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Measurement of Different Neutron Induced Reactions in ^{252}Cf Spontaneous Fission Neutron Spectrum

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Abstract - *This work aims to compare various measured neutron induced reaction rates with computed ones in different nuclear data libraries using MCNP6 for some materials used in nuclear industry. ^{252}Cf spontaneous fission source with defined emission was used as a neutron source for all reactions under the investigation.*

I. INTRODUCTION

The presented paper compares various measured neutron induced reaction rates with computed ones in different nuclear data libraries for some materials interesting for nuclear industry. It deals with following materials: Aluminium, ^{54}Fe and Sodium. Namely, paper investigates ^{27}Al(n,g), ^{27}Al(n,p), ^{27}Al(n,a), ^{54}Fe(n,p), ^{54}Fe(n,α) and ^{23}Na(n,2n) reactions.

II. DESCRIPTION OF THE ACTUAL WORK

The experimental reaction rates for neutron induced activation products of ^{27}Al, ^{54}Fe and ^{23}Na are compared with the reaction rates calculated in MCNP6 [1] using various nuclear data libraries. The experimental reaction rates are derived from Net Peak Areas (NPA) measured using a semiconductor HPGe (High Purity Germanium) spectroscopy. As a neutron source, an encapsulated ^{252}Cf was used either as a bare source or moderated on a heavy water sphere. The Cf-252 has a half-life of 2.645 years, 3.09 % of the isotope decays by spontaneous fission releasing approximately 3.7 neutrons per fission. The neutrons emission rate of the source was 9.53E8 n/s on August 13th 2015 according to the data in Certificate of Calibration involving manganese sulphate bath. Brief introduction of explored materials and experimental arrangement follows.

1. Aluminium

Aluminium is a structural material which is suitable for a use in research reactors at low power and temperature operating range because it has low thermal neutron absorption and it is low-cost. Often it is used as a cladding material of nuclear fuel.

First reaction under study was ^{27}Al(n,g)^{28}Al reaction which is primarily caused by thermal neutrons. This activating reaction is used for determining of Aluminium content in samples (for example a sample of rock). Neutron thermal field was achieved by moderating neutrons emitted from ^{252}Cf in heavy water sphere with diameter d = 299.97 mm covered with steel coating of thickness t = 0.078 cm. It contains 99.36 wt. % of heavy water, rest is light water. The sketch of experimental arrangement can be seen in the Fig. 1.

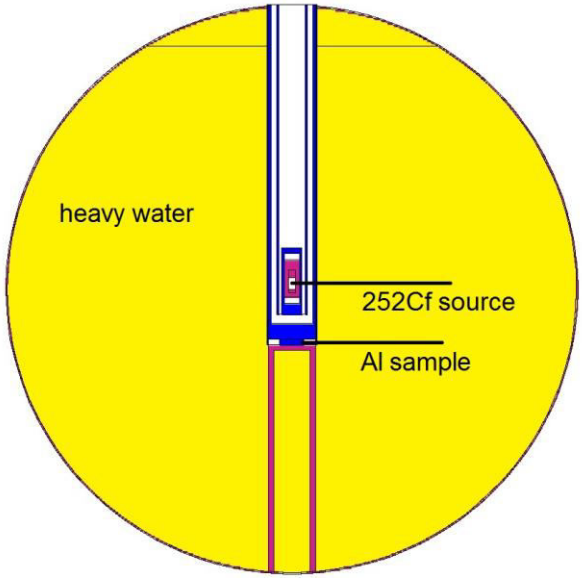

Fig. 1. Experimental activation sample arrangement for ^{27}Al(n,g)^{28}Al reaction in the heavy water sphere.

Other reactions under study were those triggered by fast neutrons, i.e. ^{27}Al(n,p)^{27}Mg and ^{27}Al(n,α)^{24}Na reactions. In the case of ^{24}Na HPGe measurement the detector efficiency to 1368.6 keV peak was corrected to true summation coincidence with 2754 keV peak using method in [2]. This correction was calculated in MCNP6. The sketch of the experimental arrangement is in the Fig. 2.

Figure 3 compares pure ^{252}Cf neutron fission spectrum with calculated neutron spectrum in position of the irradiated sample. It is clearly shown that the spectra are same within the uncertainties justifying the assumption that measurement was carried out in pure ^{252}Cf neutron spectrum.

