Ratio of spectral averaged cross sections measured in standard ^{252}Cf(sf) and ^{235}U(n$_{th}$,f) neutron fields

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Abstract. The results of systematic evaluations of spectrum averaged cross section (SACS) measurements in the fission neutron fields of ^{252}Cf and ^{235}U are presented. The data form a complete database of high-threshold experimental SACS measured in the same installation under the same conditions and using the same high purity germanium gamma spectrometer. This is crucial to reduce the uncertainty of the ratio and the data scattering and therefore, to minimize discrepancies compared to cross section measured under different conditions in different laboratories. This new dataset complements and extends earlier experimental evaluations. The total emission of the ^{252}Cf neutron source during the experiments varied from 9.5E8 to 4.5E8 neutrons per second. The emission was derived in accordance to the data in the Certificate of Calibration involving absolute flux measurements in a manganese sulphate bath. Concerning ^{235}U fission neutron field, the irradiations were carried out in a specifically designed core assembled in the zero power light water LR-0 reactor. This special core has a well described neutron field. After the irradiation, the low volume irradiated samples to be measured by gamma spectrometry were placed directly on the upper cap of a coaxial high purity germanium (HPGe) detector in a vertical configuration (ORTEC GEM35P4). High volume samples were homogenized and strewn into the Marinelli beaker. The HPGe detector is surrounded by the lead shielding box with a thin inner copper cladding and covered with rubber for suppression of background signal and bremsstrahlung. The experimental reaction rates were derived for irradiated samples from the Net Peak Areas (NPA) measured using the semiconductor HPGe detector. The measured reaction rates are used to derive the spectrum-averaged cross sections. Furthermore, measured reaction rates are also compared with MCNP6 calculations using various nuclear data libraries, in particular IRDFF evaluations.

1 Introduction

All presented spectrum-averaged cross sections (SACS) and its ratios were measured under the same conditions and using the same gamma spectrometer. This is important to minimize discrepancies compared to cross sections measured under different conditions and using different spectrometers in different institutes. The ratios of ^{252}Cf and ^{235}U spectral averaged cross sections can be use to specify high energy tail of ^{235}U as the ^{252}Cf spontaneous fission neutron spectrum is taken as a standard. The experiments were performed using a very well characterized light water pool type LR-0 reactor using a special core (6 fuel assemblies with enrichment of 3.3%) and isotopic ^{252}Cf source with emission varying from 8.0E8 to 4.0E8 n/s during experimental work.

2 Experimental and calculation methods

2.1 Experiments

The LR-0 reactor is a zero power light water pool type reactor located in Czech Republic. All irradiation experiments were performed at atmospheric pressure and at room temperature in a specially designed core assembled in the LR-0 reactor. This special core consists of six uranium fuel assemblies with nearly 3.3% ^{235}U enrichment surrounding a special dry assembly with an activation foils and sample holder. Used fuel assemblies are the same as VVER-1000 type in radial direction but not in axial direction, the fission column is shortened to 125 cm. Fuel assemblies have lattice pitch of 23.6 cm. Reactor criticality is always achieved by a moderator level in all experiments. Reactor core in this experiment consists of six uranium fuel assemblies with nearly 3.3% ^{235}U enrichment and surrounds a special dry assembly with target nuclei. There are eight aluminum dry channels surrounding the core for power monitoring. Scheme of the active zone with the power monitor channels can be seen in Fig. 1. The special core has a precisely defined neutron spectrum is crucial for performing experiments concerning the validation of selected neutron induced cross sections as well as for both calibration and testing of spectrometric and dosimetric detectors. Special core was suggested and verified for IRDFF nuclear data library [1] testing by experiments dealing with reactivity characterization [2], fission rates distribution [3] and also neutron spectra measurement in different material insertions in its center [4].

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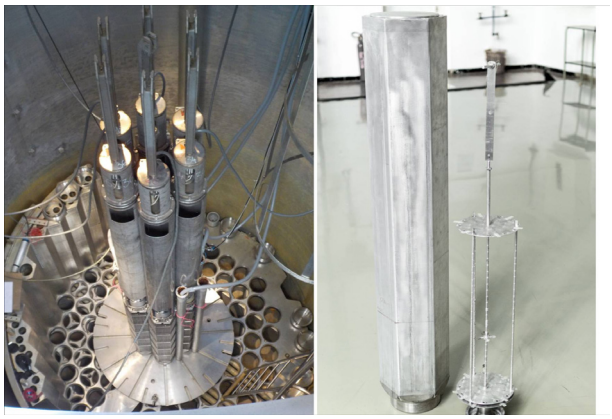

Figure 1. Overhead view inside the LR-0 reactor with a special core without moderator (left) and a dry experimental channel with activation foil holder (right).

