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Integral cross section measurements of a few threshold reactions induced by Am/Be neutrons

Abstract: Integral cross sections of the reactions $^{46}\text{Ti}(n, p)^{46}\text{Sc}$, $^{47}\text{Ti}(n, p)^{47}\text{Sc}$, $^{48}\text{Ti}(n, p)^{48}\text{Sc}$, $^{60}\text{Ni}(n, p)^{60}\text{Co}$ and $^{64}\text{Zn}(n, p)^{64}\text{Cu}$ were measured with fast neutrons ($E_n > 1.5$ MeV) from an Am/Be source. The results were compared with data calculated using the neutron spectral distribution and the excitation function of each reaction given in the data libraries ENDF/B-VII.0, IRDF-2002, JEFF-3.2 and JENDL-4.0. In general, the integral measurement and the integrated value agreed within $\pm 4\%$, except for the $^{46}\text{Ti}(n, p)^{46}\text{Sc}$ reaction where JEFF-3.2 shows a deviation of 7% and the $^{60}\text{Ni}(n, p)^{60}\text{Co}$ reaction where ENDF/B-VII.0 and IRDF-2002 exhibit deviations upto 8%.

Keywords: Am/Be neutron source, Nuclear reaction, γ -ray spectrometry, Spectrum averaged cross section, Excitation function, Integral test of differential data.

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1 Introduction

An Am/Be neutron source was recently installed at the Department of Applied Chemistry and Chemical Engineering of Rajshahi University (Bangladesh) and the neutron field produced was determined at various irradiation positions around the source [1]. The shape of the neutron spectrum was reproduced very well (within the uncertainty

limit of 6%) in comparison to the spectrum recently characterized at a “standards” laboratory [2]. The low energy part of the spectrum has a somewhat higher uncertainty, but the spectrum is well defined over the neutron energy range of 1.5 to 11.0 MeV. Such a source could possibly be conveniently used for integral cross section measurement of a neutron threshold reaction. A comparison of the integral measurement with the integrated cross section, obtained from the known excitation function of that reaction averaged over the neutron spectrum, could then be used to test the reliability of the excitation function. This was indeed done in the case of a few (n, p) reactions on medium mass target nuclei [1, 3].

In the present study the cross sections, averaged over the Am/Be neutron spectrum, of a few (n, p) reactions induced in Ti, Ni and Zn targets were experimentally determined relative to the $^{58}\text{Ni}(n, p)^{58}\text{Co}$ reaction spectrum-averaged cross section. The integral data obtained were then used to test the excitation functions of those reactions, as given in several data files.

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 [4, 5]; they are also given in Table 1. The elements Ti, Ni and Mo were available in the form of foil, the first two having a thickness of 125 μm and the third one a thickness of 250 μm . Samples for irradiations were prepared by cutting discs of 2 cm diameter. The Zn sample was prepared by pressing 600 mg of ZnO to form a circular pellet of 1.3 cm diameter. Whereas Ti and Zn served as targets for cross section measurements of (n, p) reactions, the Ni foil was used as a monitor of neutron flux. The Mo foil served as additional neutron flux monitor via the $^{92}\text{Mo}(n, p)^{92\text{m}}\text{Nb}$ reaction [cf. 3].

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Table 1: Irradiated samples and investigated activation products.