Al samples used for the irradiation had a cylindrical shape with diameter d = 12.74 mm, height h = 3.07 mm and mass m = 0.9955 g. The ^{252}Cf neutron source used for the measurement was transported from the container to the measuring position by a pneumatic flexo-rabbit system. Each sample was placed in the center of the bottom of flexo-rabbit ending.

More technical details concerning measurements on Aluminium can be found in [3].

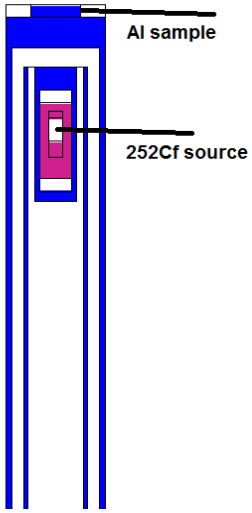

Fig. 2. Experimental arrangement for reactions induced by fast neutrons.

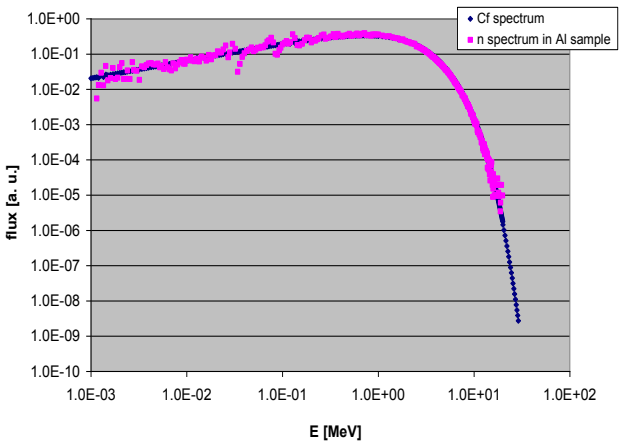

Figure 3: Comparison of pure ^{252}Cf spectrum with neutron spectrum calculated by MCNP6 for Al sample.

2. Fe-54

The ^{54}Fe(n,α)^{51}Cr reaction contributes to the helium accumulation in steel, thus this reaction is of technical importance for reactor dosimetry and material damage studies.

Neutron reactions leading to radioactive products are frequently used to measure integrated fast neutron flux in reactor environment. In this respect the ^{54}Fe(n,p) ^{54}Mn reaction appears to be very attractive, the cross section for fission neutrons is relatively large (approximately 60 mb), ^{54}Mn has a long half life (312 days), and the physical and chemical properties of iron are convenient for the purpose.

^{54}Fe sample with isotopic abundance of 99.6 % used for irradiation had a cylindrical shape with diameter d = 12.74 mm, height h = 1 mm and mass m = 0.9969 g. The experimental arrangement for exploring both reactions on ^{54}Fe was same as in case of exploring fast neutrons induced reactions on Aluminium.

3. Sodium

Reaction of interest was the ^{23}Na(n,2n)^{22}Na reaction. Estimation of this cross-section is important as it is included in International Reactor Dosimetry and Fusion File [4] and is also relevant to the correct estimation of long-term activity of Na coolant in Sodium Fast Reactors.

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 a relatively low flux loss and small spectral shift correction factor of only 1.011. The energy dependent shielding factor for ^{252}Cf spectra is plotted in the Figure 4.

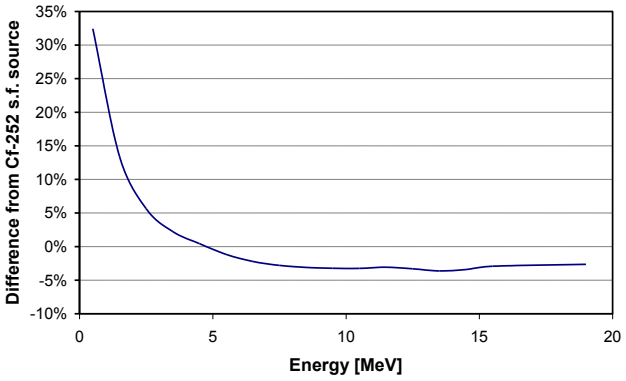

Fig. 4. Self-shielding effect of Al capsula and NaF salt in ^{252}Cf spontaneous fission spectrum.