The ^{252}Cf source involved in all experiments had initial emission of 9.53E8 n/s on August 13th 2015 at 12:00 GMT according to the data in certificate of calibration involving manganese sulphate bath performed in National Physical Laboratory, UK. The experiments were performed during source emission of 8.01E8 n/s to emission of 4.0E8 n/s. The uncertainty budget of the source is around 2.1%. It consists of uncertainty in position of source in the capsule, emission of the source, density of Pd matrix and ^{250}Cf influence. Fig. 2 shows the example of activation foils irradiation. Irradiation length depends mainly on reaction under study and distance from the source. It took from one week up to three months of uninterrupted irradiations.

Figure 2. Irradiation of the activation foils using ^{252}Cf source.

2.2 Calculations

All calculations were performed using MCNP6 transport code [5]. The influence of the IRDFF-1.05 library on the reactions under study was investigated by only changing the isotope dosimetry cross section (henceforth XS). The rest of ACE files employed in the transport and correction calculations were taken from the ENDF/B-VII.1 library [9] except for ^{235}U prompt fission neutron spectrum (PFNS) which was taken from CIELO-g6 or ENDF/B-VII.1 for comparison. ^{252}Cf spontaneous fission spectrum was taken from the IAEA project on validation webpage [12]. Efficiency of the germanium detector was also calculated in MCNP6 using verified geometry model. The samples containing isotope under study were either low-volume or high-volume. Low volume samples were placed on the upper cap of a coaxial high purity germanium (HPGe) detector in a vertical configuration (ORTEC GEM35P4) to achieve reasonable statistics in a reasonable time. Corrections to the true summation were employed in cases where applicable. High volume samples were irradiated in an aluminium can and subsequently strewn into a marinelli beaker to achieve better efficiency of the detector. Irradiations take from several hours to several weeks to shorten gamma spectroscopy measure time. Efficiency of the used detector is calculated by means of the MCNP6 model. This model was compiled from experimentally measured insensitive layer and radiograms of the detector to estimate inner distances. This approach was verified in [7].

To evaluate ^{235}U SACS, firstly we need to determine reaction rates from experimentally measured NPAs using following formula:

$$q(\bar{P}) = \frac{A_{Sat}(\bar{P})}{A(\bar{P})} \frac{C(T_m)\lambda}{\epsilon\eta N} \frac{1}{1-e^{-\lambda T_m}} \frac{1}{e^{-\lambda\Delta T}}, \qquad (1)$$

where $q(\bar{P})$ is is the reaction rate of activation during power density $\bar{P}$, T_m denotes time of the HPGe measurement, ΔT is the time between the end of irradiation and the start of HPGe measurement, $C(T_m)$ is the measured number of counts, ϵ is the gamma branching ratio, η is the detector efficiency calculated using MCNP6, N is the number of target isotope nuclei. The scaling factor which corresponds to the core neutron emission rate is used for the absolute neutron flux evaluation. It has been determined via reaction rates derived from gamma activity of monitor activation foil detectors. Two kinds of activation material were employed: gold (1% Au in Al) and nickel (100% Ni). The measured reaction rates were corrected to take account of the spectral shift effect; a ratio between the spectral-averaged cross-section in the full geometry used and the spectral-averaged cross-section in the geometrically identical but empty geometry to derive ^{235}U SACS:

$$\bar{\sigma} = \frac{C}{K} \frac{q(\bar{P})}{\int_E \Phi(E)dE}, \qquad (2)$$

where K is the scaling factor based on absolute flux density, $q(\bar{P})$ is the experimentally derived reaction rate, $\Phi(E)$ is the calculated neutron spectrum and C is the correction factor to the spectral shift effect.

Concerning ^{252}Cf source measurements, the approach is similar. The experimental reaction rates were derived for irradiated samples from the Net Peak Areas (NPA).

The experimental reaction rates were derived using the following formula:

$$q = C(T_m)\frac{\lambda}{\epsilon\eta N}\frac{1}{1-e^{-\lambda T_m}}\frac{1}{e^{\lambda\Delta T}}\frac{1}{1-e^{\lambda T_{ir}}}, \quad (3)$$

where: q is the experimental reaction rate per atom per second, N is the number of target isotope nuclei, η is the detector efficiency, ϵ is gamma branching ratio, λ is the decay constant, ΔT is the time between the end of irradiation and start of HPGe measurement, $C(T_m)$ is the measured number of counts, T_m is the time of measurement by HPGe, and T_{ir} is the time of irradiation. The ^{252}Cf SACS were derived from experimental reaction rate by means of the following formula:

$$\bar{\sigma} = C\frac{\int_E \sigma(E)\Phi(E)dE}{\int_E \Phi(E)dE}, \quad (4)$$

where C denotes the correction factor, $\Phi(E)$ is the calculated neutron spectrum, $\sigma(E)$ is the cross section and $\bar{\sigma}$ is the spectral averaged cross section. The correction factor C represents correction caused by spectral shift, flux loss and self-shielding together. It is calculated using MCNP6. Detailed description of the method can be found in [8].

3 Results

Table 1 shows comparison of ^{252}Cf SACS measured in this work with calculations using IRDFF-1.05 library. The agreement with experiment is very good for all presented reactions.