Sample	Purity (Supplier)	Nuclear reaction	Natural abundance of target isotope (%)	Q-value (MeV)	Reaction threshold energy ^{a)} (MeV)	Decay data of the activation product ^{b)}		
						$T_{1/2}$	E_γ (keV)	I_γ (%)
ZnO (Powder)	99.9% Mathey Johnson	$^{64}\text{Zn}(n, p)^{64}\text{Cu}$	48.6	+0.20	2.0	12.7 h	1345	0.54 ^{c)}
							511	35.7 ^{c)}
Ni	99.6% Goodfellow	$^{58}\text{Ni}(n, p)^{58}\text{Co}$	68.08	+0.40	0.5	70.82 d	811	99.4
		$^{60}\text{Ni}(n, p)^{60}\text{Co}$	26.22	-2.04	4.2	5.27 y	1173.2	99.97
Ti	99.9% Goodfellow	$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	8.0	-1.58	2.0	83.82 d	889.0	99.98
		$^{47}\text{Ti}(n, p)^{47}\text{Sc}$	7.3	+0.18	1.5	3.35 d	159.3	68.3
		$^{48}\text{Ti}(n, p)^{48}\text{Sc}$	73.8	-3.21	6.2	1.80 d	983.5	100
							1037.6	97.6
Mo	99.9% Goodfellow	$^{92}\text{Mo}(n, p)^{92m}\text{Nb}$	14.84	+0.29	2.25	10.15 d	934.5	99.0

^{a)} Sum of threshold energy and Coulomb barrier height.

^{b)} Taken from NUDAT data base [4], unless otherwise stated.

^{c)} Taken from Qaim et al. [5].

2.2 Irradiations

Irradiations with neutrons were carried out in two different ways, depending on half-life of the product. 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.

a) Study of the $^{64}\text{Zn}(n, p)^{64}\text{Cu}$ reaction

Due to the relatively short half-life of ^{64}Cu ($T_{1/2} = 12.7$ h), the ZnO pellet sandwiched between Ni-foils was irradiated for 37 h. Two such samples were irradiated. They were transported immediately to the Institute of Nuclear Science and Technology (INST), Savar, Dhaka, where γ -ray spectrometric analysis was started about 5 h after the end of irradiation. During the transportation period the short-lived products (e.g. ^{16}N , ^{66}Cu , etc.) had decayed out. In each stack the Ni foils served as the neutron flux monitor by virtue of the well-known excitation function of the $^{58}\text{Ni}(n, p)^{58}\text{Co}$ reaction [cf. 6].

b) Investigation of (n, p) reactions on Ti and Ni

Many of the investigated products are long-lived. To study them, two stacks, each consisting of Ni-Mo-Ti-Mo-Ti-Ni, were irradiated for about 71 d. At end of irradiation the

samples were transported to the Institute of Nuclear Science and Technology (INST), Savar, Dhaka to measure the activities of the product radionuclides. Thereafter, they were transferred to Forschungszentrum Jülich, Germany, where low-level γ -ray spectrometric measurements were carried out on long-lived products.

2.3 Measurement of radioactivity

The radioactivity of the radionuclide ^{64}Cu formed in the ZnO samples through the $^{64}\text{Zn}(n, p)^{64}\text{Cu}$ reaction was measured using a HPGe detector (Canberra, 25% relative efficiency, 1.9 keV resolution at 1332.5 keV of ^{60}Co , coupled with ORTEC DSPEC jrTM) γ -ray spectrometer at the Institute of Nuclear Science and Technology (INST), Savar, Dhaka, Bangladesh. Special care was taken to provide sufficient absorber between the source and the γ -ray detector to annihilate all positrons. The samples were counted on the surface of the detector to get good counting statistics, which, however, demands a correction on the efficiency of the extended sample. The γ -ray spectra were analysed by the software GammaVision, version 6.01. The details on γ -ray counting and detector efficiency measurement have been described earlier [1]. The annihilation gamma ray at 511 keV emitted in the decay of ^{64}Cu could also have some contribution from the background. So it was subtracted. The area under the 511 keV peak was followed as a function of time. A half-life of 12.7 h was obtained, show-

ing that the contribution of ^{65}Zn to the annihilation peak, if any, was negligible. This was further confirmed by the absence of the 1115 keV γ -ray of ^{65}Zn in the irradiated ZnO sample. Due to the short irradiation time and low intensity of thermal neutrons from the Am/Be source, the ^{65}Zn production via the $^{64}\text{Zn}(n,\gamma)$ reaction was negligible. Attempt was also made to characterize ^{64}Cu via the very weak γ -ray at 1345 keV. The uncertainty was, however, rather high.

