma spectrometry system (ORTEC DSPEC jr ^{TM}) and Maestro data acquisition software at the Institute of Nuclear Science and Technology (INST), Savar, Dhaka, Bangladesh. The decay data were taken from the Lund/LBNL Nuclear Data [9]. Due to the short half-life of ^{27}Mg ($T_{1/2}$= 9.5 min), formed via the $^{27}Al(n,p)^{27}Mg$ reaction, the sample after a short irradiation was removed from the reactor core within 20 min after the end of bombardment (EOB). Thereafter counting was done immediately at a position of 40 cm from the detector surface to keep the dead time below 5 %.

The radioactivity of the radionuclide ^{24}Na is formed in both the Al and the Mg samples through the $^{27}Al(n,\alpha)^{24}Na$ and $^{24}Mg(n,p)^{24}Na$ reactions, respectively. The radionuclide ^{58}Co produced in the Ni-monitor foil was measured about 4 days after EOB to allow complete decay of ^{58m}Co ($T_{1/2}$= 8.94 h) to ^{58}Co. Counting was done using the HPGe detector at 20 cm from the detector surface to minimize the sample-size effect on the efficiency, where the random coincidence loss is also negligible. Measurement was carried out several times giving sufficient interval over a period of several days to check the half-lives of the relatively short-lived activation products. The γ-ray spectra were analysed by both the software GammaVision, FitzPeaks and Hypermet PC 5.12. The details on γ-ray counting and detector efficiency measurement have been described earlier [cf. 1].

Measurement of the radioactivity of the longer lived radionuclides ^{60}Co and ^{57}Co produced in the Ni foils was carried out at the same HPGe γ-ray detector but, due to weak activities, the samples

were placed directly on the surface of the detector, where an efficiency loss of 2% for the extended sample had to be corrected [10].

The efficiency versus energy curve of the HPGe gamma-ray detector was determined using the standard point sources ^{57}Co, ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu (supplied by North American Scientific).

2.3 Fast neutron flux

The effective total fast neutron flux for energies above 0.5 MeV was determined using the monitor reaction ^{58}Ni(n,p)^{58}Co induced in the Ni-foil [1,2]. The spectrum averaged cross section of the monitor reaction was calculated by using the well-characterized part of the neutron spectrum at the irradiation position in the core of the reactor and adopting the excitation function of the ^{58}Ni(n,p)^{58}Co reaction from IRDFF-1.05 library [3], which amounted to 91.2 ± 5.5 mb. From this cross section value and the measured ^{58}Co radioactivity, the fast neutron flux above 0.5 MeV was calculated and the obtained value amounted to 8.91×10^{12} ncm^{-2}s^{-1} at the reactor power of 1 MW. The flux in the short irradiation was 6.54×10^{12} ncm^{-2}s^{-1}.

2.4 Determination of integral cross sections

The measured γ-ray count rate of each activation product was extrapolated to EOB and then converted to decay rate by applying the usual corrections, like the intensity of the characteristic γ-ray emitted in the decay of the investigated radionuclide and the efficiency of the detector. From the decay rates and the neutron flux (>0.5 MeV) effective at the sample, the spectrum-averaged cross sections of the reactions ^{24}Mg(n,p)^{24}Na, ^{27}Al(n,p)^{27}Mg, ^{27}Al(n,α)^{24}Na, ^{58}Ni(n,d+np)^{57}Co and ^{60}Ni(n,p)^{60}Co were determined. The overall uncertainty associated with the measured cross section was estimated by taking the square root of the quadratic sum of the individual uncertainties: uncertainty in efficiency (4%), counting statistics (0.5-5 %), peak area analysis (2%), half-life of the product (<0.1%), γ-ray intensity (<0.2%), sample mass (0.01%) and uncertainty in monitor reaction cross section (6%). The random and real coincidence losses were negligible. The overall uncertainties in the measured cross sections were in the range of 7-10 %.

3. Results and discussion

3.1 Spectrum averaged cross sections

The experimentally determined integral cross sections, averaged over the fast neutron spectrum (E_n > 0.5 MeV) in the TRIGA reactor, are given in Table 2. The total uncertainty of each value is also quoted in the same table. The reported results have reasonable accuracies. As far we know, these measurements have been done for the first time. In a previous work [1] we had reported the data for the ^{27}Al(n,α)^{24}Na reaction. The present measurement agrees with the previously determined cross section value and gives more confidence to the present data.

