Measurement of spectrum-averaged cross sections of the (n,p) and (n,n') reactions on strontium by fast neutrons of a TRIGA reactor: comparison with integrated data from excitation functions of various data libraries

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Summary. Integral cross sections of the reactions ^{84}Sr(n,p)^{84}Rb, ^{86}Sr(n,p)^{86}Rb and ^{87}Sr(n,n')^{87m}Sr, over the fast neutron spectrum of a TRIGA reactor (0.5 to 20 MeV), were measured for the first time. The measured data were compared with the values for a pure ^{235}U fission spectrum reported by Calamand. The results of this work were useful for integral test of the evaluated excitation functions in the data libraries, ENDF/B-VIII.0, ROSFOND-2010, JEFF-3.3, JENDL-5 and TENDL-2021. The integral measurements are generally consistent with the integrated values within 12 %, except for a few cases. The two data libraries ENDF/B-VIII.0 and ROSFOND-2010 appear to need improvement with regard to the reactions investigated in this work.

Keywords: TRIGA Mark II reactor/Fission spectrum / Nuclear reaction / Spectrum- averaged cross section / Integral test of differential data

1. Introduction

A 3 MW TRIGA Mark II reactor was installed on the campus of Atomic Energy Research Establishment (AERE), Savar, in 1986. Its basic characteristics were described in detail [cf.1]. The fast neutron spectrum at the core of the reactor, known as Dry Central Thimble (DCT), was

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well characterized by multiple foil activation and unfolding over the energy range of 0.5 to 20 MeV [1]. This part of the neuron spectrum is interesting for production of special radionuclides with high specific activity, for radiation safety analysis, for integral tests of excitation functions of some neutron threshold reactions, etc. [1-3]. Compared to radiative neutron capture, the (n,n'), (n,p) and (n,α) reactions are usually unimportant for isotope production because of their relatively low cross sections and the relatively low flux of higher-energy neutrons at the general irradiation facilities of a TRIGA reactor. However, these nuclear reactions may contribute appreciably to the yield of radioisotopes in an irradiation in the core of the reactor where the integral fast-neutron flux is comparable to the thermal-neutron flux.

In view of the above considerations, we planned to measure the integral cross sections of the ^{84}Sr(n,p)^{84}Rb, ^{86}Sr(n,p)^{86}Rb and ^{87}Sr(n,n')^{87m}Sr nuclear reactions in the mixed radiation field of the TRIGA reactor, and to compare the results with data reported for a pure ^{235}U prompt fission neutron spectrum [4]. Calamand [4] reported a recommended value for the above-mentioned (n,n'γ) reaction and estimated values with large uncertainties for the (n,p) reactions. Due to its relatively small cross sections and low isotopic abundance of ^{84}Sr (0.56 %), the process ^{84}Sr(n,p)^{84}Rb is difficult to investigate experimentally. For the (n,n') process the cross section is usually large but there is another difficulty; the (n,γ) process dominates in the formation of ^{87m}Sr while using a natural strontium target and a clean analysis of this contribution is needed.

Several groups have reported experimental neutron-induced reaction cross section data, but those are limited over a small energy range of 13.5 to 15 MeV [5-12]. Furthermore, several standardized activation data libraries, namely ENDF/B-VIII.0, ROSFOND-2010, JEFF-3.3, JENDL-5, TENDL-2021 etc. have been established. Among them, TENDL-2021 is based on both default and adjusted TALYS calculations and data from the previous version of TENDL library. The shapes of the excitation functions of the three investigated reactions reported in the above data libraries are rather discrepant. Data testing and validation of the evaluated data libraries by integral measurements thus appear to be timely and useful. An interesting area of application of neutron fields consists of performing integral tests of excitation functions of neutron induced reactions. The another aim of this work is testing of data files utilizing the TRIGA reactor neutron spectrum. As far as known, spectrum-averaged cross section

measurements and data testing of the three reactions using a TRIGA reactor have hitherto not been reported besides our studies.