The radioactivities of the radionuclides ^{46}Sc , ^{47}Sc , ^{48}Sc and ^{58}Co induced in Ti and Ni were also measured at the Institute of Nuclear Science and Technology (INST), Savar, Dhaka. Furthermore, measurement of the radioactivity of the long-lived product ^{46}Sc formed in the Ti samples, as well as of ^{58}Co and ^{60}Co formed in the Ni foils, was carried out at the Institut für Nuklearchemie, Forschungszentrum Jülich, Germany, using a low-level HPGe detector as described earlier [7]. Due to weak activities, the samples were placed directly on the surface of the detector, where an efficiency loss for the extended sample and loss of counts due to real coincidences had to be corrected. For this purpose, an independently prepared active enough sample was also counted at 10 cm from the detector surface, where both the sample-size effect on the efficiency and the coincidence loss were negligible. The obtained activity at 10 cm was considered as standard value, using which the detector efficiency for the extended sample on the surface combined with the coincidence loss was calculated. The analysis of the γ -ray spectra was done also at Jülich using the GammaVision software (Version 6.01).

2.4 Neutron flux measurement

The $^{58}\text{Ni}(n,p)^{58}\text{Co}$ 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 a value of 391 ± 19 mb [1] for the spectrum-averaged cross section of this reaction and the experimentally determined decay rate of the activation product ^{58}Co , the flux of neutrons above 1.5 MeV was calculated. As a further check the neutron flux was also determined using the $^{92}\text{Mo}(n,p)^{92\text{m}}\text{Nb}$ reaction, adopting its spectrum averaged cross section as 43 ± 4 mb [1, 3]. The two flux values agreed within 5%.

2.5 Calculation of integral reaction cross sections and their uncertainties

From the measured count rate of each product radionuclide and the flux of neutrons of energies above 1.5 MeV, the neutron-spectrum averaged cross section $\langle\sigma\rangle$ of each investigated reaction, i.e. $^{46}\text{Ti}(n,p)^{46}\text{Sc}$, $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, $^{48}\text{Ti}(n,p)^{48}\text{Sc}$, $^{60}\text{Ni}(n,p)^{60}\text{Co}$ and $^{64}\text{Zn}(n,p)^{64}\text{Cu}$, 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 which were very similar to those described earlier [3].

3 Results and discussion

3.1 Spectrum averaged cross sections

The experimentally determined integral cross sections for the $^{46}\text{Ti}(n,p)^{46}\text{Sc}$, $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, $^{48}\text{Ti}(n,p)^{48}\text{Sc}$, $^{60}\text{Ni}(n,p)^{60}\text{Co}$ and $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ reactions, averaged over the fast neutron spectrum ($E_n > 1.5$ MeV), are given in Table 2. The low energy spectrum below 1.5 MeV was not considered because it has no effect on the investigated neutron threshold reactions. Each value in Table 2 is based on at least two independent measurements. The total uncertainty of each value amounts to between 9 and 12% (1 sigma). Special care needed in the study of the $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ reaction has been described above. Great care was also needed in the case of the $^{60}\text{Ni}(n,p)^{60}\text{Co}$ reaction because ^{60}Co sources in the counting room often contribute to the background. The $^{60}\text{Ni}(n,p)^{60}\text{Co}$ reaction cross section could be measured because the background was very low in the counting system at the Forschungszentrum Jülich, Germany.

An interesting observation is the sharp decrease in the (n,p) cross section as the isotopic mass of the target element increases (^{46}Ti to ^{48}Ti). This was clearly observed also with 14 MeV neutrons and it was attributed to signif-

Table 2: Measured spectrum averaged cross sections.

Nuclear reaction	Cross section (mb)
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	100 ± 10
$^{47}\text{Ti}(n,p)^{47}\text{Sc}$	63 ± 6
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	4.6 ± 0.6
$^{60}\text{Ni}(n,p)^{60}\text{Co}$	26 ± 2.5
$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	143 ± 13

icant differences in proton binding energies of the respective target nuclei [cf. 8].

