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Measurement of the ^{23}Na(n,2n) cross section in ^{235}U and ^{252}Cf fission neutron spectra

Michal Košťál, Martin Schulc, Vojtěch Rypar, Evžen Losa, Marie Švadlenková, Petr Baroň, Bohumil Jánský, Evžen Novák, Martin Mareček, and Jan Uhlíř

Research Centre Rez, Hlavní 130, 25068 Husinec-Řež, Czech Republic

Abstract. The presented paper aims to compare the calculated and experimental reaction rates of ^{23}Na(n,2n)^{22}Na in a well-defined reactor spectra and in the spontaneous fission spectrum of ^{252}Cf. The experimentally determined reaction rate, derived using gamma spectroscopy of irradiated NaF sample, is used for average cross section determination.Estimation of this cross-section is important as it is included in International Reactor Dosimetry and Fusion File and is also relevant to the correct estimation of long-term activity of Na coolant in Sodium Fast Reactors. The calculations were performed with the MCNP6 code using ENDF/B-VII.0, JEFF-3.1, JEFF-3.2, JENDL-3.3, JENDL-4, ROSFOND-2010, CENDL-3.1 and IRDFF nuclear data libraries. In the case of reactor spectrum, reasonable agreement was not achieved with any library. However, in the case of ^{252}Cf spectrum agreement was achieved with IRDFF, JEFF-3.1 and JENDL libraries.

1. Introduction

An ongoing process of evaluation, reevaluation and measurement of selected dosimetry cross sections is covered by the Nuclear Data Section of IAEA. Many reactor dosimetry reactions in the International Reactor Dosimetry and Fusion File (IRDFF library) [1] were already evaluated, but a substantial portion of them have not yet been validated. One of them is the ^{23}Na(n,2n) reaction. Even though essential cross section data have been obtained by direct measurement, there is considerable interest in ^{235}U and ^{252}Cf spectral averaged cross-sections (SACS hereafter). These SACS data for ^{23}Na(n,2n) reaction were not measured yet. In EXFOR database, there are only monoenergetic cross sections and SACS result in fast reactor flux [2].

The LR-0 [3] is multipurpose experimental reactor operated by Research Center Rez. It is known for VVER-1000 mock-up experiments used for reactor dosimetry experiments. Moreover, there can also be assembled special cores for other types of experiments, like those intended for study of neutronic parameters of new reactor systems coolants or the measurement of spatial distribution of fission density in special arrangements. These special arrangements also have the advantage that they have well defined neutron spectra, which are in the higher energies nearly identical with ^{235}U fission spectra [4]. The correction to spectral shift between ^{235}U and reactor spectra is determined calculationally and is 0.965.

A well defined neutron spectrum can be also obtained from ^{252}Cf neutron source which is available at the Research Center Rez. Both available neutron fields have been used for determination of ^{23}Na(n,2n) SACS. The influence of different shapes of the ^{235}U and ^{252}Cf fission spectra can be observed in the comparison of SACS averaged over the (n,2n) threshold to 20 MeV.

The presented neutron SACS of ^{23}Na(n,2n) reaction in the both reactor and ^{252}Cf spectrum is derived from the experimentally determined reaction rates. Those are derived from Net Peak Areas (NPA) measured using the semiconductor HPGe spectroscopy. ^{22}Na can originate not only from (n,2n) reaction but also from (γ,n) reaction. It was proven that this contribution to ^{22}Na production rate can be neglected [5].

The experimental reaction rates are compared with reaction rates calculated using various nuclear data libraries.

2. Experimental methods

The studied sodium (in the form of encapsulated NaF) was irradiated in a well-defined spectrum in the center of a special core assembled in the LR-0 reactor (core in Fig. 1). It is a well-defined core with an already characterized reactivity [6], fission rates distribution [7] and also neutron spectra [8] (see Fig. 2).

Sodium fluoride was chosen as a suitable, chemically stable form of sodium. Another advantage of this choice is also the suitable neutronic properties of fluorine in the aluminum can. These favorable properties are reflected by the small self-shielding multiplication correction factor of only 0.991 for reactor measurement and 0.999 for ^{252}Cf measurement.