2. Experimental techniques

2.1 Samples and irradiation

High purity $^{nat}SrCl_2.6H_2O$ (purity: 99%; MERCK) available in the powder form was used as target. The powder was pressed by hydraulic press at a pressure of 10 tons to make a pellet. Two pellets of 1091 mg and 2074 mg were prepared and sealed in separate polyethylene bags. The diameter of each pellet was 13 mm. High purity foils of Ni (0.125 mm) and Au (0.025 mm) were cut in circular discs with a diameter similar to the target pellet. Each target was sandwiched between Ni and Au foils. Ni foil was used to monitor fast neutrons and Au foil for thermal neutrons. Two stacked samples were placed in an irradiation vial. The irradiation with neutrons was performed for 2 h at the DCT of the TRIGA Mark II research reactor mentioned above. During the irradiation the reactor power was kept constant at 500 kW. After irradiation the samples were kept in the core of the reactor for one day to let the unwanted short lived radionuclides decay and to reduce the radiation dose below the permissible level. Thereafter, the radioactive samples were taken out from the reactor and each sample was put separately in a fresh polyethylene bag to avoid contamination when handling sample for measurement.

2.2. Gamma-ray spectrometric measurement

The radioactivity of the product radionuclide under investigation 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) at the Institute of Nuclear Science and Technology (INST), Savar, Dhaka, Bangladesh. The detector was associated with a digital gamma spectrometry system (ORTEC DSPEC jr ^{TM}) and Maestro data acquisition software. The spectrum was analyzed using the FitzPeak gamma analysis software [18].

The ^{87m}Sr ($T_{1/2}$=2.815 h) activity was measured at a distance of 25 cm from the detector surface to keep the dead time below 5 %. To check the half-life, the measurement was repeated several times by giving sufficient intervals within a day. A very sharp peak was observed in the spectrum at an energy of 388 keV, emitted in the decay of ^{87m}Sr via isomeric transition.

Due to low activities of the relatively long-lived radionuclides ^{84}Rb ($T_{1/2}$= 32.82 d) and ^{86}Rb ($T_{1/2}$= 18.642 d), their measurement was started 4 days after EOB. During this time the short-lived radionuclides had decayed out and the background count was low. Sharp peaks for those radionuclides, therefore, appeared at energies of 881 keV and 1076 keV, respectively. The counting was performed for 20000 sec at 7 cm from the detector surface and a good counting statistics was achieved. Measurement was repeated several times giving 7 to 20 days interval over a period of several months to check the half-lives of the above two long-lived activation products.

The decay data were taken from the ENSDF library [19-23] through the NuDat 3 web interface [24]. The efficiency versus energy curve of the HPGe gamma-ray detector was determined using the standard point sources of ^{57}Co, ^{60}Co, ^{137}Cs and ^{152}Eu (supplied by North American Scientific).

2.3 Fast neutron flux

The fast neutron flux for energies above the ^{58}Ni(n,p)^{58}Co reaction threshold (~0.5 MeV) was determined [1]. The standard integral cross section of the monitor reaction ^{58}Ni(n,p)^{58}Co for the TRIGA Mark II reactor was adopted as 91.2 ± 5.5 mb. It was obtained from the well-characterized part of the neutron spectrum at the irradiation position in the core of the reactor [cf. 1] and the excitation function of the ^{58}Ni(n,p)^{58}Co reaction from the IRDFF-II library [25]. From the 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 3.88×10^{12} ncm^{-2}s^{-1} at the reactor power of 500 kW used in this experiment.

2.4 Thermal neutron flux

The monitor reaction ^{197}Au(n,γ)^{198}Au was used to determine the thermal and epithermal neutron flux at the irradiation position. The method in detail was given in [cf. 1]. Both the thermal and epithermal neutron fluxes were determined from the measured activity of ^{198}Au formed in a Au-monitor foil and using the known thermal cross section (98.8 b) and resonance integral (1550 b). At the irradiation position in our reactor, the thermal neutron flux obtained was 5.99×10^{12} ncm^{-2}s^{-1}, whereas the flux of epithermal neutrons was 3.06×10^{11} ncm^{-2}s^{-1}.