As far as literature data are concerned, cross sections of the (n,p) reactions on ^{46}Ti , ^{47}Ti , ^{48}Ti and ^{64}Zn have been reported [cf. 9] for Am/Be neutrons. A comparison of those data with our results is, however, difficult because the literature values are for average neutron energies above reaction thresholds, whereas our data are averaged over the whole range of neutron energy.

3.2 Integral tests of excitation functions

The standard Am/Be neutron spectrum is shown in Figure 1(A) [cf. 1, 2] and the excitation functions of the five investigated nuclear reactions were taken from various evaluated files and libraries [6, 10–13]. The curves for four cases, namely $^{46}\text{Ti}(n,p)^{46}\text{Sc}$, $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, $^{60}\text{Ni}(n,p)^{60}\text{Co}$ and $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ reactions, are shown in Figure 1(B to E) because some discrepancies exist in data libraries. The excitation function curves for the reaction $^{48}\text{Ti}(n,p)^{48}\text{Sc}$ are more consistent and are therefore

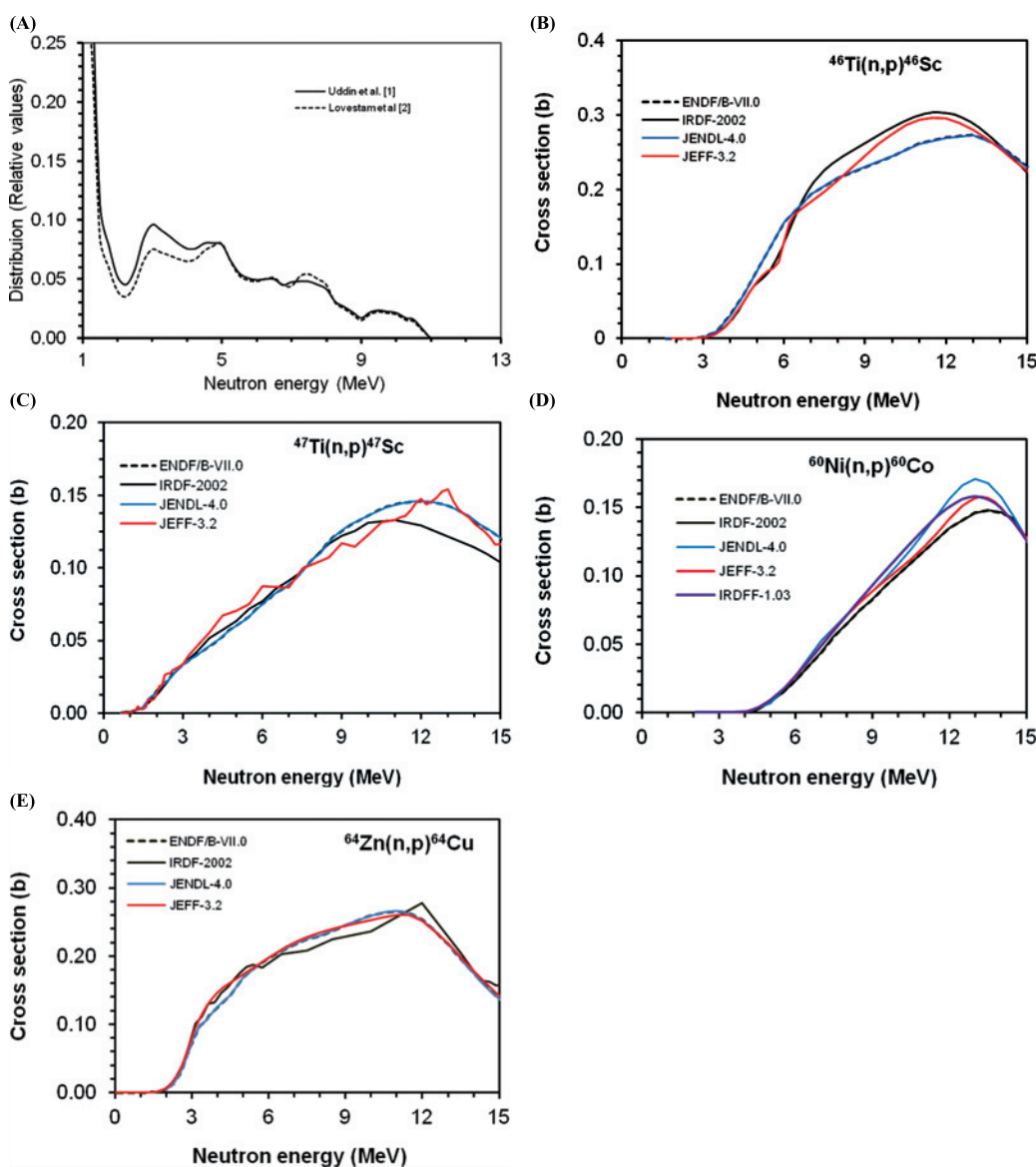


Fig. 1: (A) The standard neutron spectrum of the Am/Be neutron source, above 1.5 MeV used in this work. The spectrum below 1.5 MeV is not well characterized and it is not shown because it has no effect on the investigated neutron threshold reactions. (B) Evaluated excitation functions of the reaction $^{46}\text{Ti}(n,p)^{46}\text{Sc}$ given in different libraries. (C) Evaluated excitation functions of the reaction $^{47}\text{Ti}(n,p)^{47}\text{Sc}$ given in different libraries. (D) Evaluated excitation functions of the reaction $^{60}\text{Ni}(n,p)^{60}\text{Co}$ given in different libraries. (E) Evaluated excitation functions of the reaction $^{64}\text{Zn}(n,p)^{64}\text{Cu}$ given in different libraries.