With regard to the formation of ^{57}Co, there are three channels, two direct production reactions are the ^{58}Ni(n,np) and ^{58}Ni(n,d), and the third is the decay of ^{57}Ni ($T_{1/2}$ = 35.6 h) produced in the reaction ^{58}Ni(n,2n)^{57}Ni (E_{thr} = 12.4 MeV). However, even an intensive check, involving a long sample counting did not show any sign of the 1378 keV characteristic γ-ray of ^{57}Ni and the cross section limit for the formation of ^{57}Ni was placed at ~0.004 mb, which indicates that the

contribution of the (n,2n) reaction to the formation of ^{57}Co is negligible. Therefore, the measured cross section gives the sum of the ^{58}Ni(n,np)^{57}Co and ^{58}Ni(n,d)^{57}Co reactions.

3.2 Comparison of the measured integral data with the recommended values for the pure ^{235}U fission spectrum

A comparison of the integral data measured in this work was done with the recommended values for the pure ^{235}U fission spectrum (cf. Calamand, [11]). The agreement is generally within 3 %. For the reaction ^{27}Al(n,p)^{27}Mg, our measured value is 5 % higher than the Calamand value [11]. This deviation may be due to slightly larger uncertainty associated with the recommended value for the pure fission spectrum. Calamand [11] did not report any recommended value for the reaction ^{58}Ni(n,d+np)^{57}Co. It should, however, be mentioned that the Calamand data are for the neutron spectrum with zero as the lower boundary whereas our data refer to neutrons above 0.5 MeV. The relatively good agreement between Calamand data and our data is thus only incidental. The neglect of low energy neutrons in our work seems to be compensated by the higher neutron flux in the 0.5 to 1.5 MeV energy region of the TRIGA reactor than that of the pure ^{235}U fission spectrum (see ref.[1]).

3.3 Comparison of the measured integral data with the integrated values from the evaluated data files

The spectrum–integrated cross sections from the known excitation functions given in a few evaluated data libraries (ENDF/B-VIII.0; IRDFF-1.05; ROSFOND-2010) and a theory-based file (TENDL-2017) were compared with our measured integral cross sections. The ratio of calculated/measured integral cross section for each reaction is given in Table 2.

For the reactions ^{24}Mg(n,p)^{24}Na and ^{27}Al(n,α)^{24}Na the $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratios for all the libraries are close to 1.00, with a maximum deviation of 6 %, showing good agreement between the integrated data and the integrally measured data. For the reaction, ^{60}Ni(n,p)^{60}Co the $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratio for all the libraries lies between 0.80 and 0.91. We also calculated the integral value for this reaction from the available experimental data [12-20] compiled in EXFOR [21]. An excitation function was obtained by a polynomial fitting of the experimental data sets shown in Fig.1. A few data points in the range of 8 – 10.5 MeV reported by Paulsen and Liskien [12] were not considered in data fitting, because those data points were shown by the other authors [13-15] to deviate from the common trend. The ratio of the integrated value obtained from the fitted experimental curve to the integrally measured one amounts to 0.98. As well known, TENDL is a purely calculated file and the results occasionally deviate from the experimental data. The other three data files, on the other hand, are much above the experimental points in the neutron energy range above 13 MeV (Fig. 1). Our present results show that those evaluated data files need improvement.

With regard to the reaction ^{27}Al(n,p)^{27}Mg, the $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratio lies between 0.80 and 0.86, except for the IRDFF-1.05 (ratio 0.97). It should be pointed out that this reaction has many resonances below 10 MeV (cf. Fig. 2). They are more visible in IRDFF-1.05 and to some extent in TENDL2017. The other libraries ignore them. It was thus not possible to collect data points precisely for integration. Furthermore, in the energy range of 7 to 14 MeV, the excitation functions of the TENDL2017 and IRDFF-1.05 libraries are significantly higher in comparison to the libraries ENDF/B-VIII.0 and ROSFOND-10. The large deviations of $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratios

for data from several libraries are attributed to the uncertainty in the integration of the cross section in the region of the excitation function with several resonances as well as to the significant difference in the cross section values between 9 and 13 MeV.

The agreement between the integral value measured for the reaction ^{58}Ni(n,d+np)^{57}Co and the integrated values of both ENDF/B-VIII.0 and ROSFOND-2010 is quite good (see Table 2). In the case of TENDL-2017, the $<\sigma>_{cal.}/<\sigma>_{meas.}$ ratio amounted to 1.41 and it is not acceptable. As shown in Fig. 3, the deviation originates from the shift of the excitation function to lower energies. For comparison with our measurement, however, only the summed cross section is of relevance.