2.5 Determination of integral cross section

The measured γ-ray count rate of each activation product was 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, and then extrapolated to the end of bombardment (EOB), and the following formula was used for calculation.

$$R = \frac{\lambda C}{N \varepsilon I_{\gamma} \; e^{-\lambda t_C} \; (1 - e^{-\lambda t_m}) \cdot (1 - e^{-\lambda t_i})}$$

where, λ is decay constant, s^{-1}, C total counts of gamma-ray peak area, N number of target atom, ε peak efficiency, I_γ branching ratio of gamma-ray, t_c cooling time, t_m counting time, and t_i irradiation time.

From the decay rates at EOB and the neutron flux $\phi(E)$ (>0.5 MeV) effective at the sample, the spectrum-averaged cross sections of the $^{84}Sr(n,p)^{84}Rb$, $^{86}Sr(n,p)^{86}Rb$ and $^{87}Sr(n,n')^{87m}Sr$ reactions were determined using the following formula:

$$\sigma(E\ ) = \frac{R}{\phi(E\ )}$$

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 (0.5-8%), sample preparation (5%), half-life of the product (<0.4%), γ-ray intensity (0.01-3.0%), 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 9-11 %.

3. Results and discussion

3.1 Spectrum averaged cross sections

The measured spectrum-averaged cross-sections in the neutron field from 0.5 to 20 MeV in the TRIGA reactor are given in Table 2. The total uncertainty of each value is also quoted in the same table. Our measurements were done for the first time.

While measuring neutron-induced reaction cross sections using a natural Sr target, some contributions to the formation of ^{87m}Sr from subsidiary reactions on the undesired target isotopes ^{86}Sr (9.86%) and ^{88}Sr (82.58%) present in the Sr were also expected. With regard to the formation of ^{87m}Sr, there are three channels, namely ^{87}Sr(n,n'), ^{88}Sr(n,2n) and ^{86}Sr(n,γ). We estimated those contributions from the available literature values. The cross section of the ^{86}Sr(n,γ) reaction at 0.0253 eV energy is well known. We adopted the cross section of this reaction from the average of the literature values [26-31] and it amounted to 854 mb, whereas average resonance integral is 4164 mb [26,27, 32-34]. The (n,γ) process dominates the formation of ^{87m}Sr in the core of the reactor and its contribution is estimated as 66 %. The contribution of the ^{88}Sr(n,2n) process was estimated from the integral cross section (10 mb) recommended by Calamand [4] for the pure fission spectrum and this amounts to about 3% of total formation of ^{87m}Sr. The calculated contribution from the subsidiary reactions was subtracted from the measured total activity and from the remaining activity the integral cross section of the ^{87}Sr(n,n')^{87m}Sr process was deduced.

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

A comparison of the integral data measured in this work was done with the values for the pure ^{235}U fission spectrum (cf. Calamand, [4]). For the reaction ^{84}Sr(n,p)^{84}Rb, the Calamand value of 8.6 mb is about two times higher than our measured value (4.1±0.4 mb). The value for the ^{86}Sr(n,p)^{86}Rb reaction obtained in this work is about 5 times lower than that of Calamand. This deviation may be due to the larger uncertainty associated with the estimated values for the pure fission spectrum. Calamand [4] did not report any recommended value for the two reactions, because no experimental data of the above two reactions were available before his report.