Irradiation in the LR-0 reactor details

The used NaF capsule has an inner diameter of 80 mm and a height of 106.1 mm. The capsule itself has a longitudinal wall thickness of 1 mm, while the bottom and top end are 3 mm thick. The density of NaF powder was 1.208 g/cm^3, thus the activated mass consisted in 644 g of NaF. The core in this experiment consists of a special dry assembly with

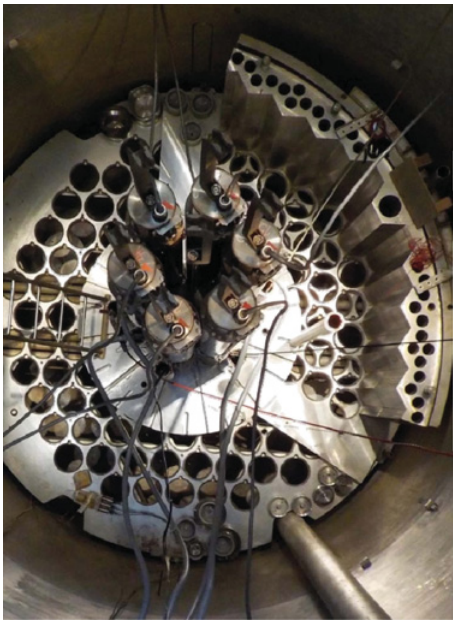

Figure 1. Insertion core at LR-0 reactor.

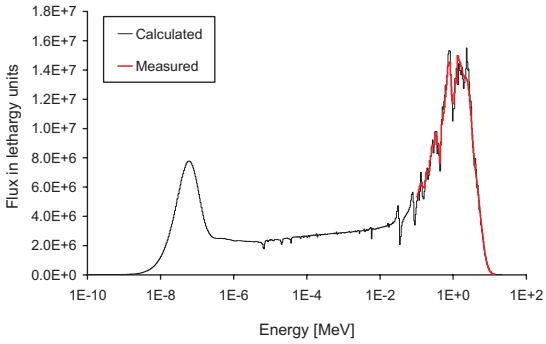

Figure 2. Calculated and measured spectra in irradiation position in LR-0 reactor.

an irradiated capsule in the centre surrounded by 6 uranium fuel assemblies with nearly 3.3% ^{235}U enrichment. The spacer grid and also the experimental dry assembly follow a hexagon lattice with an internal dimension of 23.4 cm, the lattice pitch being 23.6 cm. Hence between adjoining assemblies, there is a 2 mm gap. The capsule is placed in the centre of the special dry assembly. Axially, the capsule lower end is positioned 22.4 cm above the fissile column lower end. Thus the irradiated sample axial position, related to the fission column lower end extends from 22.7 cm to 33.3 cm.

The monitoring activation foils were placed on the outer surface of the capsule at a height of 28 cm. Two kinds of activation material were employed: gold (1% Au in Al) and nickel (100% Ni). The following activation reactions were used: ^{197}Au (n,γ) ^{198}Au and ^{58}Ni (n,p) ^{58}Co. From comparison with measured values, the scaling factor K was determined [8]. Its physical meaning is the core neutron emission rate. The average scaling factors determined from Au and Ni differ by approximately 0.3%. The total uncertainty of the scaling factor is about 2% [5]. The irradiation of the capsule was performed at a power of approximately 5W, during almost 21 h.

A semiconductor HPGe gamma spectroscopy was used for the determination of the ^{22}Na amount in the irradiated sample. The activity of NaF powder was measured in the Marinelli beaker placed on the upper cap of a coaxial HPGe detector placed in lead shielding with a thin copper lining. As the irradiated sample had only activity of 0.55 Bq and it takes four weeks,to obtain satisfactory statistics. Background radiation was measured with an empty Marinelli beaker.