The formation of ^{57}Co in the interactions of neutrons with ^{58}Ni needs special consideration. As discussed above (section 3.1), the measured cross section gives a sum of the ^{58}Ni(n,np)^{57}Co and ^{58}Ni(n,d)^{57}Co reactions. The measured summed cross sections over broader energy ranges [16-18] are shown in Fig. 3. Some other individual data points around 14 MeV are not shown but they agree with the values given in Fig. 3. The exceptionally high cross section for the (n,np) process was interpreted in terms of the rather high separation energy of the second chance neutron in comparison to the proton separation energy [19]. The curves from the three data files are also shown in Fig. 3. The purely (n,d) cross section was determined by Qaim and Wölfle [20] through activation measurements below the threshold of the (n,np) reaction. They gave an eye-guide curve by combining their results with those from a spectral measurement by Grimes et al. [22]. Those data and the trend are also shown in Fig. 3 and compared with the values in three files.

4. Conclusions

For the reactions ^{24}Mg(n,p)^{24}Na, ^{27}Al(n,p)^{27}Mg, ^{27}Al(n,α)^{24}Na, ^{58}Ni(n,d+np)^{57}Co and ^{60}Ni(n,p)^{60}Co integral cross sections over fast neutron spectrum from the energy 0.5 to 20 MeV were measured for the first time using a TRIGA reactor. From both the present and previous measurements [1] at a TRIGA reactor it is observed that the measured integral cross sections of the reactions having thresholds above 2 MeV incidentally agree within about 4 % with the recommended values for the pure fission spectrum, whereas the measured values below 2 MeV significantly deviate (8-15%) from those for the pure fission spectrum. This is possibly due to the difference between the TRIGA neutron spectrum and the pure fission spectrum below about 2 MeV.

Except for a few investigated reactions, the cross sections integrated by using the unfolded TRIGA spectrum and the known excitation functions from data libraries are consistent with the integrally measured values. In general, it is therefore concluded that in case of well-investigated neutron threshold reactions, the TRIGA Mark II reactor neutron field above 0.5 MeV can be used for validation of evaluated data. Regarding the ^{60}Ni(n,p)^{60}Co reaction, the evaluated data files appear to need improvement.

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Table 1

Irradiated samples and investigated activation products.

Sample	Purity (Supplier)	Nuclear reaction	Q-value (MeV)	Threshold energy (MeV)	Reaction threshold (MeV)	Decay data of the activation product[b]		
						$T_{1/2}$	E_γ(keV)	I_γ (%)
MgO	99.5 % (Aldrich)	$^{24}Mg(n,p)^{24}Na$	-4.73	4.93	6.0	14.96 h	1368.6	100
Al[a]	99.9 % (Goodfellow)	$^{27}Al(n,\alpha)^{24}Na$	-3.13	4.60	6.2	14.96 h	1368.6	100
		$^{27}Al(n,p)^{27}Mg$	-1.83	1.89	2.7	9.46 min	843.7	71.8
Ni[a]	99.98% (Chempur)	$^{60}Ni(n,p)^{60}Co$	-2.04	2.07	4.2	5.2714 y	1173.2	99.97
							1332.4	99.98
		$^{58}Ni(n,d+np)^{57}Co$[c]	-8.17	8.31	9.0	271.79 d	122.0	85.60
							136.5	10.68
		$^{58}Ni(n,p)^{58}Co$	0.40	0.0	0.5	70.86 d	810.7	99.0

[a] As foil.

[b] Taken from Lund/LBNL Nuclear Data [9].

[C] In case of d-emission the Q-value is -5.95 MeV, the threshold energy 6.05 MeV and the reaction threshold 8 MeV.

Table 2. Measured and calculated neutron spectrum averaged cross sections [a)]

	$\langle\sigma\rangle_{meas}$ (This work) mb	$\langle\sigma\rangle_{Recom}$ (Calamand) mb	ENDF/B-VIII.0		IRDFF-1.05		TENDL-2017		ROSFOND-2010		EXFOR	
			$\langle\sigma\rangle_{cal}$ mb	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$	$\langle\sigma\rangle_{cal}$ mb	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$	$\langle\sigma\rangle_{cal}$ mb	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$	$\langle\sigma\rangle_{cal}$ mb	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$	$\langle\sigma\rangle_{cal}$ mb	$\langle\sigma\rangle_{cal}/\langle\sigma\rangle_{meas}$
$^{24}Mg(n,p)^{24}Na$	1.57±0.09	1.53±0.09	1.58	1.01	1.47	0.94	1.50	0.96	1.57	1.00		
$^{27}Al(n,\alpha)^{24}Na$	0.71±0.06	0.73±0.05	0.71	1.00	0.70	0.99	0.67	0.94	0.67	0.94		
$^{27}Al(n,p)^{27}Mg$	4.20±0.25	4.00±0.45	3.35	0.80	4.07	0.97	3.61	0.86	3.40	0.81		
$^{60}Ni(n,p)^{60}Co$	2.20±0.12	2.30±0.40	1.87	0.85	1.99 [b)]	0.90	2.0	0.91	2.0	0.91	2.16 [c)]	0.98
$^{58}Ni(n,d+np)^{57}Co$	0.29±0.014		0.30	1.03			0.41	1.41	0.30	1.03		