In this case, the source was placed inside the capsule containing NaF with mass 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 following HPGe measurement takes about 2.5 days to obtain satisfactory NPA. NPA was corrected to true summations coincidences because of coincidence of 1274.5 keV photons with annihilation 511 keV photons. This correction was calculated in MCNP6 [1]. The measurement procedure follows that published in [5]. The scheme of this measurement can be found in the Fig.5.

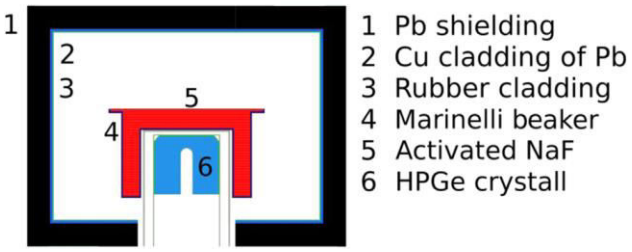

Fig. 5. Scheme of HPGe measurement of irradiated NaF in Marinneli beaker.

4. Calculation methods

All calculations were done with MCNP6 [1] transport code with various neutron nuclear data libraries. Neutron nuclear data libraries which were employed for calculation and mutual comparison include ENDF/B-VII.0 [6], ENDF/B-VII.1 [7], CENDL-3.1 [8], JENDL-3.3 [9], JENDL-4 [10], JEFF-3.2 [11], TENDL-2013 [12], ROSFOND-2010 [13], IRDFF [4] and CIELO [14]. All of the mentioned libraries were processed using NJOY code [15]. The influence of libraries was studied by changing only relevant cross-section in the sample. The input Cf-252 spontaneous neutron fission spectrum was taken from IRDFF webpage [4].

The efficiency calibration of the HPGe detector was calculated using the MCNP6 code. The samples were placed on the upper cap of the detector. The count times of HPGe spectrometry for all nuclides of interest were set so long as to obtain reasonable statistic uncertainty, in most cases less than 1.0 %. The reaction rate during irradiation was determined using Eq. (1)

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

where q is experimental reaction rate, N is number of target isotope nuclei, η is detector efficiency, ε is gamma branching ratio, λ is decay constant, ΔT is time between the end of irradiation and start of HPGe measurement, $C(T_m)$ is measured number of counts, T_m is time of measurement by HPGe and T_{ir} is time of irradiation.

III. RESULTS

All reactions under study are consecutively explored by introducing both calculated and experimentally determined reaction rate together with the ratio of calculation over experiment (C/E-1) and uncertainty (Unc.). The influence of libraries was studied by changing only the cross-section of a studied material in the sample.

1. ^{27}Al(n,g) reaction

To explore this reaction, ^{252}Cf neutron spectrum was moderated in the heavy water sphere. Reasonable agreement was reached with the use of all libraries, see Table I.

Table I. Calculation and C/E-1 comparison with experimental data

^{27}Al(n, g)^{28}Al	q[s^{-1} atom^{-1}]	C/E-1	Unc.
EXPERIMENT	7.693E-20		2.5 %
ENDF/B-VII.0	7.851E-20	2.06 %	1.2 %
JEFF-3.2	7.841E-20	1.93 %	1.2 %
JENDL-3.3	7.876E-20	2.38 %	1.2 %
JENDL-4	7.888E-20	2.54 %	1.2 %
ROSFOND-2010	7.873E-20	2.34 %	1.2 %
CENDL-3.1	7.866E-20	2.26 %	1.2 %
ENDF/B-VII.1	7.851E-20	2.06 %	1.2 %

2. ^{27}Al(n,p) reaction

The calculated values of the reaction rates and C/E-1 comparisons with corresponding uncertainties are presented in the Table II. The best agreement within uncertainty is reached with the use of JENDL-4 library, the worst with JEFF-3.2 library. Possible contribution from impurities stemming from reaction ^{26}Mg(n,γ)^{27}Mg was analyzed. This contribution was found negligible.

The energy share of ^{27}Mg production from (n,p) reaction is listed in Table III. It can be observed that there are large differences in energy distribution of reaction rates between ENDF/B-VII.0 and JENDL-4.0 libraries. Major contribution to total reaction rate in ^{252}Cf spectrum is in energy range 4-8 MeV.