From statistical point of view, our measured cross-section value of the reaction $^{87}Sr(n,n')^{87m}Sr$ is approximately consistent with the recommended value of Calamand [4]. The recommended value is about 26 % lower than this work. 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 somewhat fair 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 libraries

The integrated cross sections over the neutron spectrum from 0.5 to 20 MeV of TRIGA reactor were calculated from the known excitation functions given in the evaluated data libraries, ENDF/B-VIII.0, ROSFOND-2010, JEFF-3.3, JENDL-5 and TENDL-2021. The evaluated excitation functions of the three investigated reactions described in those data libraries are shown in Figs. 1-3, together with the available experimental data points [5-12]. All evaluations were based on experimental data in the first four data libraries, whereas TENDL-2021 is a theory-based library. The integrated cross sections for the investigated reactions 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 reaction $^{84}Sr(n,p)^{84}Rb$, the $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratio for all the libraries, except ROSFOND-2010, lies between 0.78 and 0.93. In the case of ROSFOND-2010, the $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratio is 0.39, i.e. the excitation curve for this library (shown in Fig.1) is too low and thus needs improvement.

As shown in Fig.2, the excitation function curve of the reaction $^{86}Sr(n,p)^{86}Rb$ in the ENDF/B-VIII.0 overlapped that of ROSFOND-2010. The integrated cross section from those libraries is about 28 % higher than the measured integral value. In Fig.2, the excitation function from 5 to 10 MeV is too high compared to the general trend, and this part of the excitation function curve is responsible for the deviation. The $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratio for the data library TENDL-2021 is 1.0, whereas the ratio for JEFF-3.3 is 0.98, and the small deviation may be attributed to the

systematically lower excitation function. The experimental cross section data between 13 and 15 MeV are also reproduced well by TENDL-2021. Therefore, it can be interpreted that the TENDL-2021 evaluation is consistent with both 14 MeV experimental neutron cross sections and newly measured spectrum averaged cross section.

The excitation function of the reaction ^{87}Sr(n,n')^{87m}Sr was found only in the evaluated data libraries, JENDL-5, JEFF-3.3 and TENDL-2021. The agreement between the integral value measured for the reaction ^{87}Sr(n,n')^{87m}Sr (this work) and the integrated values of both TENDL-2021 and JEFF-3.3 is quite good (see Table 2). In the case of JENDL-5, the $\langle\sigma\rangle_{cal.}/\langle\sigma\rangle_{meas.}$ ratio amounted to 0.84 and it is consistent with the measured value from the statistical point of view. As shown in Fig. 3, possibly a small deviation originates from the shift of the excitation function to downwards between 2 and 9 MeV compared to the other two libraries.

4. Conclusions

A new measurement of the integral cross sections for the reactions ^{84}Sr(n,p)^{84}Rb, ^{86}Sr(n,p)^{86}Rb and ^{87}Sr(n,n')^{87m}Sr in the neutron field from the energy 0.5 to 20 MeV at TRIGA reactor was performed. The results obtained in this work are the first one measured at a TRIGA reactor. The available integral cross sections of the reactions ^{84}Sr(n,p)^{84}Rb and ^{86}Sr(n,p)^{86}Rb for the pure fission spectrum are not comparable to this work. In the case of the ^{87}Sr(n,n')^{87m}Sr reaction, the recommended values for the pure fission spectrum deviate by about 16% from our measured value; it can be explained by i) the difference between the TRIGA neutron spectrum and the pure fission spectrum below 2 MeV and ii) recommended value is based on only two experimental values which are not enough to extract a standard value. For a few investigated reactions, the evaluated data files appear to need improvement.

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Table 1

Nuclear reactions and decay data of investigated activation products [19-23].

Sample	Purity (Supplier)	Nuclear reaction	Q-value (MeV)	Threshold energy (MeV)	Reaction threshold (MeV)	Decay data of the activation product		
						$T_{1/2}$	E_γ (keV)	I_γ (%)
$SrCl_2.6H_2O$	99 % (MERCK)	^{84}Sr(n,p)^{84}Rb	-0.11	0.11	3.0	32.82(7) d	881.6	68.9(2.1)
		^{86}Sr(n,p)^{86}Rb	-0.99	1.00	4.0	18.642(18) d	1077	8.64(4)
		^{87}Sr(n,n')^{87m}Sr	-0.39	0.40	0.5	2.815 (12) h	388.5	82.19(20)
Ni	99.98% (Chempur)	^{58}Ni(n,p)^{58}Co	0.40	0.0	0.5	70.86(6) d	810.7	99.00(1)
Au	99.9 % (Goodfellow)	^{197}Au(n,γ)^{198}Au	6.51	0.0	0.0	2.6941(2) d	411.8	95.62(6)