^{252}Cf irradiation details

In the case of irradiation with ^{252}Cf source, the source was placed inside the capsule containing NaF with similar dimensions as in reactor case but with different filling. The mass was 529.5 g. The irradiation lasted 6 weeks divided into 2 irradiation batches 2 weeks and 4 weeks. The induced activity was about 4.9 Bq, thus the subsequent HPGe measurement takes about 2.5 days to obtain satisfactory NPA. In this experiment, an activation foil monitor is not needed because the emission is defined (9.10^8 n/s at the beginning of irradiation). The neutrons emission rate of the source was 9.53E8 n/s on August 13th 2015 according to the data in Certificate of Calibration (NPL UK) involving manganese sulphate bath.

Gamma spectroscopy details

The efficiency calibration of the HPGe detector was calculated using the MCNP6 code [10]. The measured spectra were analyzed by Genie 2000 software. The reaction rate was inferred from the measured NPA [5]. All NPAs were corrected to true summation coincidences as the 1274.5 keV peak coincides with the annihilation peak 511 keV. This correction was calculated in MCNP6.

3. Calculation methods

The simulations of neutron and photon flux distribution in the irradiated sample were performed in critical calculations using the MCNP6 Monte Carlo code and ENDF/B-VII.0 data library. Satisfactory agreement of the calculated k_{eff} for a model using the experimentally determined moderator height with a critical value of 1.003 was obtained. This indicates that the fission source uncertainty has only a minor role in the reaction rates results. The results were normalized to source neutron by dividing the reaction rate by the resulting k_{eff}, see [11]. The normalization to neutron flux level is performed via multiplication by the mentioned scaling factor K.

The NaF salt sample and neutron spectra were modeled using these selected nuclear data libraries: ENDF/B-VII.0 [12], JEFF-3.1 [13], JEFF-3.2 [14], JENDL-4 [15], JENDL-3.3 [16], ROSFOND-2010 [17], CENDL-3.1 [18], CIELO [19] and IRDFF [1]. Nuclear data libraries were processed using the NJOY99 code [20].

4. Results

In the case of reactor spectrum the cross section averaged in spectrum above 10 MeV (threshold of the reaction under study) was determined to be 2.9 mb, the cross section averaged in LR-0 reactor spectrum was evaluated as 0.96 μb. This value is determined with an uncertainty of 4.7% [5], where the major portion is formed by the uncertainty of the scaling factor used for absolute

Table 1. Reaction rates, C/E-1 and uncertainty in reactor spectrum using ENDF/B-VII.0 fission spectra.

Library	RR [1/atom]	C/E-1	Unc.
ENDF/B-VII.0	2.73E-34	33.0%	5.1%
JEFF-3.1	1.75E-34	−14.8%	5.1%
JEFF-3.2	2.05E-34	−0.4%	5.1%
JENDL-3.3	1.75E-34	−14.9%	5.1%
JENDL-4	1.75E-34	−14.9%	5.1%
ROSFOND-2010	1.91E-34	−7.0%	5.1%
CENDL-3.1	2.00E-34	−2.7%	5.1%
IRDFF	1.80E-34	−12.5%	5.1%

Table 2. Reaction rates, C/E-1 and uncertainty in reactor spectrum with CIELO fisison spectra evaluation.

Library	RR [1/atom]	C/E-1	Unc.
ENDF/B-VII.0	3.30E-34	62.9%	5.1%
JEFF-3.1	2.05E-34	1.0%	5.1%
JEFF-3.2	2.45E-34	20.7%	5.1%
JENDL-3.3	2.07E-34	2.1%	5.1%
JENDL-4	2.07E-34	2.1%	5.1%
ROSFOND-2010	2.26E-34	11.4%	5.1%
CENDL-3.1	2.36E-34	16.2%	5.1%
IRDFF	2.16E-34	6.7%	5.1%

Table 3. Reaction rates, C/E-1 and uncertainty in ^{252}Cf fission spectrum.