Table II. Calculation and C/E-1 comparison with experimental data

^{27}Al(n, p) ^{27}Mg	q[s^{-1} atom^{-1}]	C/E-1	Unc.
EXPERIMENT	2.875E-20		2.5 %
ENDF/B-VII.0	2.692E-20	-6.37 %	1.2 %
JEFF-3.2	2.684E-20	-6.64 %	1.2 %
JENDL-3.3	2.921E-20	+1.60 %	1.2 %
JENDL-4	2.911E-20	+1.25 %	1.2 %
ROSFOND-2010	2.692E-20	-6.37 %	1.2 %
CENDL-3.1	2.925E-20	+1.74 %	1.2 %
TENDL-2013	2.720E-20	-5.39 %	1.2 %
ENDF/B-VII.1	2.691E-20	-6.40 %	1.2 %
IRDFF	2.721E-20	-5.35 %	1.2 %

Table III. Reaction rates energy distributions

^{27}Al(n, p) ^{27}Mg	ENDF/B-VII.1	JENDL-4.0
2-4 MeV	12.48 %	9.61 %
4-6 MeV	41.58 %	40.43 %
6-8 MeV	30.02 %	32.28 %
8-12 MeV	14.96 %	16.70 %

12-15 MeV	0.90 %	0.87 %
15-20 MeV	0.10 %	0.11 %

3. ^{27}Al(n,α) reaction

The calculated values of the reaction rates are presented in Table IV. The C/E-1 comparison together with uncertainty is also shown. The best agreement within uncertainty is reached with the use of JENDL-4 and CENDL-3.1 libraries, the worst with ROSFOND-2010 and JEFF-3.2 libraries.

Possible contribution from impurities stemming from reaction ^{23}Na(n,γ)^{24}Na was also analyzed. This contribution was found at most 0.02 % of total ^{24}Na yield.

Table IV. Calculation and C/E-1 comparison with experimental data

^{27}Al (n, α) ^{24}Na	q[s^{-1} atom^{-1}]	C/E-1	Unc.
EXPERIMENT	5.412E-21		2.7 %
ENDF/B-VII.0	5.683E-21	5.00 %	1.2 %
ENDF/B-VII.1	5.683E-21	5.00 %	1.2 %
JEFF-3.2	5.726E-21	5.81 %	1.2 %
JENDL-3.3	5.478E-21	1.25 %	1.2 %
JENDL-4	5.478E-21	1.25 %	1.2 %
ROSFOND-2010	5.726E-21	5.81 %	1.2 %
CENDL-3.1	5.480E-21	1.25 %	1.2 %
TENDL-2013	5.608E-21	3.66 %	1.2 %
IRDFF	5.582E-21	3.14 %	1.2 %

4. ^{54}Fe(n,p) reaction

The best agreement with the experiment was achieved using ROSFOND, JEFF-3.2 and CIELO libraries, all within uncertainty. The worst results give JENDL, TENDL-2013 and ENDF/B-VII.1 libraries, see Table V.

Table V. Calculation and C/E-1 comparison with experimental data

^{54}Fe(n,p) ^{54}Mn	q[s^{-1} atom^{-1}]	C/E-1	Unc.
EXPERIMENT	4.835E-19		2.5 %
ENDF/B-VII.1	5.447E-19	12.65 %	1.2 %
JEFF-3.2	4.844E-19	0.19 %	1.2 %
JENDL-3.3	5.513E-19	14.03 %	1.2 %
JENDL-4	5.513E-19	14.03 %	1.2 %
ROSFOND-2010	4.844E-19	0.19 %	1.2 %
CENDL-3.1	5.443E-19	12.57 %	1.2 %
TENDL-2013	5.492E-19	13.59 %	1.2 %
CIELO	4.925E-19	1.87 %	1.2 %
IRDFF	5.333E-19	10.31 %	1.2 %

5. ^{54}Fe(n,α) reaction

There are very large differences among all libraries for this reaction. The agreement within uncertainty was not achieved using any library. The best agreement gives TENDL-2013 library, although difference is 6.52 %, see Table VI. The energy share of ^{51}Cr production from (n, α) reaction for JEFF-3.2, CIELO and TENDL-2013 libraries is listed in Table VII. There are large differences in energy distribution of reaction rates among all mentioned libraries.