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

	<σ>$_{meas}$ (This work) mb	<σ>$_{Recom}$ (Calamand) mb	ENDF/B-VIII.0		ROSFOND-2010		JEFF-3.3		JENDL-5		TENDL-2021	
			<σ>$_{cal}$ mb	<σ>$_{cal}$/<σ>$_{meas}$	<σ>$_{cal}$ mb	<σ>$_{cal}$/<σ>$_{meas}$	<σ>$_{cal}$ mb	<σ>$_{cal}$/<σ>$_{meas}$	<σ>$_{cal}$ mb	<σ>$_{cal}$/<σ>$_{meas}$	<σ>$_{cal}$ mb	<σ>$_{cal}$/<σ>$_{meas}$
^{84}Sr(n,p)^{84}Rb	4.1±0.4	8.6[b)]	3.2	0.78	1.6	0.39	3.6	0.88	3.8	0.93	3.6	0.88
^{86}Sr(n,p)^{86}Rb	0.40±0.04	2.0[b)]	0.51	1.28	0.51	1.28	0.39	0.98	0.38	0.95	0.40	1.00
^{87}Sr(n,n')^{87m}Sr	152±15	112±17[c)]	-	-	-	-	156	1.03	128	0.84	148	0.97

a) For the neutron field from 0.5 to 20 MeV in a TRIGA reactor, except for Calamand values [4] which are for pure fission neutron spectrum.

b) Calamand values estimated.

c) The value recommended by Calamand.