Library	RR [1/atom]	C/E-1	Unc.
ENDF/B-VII.0	5.54E-32	53.5%	4.3%
ENDF/B-VII.1	5.74E-32	59.0%	4.3%
JEFF-3.1	3.51E-32	−2.7%	4.3%
JEFF-3.2	4.19E-32	16.1%	4.3%
JENDL-3.3	3.59E-32	−0.4%	4.3%
JENDL-4	3.59E-32	−0.4%	4.3%
ROSFOND-2010	3.9E-32	8.1%	4.3%
CENDL-3.1	4.07E-32	12.7%	4.3%
IRDFF	3.73E-32	3.3%	4.3%
TENDL	3.1E-32	−17.0%	4.3%

flux determination. The calculated values of the reaction rates are presented in Table 1 and Table 2. These tables compare reaction rates achieved with different fission spectra fixing ^{23}Na(n,2n) cross section library. The C/E-1 comparison is also shown, where C corresponds to calculation with selected library and E is experimental reaction rate averaged with reactor spectra. A reasonable agreement is not reached with any library. It is important to note that using new CIELO evaluation of prompt fission neutron spectra of ^{235}U and ^{238}U gives better results than ENDF/B-VII.0, see Table 2.

In the case of ^{252}Cf spontaneous fission spectrum the experimental cross section averaged in spectrum above 10 MeV was determined to be 3.54 mb. The cross section averaged in ^{252}Cf reactor spectrum was evaluated as 6.32 μb. These values are determined with an uncertainty of 3.5%. The calculated values with C/E-1 comparison of the reaction rates are presented in Table 3.

Unlike the previous case, the reasonable agreement is reached with IRDFF, JENDLs and JEFF-3.1 libraries. Other libraries perform much worse. Figure 3 shows the energy dependence of ^{23}Na (n,2n) cross section in various

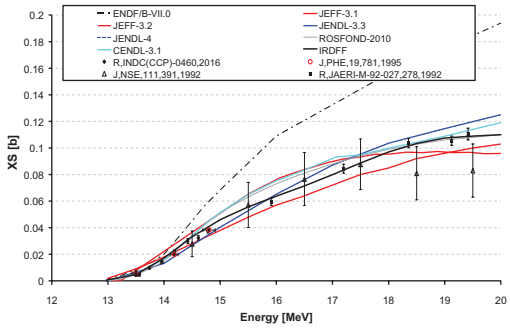

Figure 3. ^{23}Na(n,2n) cross section energy dependent in various libraries.

Table 4. Ratio of ^{23}Na(n,2n) SACS in ^{252}Cf spectra and ^{235}U(n_{th},fiss) spectra in ENDF/B-VII.0 and CIELO prompt fission neutron spectra.

Ratio of XS ^{252}Cf / XS ^{235}U				
	ENDF/B-VII.0.		CIELO	
	Mean	C/E-1	Mean	C/E-1
ENDF/B-VII.0	1.55	19.5%	1.27	−1.7%
JEFF	1.48	14.2%	1.26	−3.0%
JEFF-3.2	1.51	16.5%	1.25	−3.1%
JENDL-3.3	1.52	17.0%	1.27	−1.7%
JENDL-4	1.52	17.0%	1.27	−1.7%
ROSFOND-2010	1.51	16.3%	1.27	−2.2%
CENDL-3.1	1.50	15.9%	1.27	−2.3%
IRDFF	1.53	18.1%	1.26	−2.4%

libraries and various monoenergetic cross section data, there are very large differences amongst the libraries.

Table 4 shows the ratio of SACS in ^{252}Cf (s.f.) spectra and SACS in ^{235}U(n_{th},fiss). The SACS for ^{235}U is obtained from SACS in LR-O spectra which is corrected to spectral shift between ^{235}U(n_th,fiss) in region above 10MeV. Using such correction SACS of ^{23}Na(n,2n) in ^{235}U(n_{th},fiss) gives result of 3.82 μb.

5. Conclusions

In this work, the cross section of ^{23}Na(n,2n) reaction was derived from measured data in a reactor spectrum as well as in a ^{252}Cf fission neutron spectrum. In the case of ^{252}Cf spontaneous fission spectrum the experimental SACS was determined to be 6.32 μb with 3.5% uncertainty. In the case of reactor spectrum the experimental SACS was determined to be 0.96 μb. When used spectral shift correction the experimental SACS in ^{235}U(n_{th},fiss), it was determined to be 3.8 μb. These values are determined with an uncertainty of 4.7%. Reasonable agreement between libraries and experiment was achieved only in ^{252}Cf fission neutron spectrum.

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