Table 1. Comparison of ^{252}Cf SACS measured in this work with data calculated using IRDFF-1.05.

Reaction	SACS	Diff.	XS Unc.	Sp. Unc.
^{54}Fe(n,p)	74.6	-3.12%	3.06%	0.81%
^{89}Y(n,2n)	0.164	-0.52%	1.24%	4.35%
^{90}Zr(n,2n)	0.102	1.51%	0.91%	5.31%
^{23}Na(n,2n)	3.85E-03	2.36%	1.27%	8.43%

The results were published in [8]. Table 2 shows same comparison for ^{235}U SACS using CIELO-g6 PFNS. Prompt fission neutron spectra (PFNS) of ^{235}U was taken from CIELO-g6 library and cross sections of the appropriate reactions was taken from IRDFF-1.05 library. All experimental results are in a good agreement with calculations using CIELO-g6 library within uncertainties. For details see [10], [11] and [7]. The final result significantly depends on library used for ^{235}U PFNS, see Table 3. If the ENDF/B-VII.1 for ^{235}U is used, the discrepancy is higher with higher mean energy of the reaction.

Table 2. Comparison of ^{235}U SACS measured in this work with data calculated using CIELO-g6 PFNS and IRDFF-1.05 cross sections.

Reaction	SACS	Diff.	XS Unc.	Sp. Unc.
^{54}Fe(n,p)	74.6	-3.12%	3.19%	1.24%
^{89}Y(n,2n)	0.164	-0.52%	1.30%	7.87%
^{90}Zr(n,2n)	0.102	1.51%	0.92%	8.84%
^{23}Na(n,2n)	3.85E-03	2.36%	1.27%	11.65%

Table 3. Comparison of ^{235}U SACS measured in this work with data calculated using ENDF/B-VII.1 PFNS and IRDFF-1.05 cross sections.

Reaction	SACS	Diff.	XS Unc.	Sp. Unc.
^{54}Fe(n,p)	74.6	4.9%	3.15%	7.33%
^{89}Y(n,2n)	0.164	-9.03%	1.36%	90.97%
^{90}Zr(n,2n)	0.102	-12.8%	0.94%	97.60%
^{23}Na(n,2n)	3.85E-03	-20.3%	1.29%	113.77%

Table 4 summarizes experimental ratios of ^{252}Cf and ^{235}U SACS for ^{54}Fe(n,p)^{54}Mn, ^{89}Y(n,2n)^{88}Y, ^{90}Zr(n,2n)^{89}Zr and ^{23}Na(n,2n)^{22}Na reactions.

Table 4. Experimental ratios of ^{252}Cf and ^{235}U SACS

Reaction	E50% [MeV]	Ratio	Uncertainty
^{54}Fe(n,p)^{54}Mn	4.36163	1.06	3.10%
^{89}Y(n,2n)^{88}Y	13.8359	2.14	3.38%
^{90}Zr(n,2n)^{89}Zr	14.3544	2.14	4.42%
^{23}Na(n,2n)^{22}Na	15.3666	2.28	4.88%

The data of individual reactions are plotted as a function of the averaged mean response energy E50% in ^{252}Cf (sf) and ^{235}U CIELO-g6 neutron spectrum, see Figure 3. It corresponds to the energy where integration of the product of spectra and cross section reaches 50%. The experimental ratios show a smooth increasing of ratio with increasing E50%. The variation of the ratios as a function of energy up to a value of 2.28, reflects the hardness of the ^{252}Cf spectrum compared to the ^{235}U spectrum.

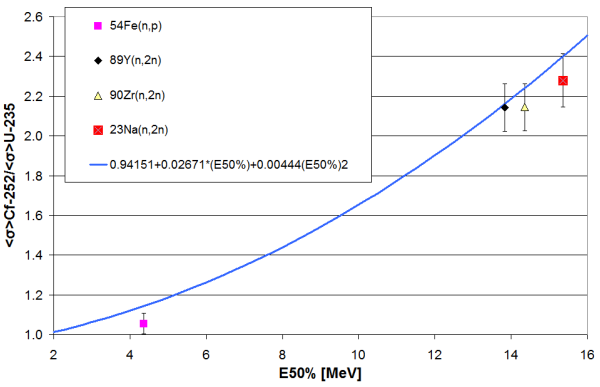

Figure 3. Ratios of the ^{252}Cf and ^{235}U SACS in dependence on E50% fitted with quadratic function.

4 Conclusions

Presented results can be used to validate IRDFF-1.05 library. Validated reactions can be also used to check the consistency of evaluated reference neutron spectra using measured set of SACS ratios, especially for E50% higher than 8 MeV. The results significantly differ for ^{235}U ENDF/B-VII.1 and CIELO-g6 PFNS. The experimental results are in a good agreement with CIELO-g6 PFNS unlike ENDF/B-VII.1 PFNS. It suggests that ^{235}U PFNS is better reproduced in CIELO-g6 library.

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