Table VI. Calculation and C/E-1 comparison with experimental data

^{54}Fe (n, α) ^{51}Cr	q[s^{-1} atom^{-1}]	C/E-1	Unc.
EXPERIMENT	6.048E-21		3.5 %
ENDF/B-VII.1	7.111E-21	17.57 %	1.2 %
JEFF-3.2	4.245E-21	-29.81 %	1.2 %
JENDL-3.3	6.820E-21	12.75 %	1.2 %
JENDL-4	6.819E-21	12.74 %	1.2 %
ROSFOND-2010	4.246E-21	-29.81 %	1.2 %
CENDL-3.1	6.873E-21	13.63 %	1.2 %
TENDL-2013	6.443E-21	6.52 %	1.2 %
CIELO	7.873E-21	30.17 %	1.2 %
IRDFF	6.698E-21	10.74 %	1.2 %

Table VII. Reaction rates energy distributions

^{54}Fe(n, α) ^{51}Cr	JEFF-3.2	CIELO	TENDL -2013
3.5-6 MeV	12.17 %	19.60 %	15.02 %
6-8 MeV	34.82 %	42.00 %	43.80 %
8-10 MeV	32.19 %	24.70 %	26.93 %
10-15 MeV	19.89 %	13.16 %	13.55 %
15-20 MeV	0.93 %	0.54 %	0.70 %

6. ^{23}Na(n,2n) reaction

Figure 6 shows energy dependence of ^{23}Na (n,2n) cross section in various libraries and very large differences among libraries. ^{22}Na can originate not only by (n,2n) reaction but also by (γ,n) reaction. However, (γ,n) reaction has very high threshold, thus it does not contribute to ^{22}Na production.

The experimental cross section averaged in ^{252}Cf spontaneous fission spectrum above 10 MeV was determined to be 3.54 mb. This value is determined with an uncertainty of 3.5 %. The calculated values with C/E-1 comparison of the reaction rates are presented in Table VIII. Reasonable agreement is achieved only with IRDFF, JENDL and JEFF-3.1 libraries.

Table VIII. Calculation and C/E-1 comparison with experimental data			
^{23}Na(n,2n) ^{22}Na	q[s^{-1} atom^{-1}]	C/E-1	Unc.
EXPERIMENT	3.24E-23		3.5 %
ENDF/B-VII.0	4.97E-23	53.5 %	4.3 %
ENDF/B-VII.1	5.15E-23	59.0 %	4.2 %
JEFF-3.1	3.15E-23	-2.7 %	4.2 %
JEFF-3.2	3.76E-23	16.1 %	4.3 %
JENDL-3.3	3.23E-23	-0.4 %	4.3 %
JENDL-4	3.23E-23	-0.4 %	4.3 %
ROSFOND-2010	3.50E-23	8.1 %	4.3 %
CENDL-3.1	3.65E-23	12.7 %	4.3 %
IRDFF	3.35E-23	3.3 %	4.3 %

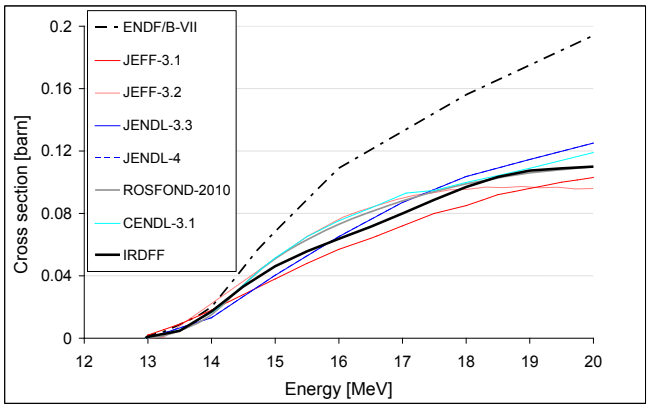

Fig. 6. ^{23}Na(n,2n) cross section energy dependence in various libraries.

IV. CONCLUSIONS

In this work, several nuclear reactions were investigated. Concerning reactions on Aluminium, in the fast region, reaction (n,p) gives the highest differences among libraries. The best agreement within uncertainty is reached with use of JENDL-4 library, in contrast to JEFF-3.2 library which is the worst. It can be explained by large differences in energy distribution of reaction rates between the libraries.

The cross section of ^{23}Na(n,2n) reaction was derived from measured data in ^{252}Cf fission neutron spectrum. The experimental cross section averaged in spectrum above 10 MeV was determined to be 3.54 mb with 3.5 % uncertainty.

The large discrepancies among libraries and experiment were observed for ^{23}Na(n,2n), ^{54}Fe(n,α) and ^{54}Fe(n,p) reactions.

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