Table 3: Measured and calculated neutron-spectrum averaged cross sections.

Reaction	Spectrum-averaged cross section $\langle\sigma\rangle$ (mb)										
		ENDF/B-VII.0		IRDF-2002		JEFF-3.2		JENDL-4.0		Research papers	
	$\langle\sigma\rangle_{\text{meas}}$ (This work)	$\langle\sigma\rangle_{\text{cal}}$	$\frac{\langle\sigma\rangle_{\text{cal}}}{\langle\sigma\rangle_{\text{meas}}}$	$\langle\sigma\rangle_{\text{cal}}$	$\frac{\langle\sigma\rangle_{\text{cal}}}{\langle\sigma\rangle_{\text{meas}}}$	$\langle\sigma\rangle_{\text{cal}}$	$\frac{\langle\sigma\rangle_{\text{cal}}}{\langle\sigma\rangle_{\text{meas}}}$	$\langle\sigma\rangle_{\text{cal}}$	$\frac{\langle\sigma\rangle_{\text{cal}}}{\langle\sigma\rangle_{\text{meas}}}$	$\langle\sigma\rangle_{\text{cal}}$	$\frac{\langle\sigma\rangle_{\text{cal}}}{\langle\sigma\rangle_{\text{meas}}}$
$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	100 ± 10	99	0.99	99	0.99	93	0.93	99	0.99	$95^{a)}$	0.95
$^{47}\text{Ti}(n, p)^{47}\text{Sc}$	63 ± 6	62	0.98	64	1.01	65	1.03	62	0.98	$64^{b)}$	1.01
$^{48}\text{Ti}(n, p)^{48}\text{Sc}$	4.6 ± 0.6	4.6	1.00	4.8	1.04	4.4	0.96	4.6	1.00	$4.6^{b)}$	1.00
$^{60}\text{Ni}(n, p)^{60}\text{Co}$	26 ± 2.5	24	0.92	24	0.92	25	0.96	26	1.00	$27^{d)}$	1.03
				$26^{c)}$	1.00						
$^{64}\text{Zn}(n, p)^{64}\text{Cu}$	143 ± 13	141	0.99	143	1.00	148	1.03	142	0.99	–	–

^{a)} Molla et al. [15, 17]^{b)} Qaim et al. [16]^{c)} IRDFF-1.03 [13]^{d)} Sudár et al. [18]

not given in Figure 1. Based on the available information, the integrated cross section for the standard Am/Be neutron spectrum was obtained. Considering the uncertainties in the neutron spectrum and the evaluated excitation function, the overall uncertainty in the integrated reaction cross section was estimated to be about 6%. The results are given in Table 3.