a) For the neutron field from 0.5 to 20 MeV in a TRIGA reactor, except for Calamand values [11] which are for pure fission neutron spectrum.

b) The integrated value for the PFNS given in IRDFF-1.05 website is slightly higher (2.17 mb).

c) This $\langle\sigma\rangle$ value is deduced from the selected experimental data of Paulsen and Liskien [12] and all data of Sudár et al. [13], Wagner et al.[14] and Vonach et al.[15]. It is in good agreement with our measured value of 2.20±0.12 mb.

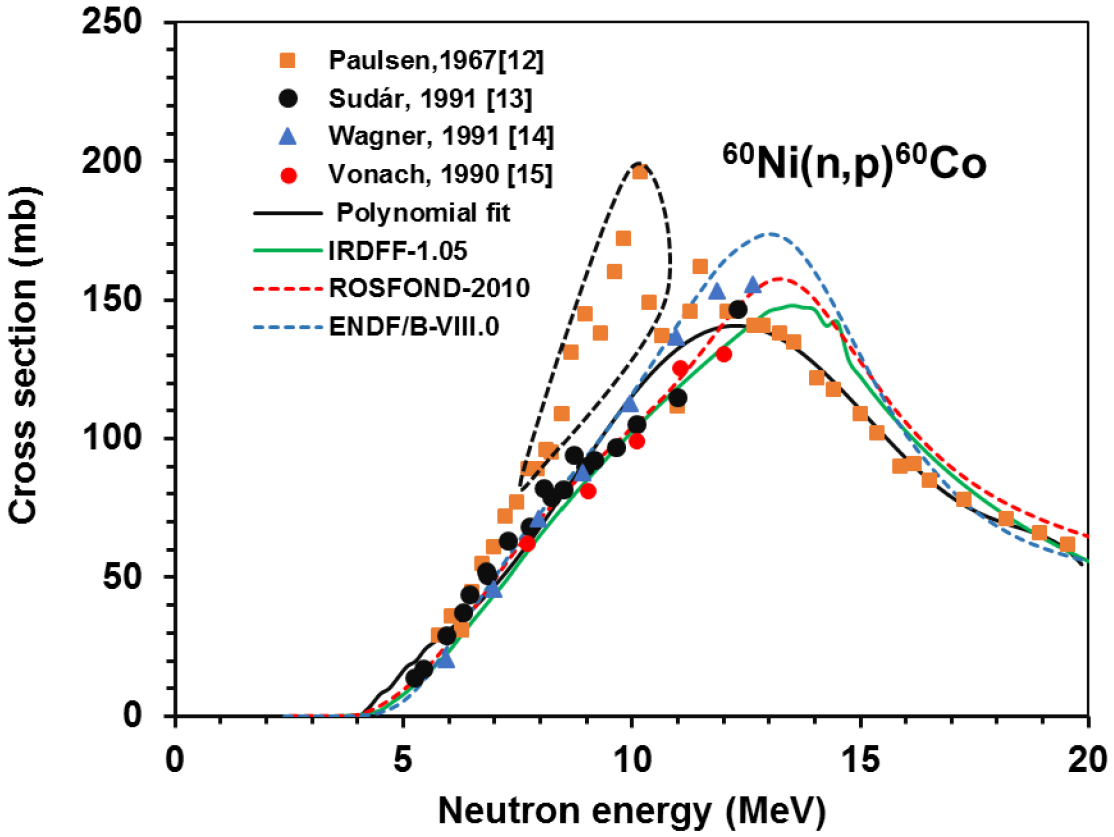

Fig. 1 Excitation function of the ^{60}Ni(n,p)^{60}Co reaction, obtained by neglecting the encirculated points and fitting of the concordant experimental values by a polynomial function. The curves in the libraries IRDFF-1.05, ROSFOND-2010 and ENDF/B-VIII.0 are also shown.