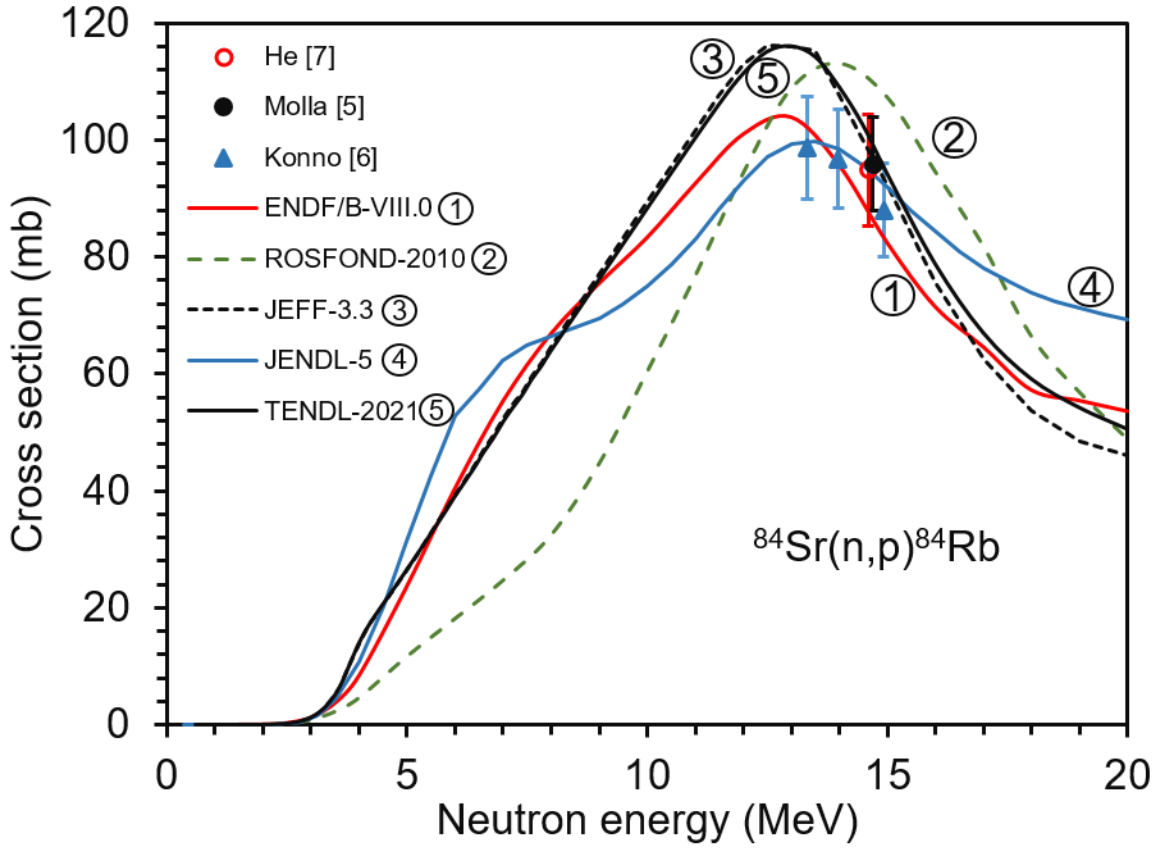

Fig.1 Excitation function of the ^{84}Sr(n,p)^{84}Rb nuclear reaction. Given are experimental data and calculated curves from various data libraries.