The integrated reaction cross sections from evaluated libraries [6, 10–13] were compared with the experimentally measured integral cross section. The results $\langle\sigma\rangle_{\text{cal.}}/\langle\sigma\rangle_{\text{meas.}}$ for the investigated reactions are also given in Table 3. It should, however, be pointed out that all evaluated data libraries are based on experimental data given in the EXFOR library [14]. At Jülich a systematic study of (n, p) reactions on ^{46}Ti , ^{47}Ti , ^{48}Ti , ^{49}Ti and ^{50}Ti was carried out over the neutron energy range of 5 to 12 MeV in collaboration with Vienna and Dresden groups [15–17]. Similarly detailed studies were performed on the $^{60}\text{Ni}(n, p)^{60}\text{Co}$ reaction under a Jülich-Debrecen cooperation [18]. A comparison was, therefore, also carried out between the experimental spectrum averaged cross sections measured in this work and the spectrum averaged cross sections deduced from the extensive excitation functions reported from Jülich and cooperating laboratories. Those results are also shown in Table 3.

For the five investigated reactions, the $\langle\sigma\rangle_{\text{cal.}}/\langle\sigma\rangle_{\text{meas.}}$ ratios for different libraries are 1.00 ± 0.04 , except for a few cases. For the reaction $^{46}\text{Ti}(n, p)^{46}\text{Sc}$ the ratio in JEFF-3.2 is 0.93. For the $^{60}\text{Ni}(n, p)^{60}\text{Co}$ reaction the ratio in IRDF-2002 and ENDF/B-VII.0 is 0.92, whereas the ratio in IRDFF-1.03 is 1.00. The present integral measurements thus reflect the status of the evaluated excitation functions of those five reactions well. The same applies to the experimental data reported for four reactions from Jülich [15–18]. Con-

sidering in more detail, for the reactions $^{47}\text{Ti}(n, p)^{47}\text{Sc}$, $^{48}\text{Ti}(n, p)^{48}\text{Sc}$ and $^{64}\text{Zn}(n, p)^{64}\text{Cu}$ the ratios calculated from all the curves showed good agreement. In other two cases shown in Figure 1, on the other hand, some differences are visible. For the reaction $^{46}\text{Ti}(n, p)^{46}\text{Sc}$, Figure 1(B), for example, the IRDF-2002 and JEFF-3.2 curves differ considerably from the other two curves, especially in the energy region beyond 10 MeV. This, however, does not influence the present ratio calculations because of the weak contribution of > 10 MeV neutrons to the total neutron spectrum shown in Figure 1(A). The ratio of the integrated to the measured cross section for the JEFF-3.2 file is low (0.93). The same applies to the Jülich data. In the case of the $^{60}\text{Ni}(n, p)^{60}\text{Co}$ reaction, the calculated to the measured ratio for three evaluated files (ENDF/B-VII.0, IRDF-2002 and JEFF-3.2) is consistent but low. The evaluated data from JENDL-4.0 and IRDFF-1.03 and the Jülich experimental data show better agreement. In general, however, it is concluded that the present integral measurements validate the evaluated excitation functions of the five investigated reactions fairly well.

4 Conclusion

Using the recently installed Am/Be neutron source at the Rajshahi University, neutron spectrum averaged cross sections were measured for five nuclear reactions. The fast neutron field (with $E_n > 1.5$ MeV) was found to be well suited for integral tests of excitation functions of the investigated neutron threshold reactions. In most of the cases, the measured integral data are in good agreement with the data integrated from the evaluated excitation functions,

which reflects the status of the available excitation functions of the reactions considered.

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