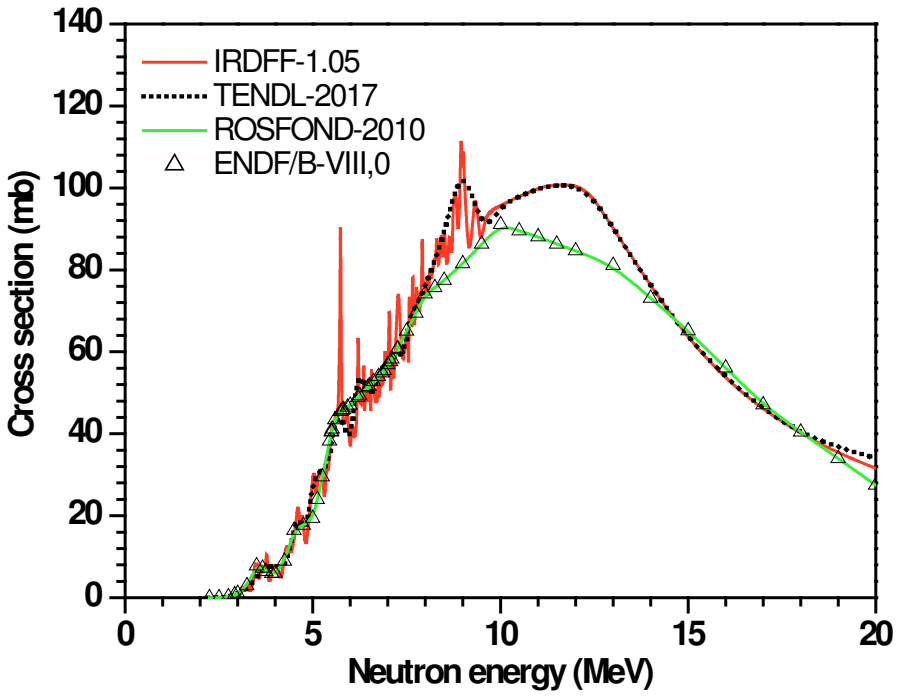

Fig. 2 Excitation function of the ^{27}Al(n,p)^{27}Mg reaction, described in various data libraries.

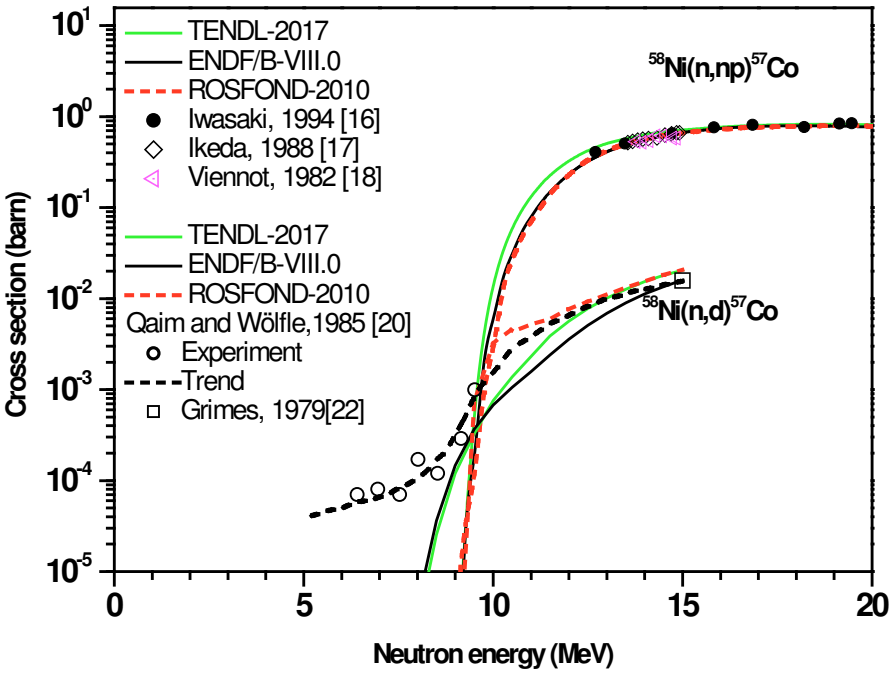

Fig. 3 Excitation functions of the ^{58}Ni(n,np)^{57}Co and ^{58}Ni(n,d)^{57}Co reactions. The (n,d) cross section data below the reaction threshold were interpreted in terms of tunnel effect [20].