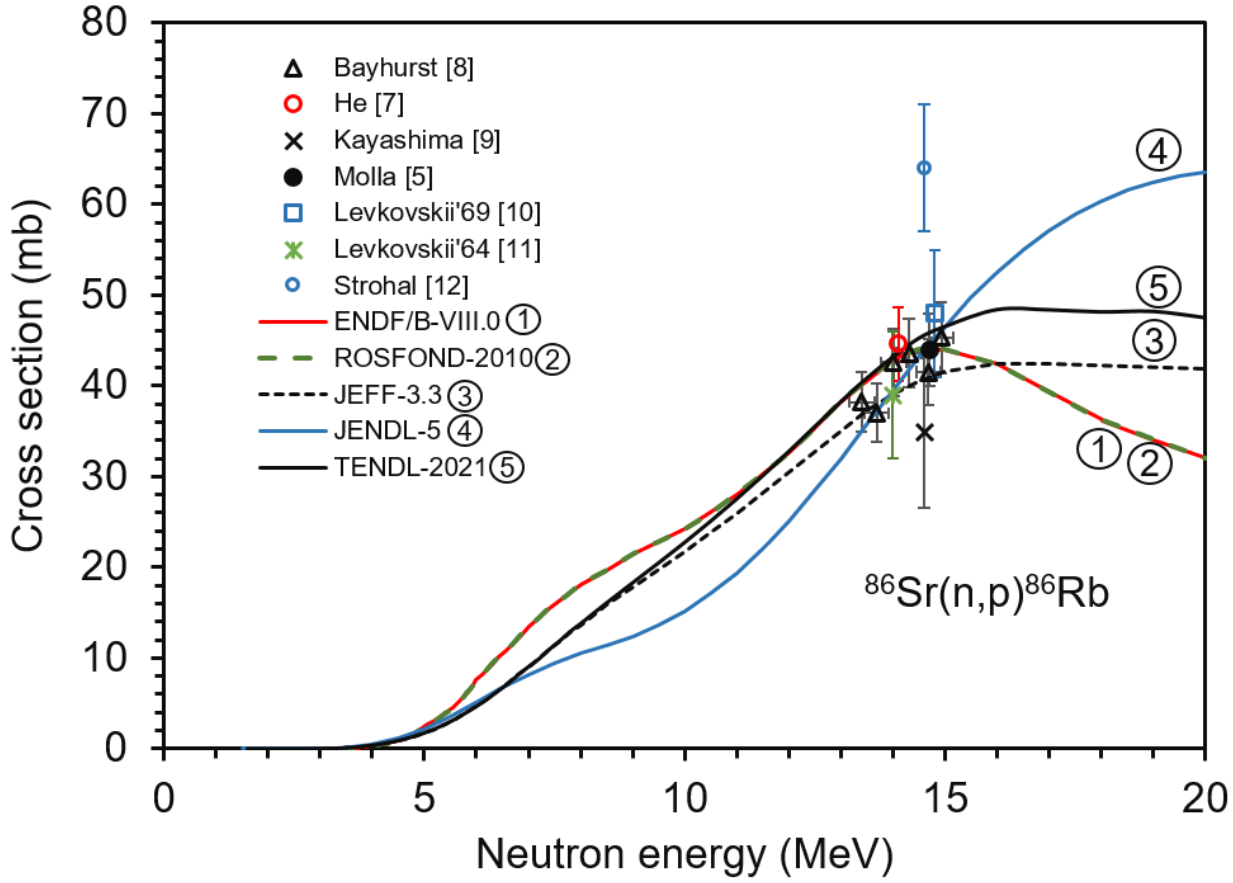

Fig.2 Excitation function of the ^{86}Sr(n,p)^{86}Rb nuclear reaction. Given are experimental data and calculated curves from various data libraries.

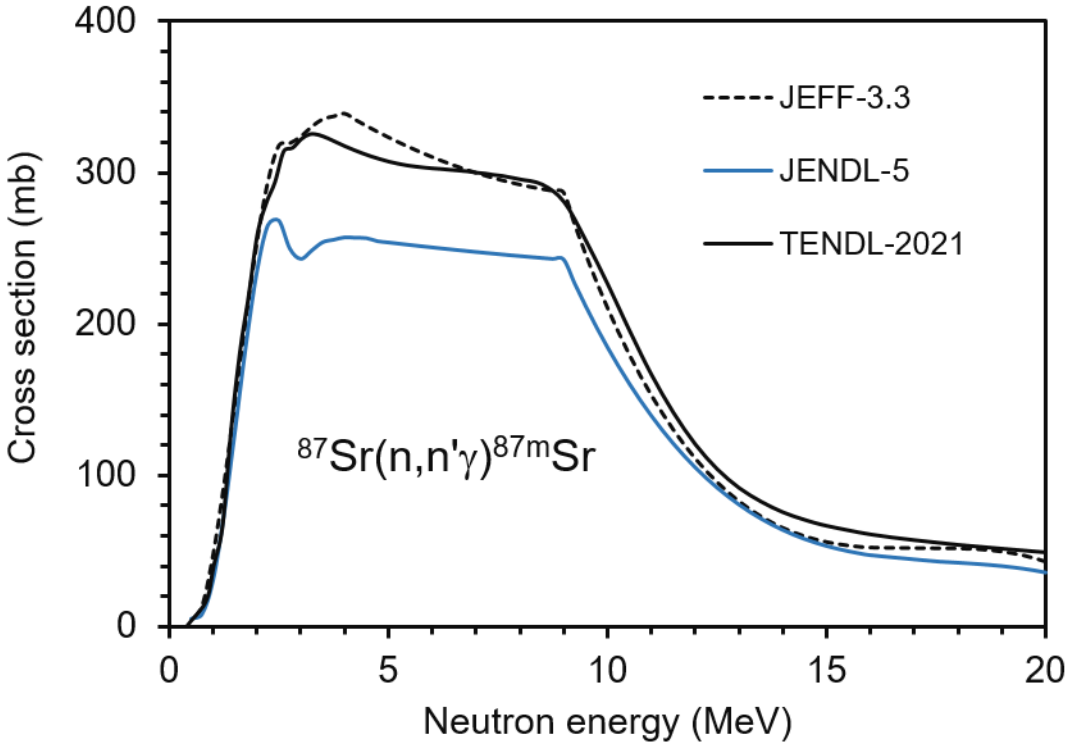

Fig.3 Excitation function of the ^{87}Sr(n,n')^{87m}Sr reaction. Shown are only calculated curves from various data libraries.