

MEASUREMENT OF SOME AVERAGE CROSS SECTIONS FOR ACTIVATION IN THE SPONTANEOUS FISSION NEUTRON FIELD OF ^{252}Cf

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The average cross sections for the reaction $^{115}\text{In}(n, n')$, ^{115m}In , $^{115}\text{In}(n, \gamma)$, ^{116m}In , $^{113}\text{In}(n, n')$, ^{113m}In , $^{197}\text{Au}(n, \gamma)$, ^{198}Au , $^{111}\text{Cd}(n, n')$, ^{111m}Cd , $^{110}\text{Cd}(n, \gamma)$, ^{111m}Cd , $^{27}\text{Al}(n, p)$, ^{27}Mg , $^{64}\text{Zn}(n, p)$, ^{64}Cu , $^{68}\text{Zn}(n, \gamma)$, ^{69m}Zn , $^{138}\text{Ba}(n, \gamma)$, ^{139}Ba , $^{135}\text{Ba}(n, n')$, $^{135m}\text{Ba} + ^{134}\text{Ba}(n, \gamma)$, ^{135m}Ba , $^{58}\text{Ni}(p, n)$, ^{58}Co and $^{87}\text{Sr}(n, n')$, $^{87m}\text{Sr} + ^{86}\text{Sr}(n, \gamma)$, ^{87m}Sr have been measured in the spontaneous fission spectrum of neutrons from a small ^{252}Cf source. The results are in general agreement with other data in the literature. In the cases where energy differential cross sections were available the average cross sections have been calculated using for the neutron spectrum a Maxwellian with $T = 1.42$ MeV corresponding to an average energy of 2.13 MeV. The comparison of measured and calculated values indicates strong disagreements in the cases of the $^{56}\text{Fe}(n, p)$, $^{138}\text{Ba}(n, \gamma)$ and $^{135}\text{Ba}(n, n') + ^{134}\text{Ba}(n, \gamma)$ reactions.

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1. Introduction

The energy spectrum of spontaneous fission neutrons emitted from ^{252}Cf has reached the status of a standard neutron field¹⁾.

The shape of the neutron spectrum is agreed upon to be basically a Maxwellian with a temperature of 1.42 MeV, corresponding to an average energy of 2.13 MeV. Grundl and Eisenhauer^{2,3)} propose continuous segment corrections to the basic shape but on the other hand recent results of Blinov et al.⁴⁾ suggest that in the region from 1 keV to 1 MeV neutron energy no corrections to a Maxwellian is necessary.

But notwithstanding the small corrections which can be brought to the detailed shape of the spectrum it is a fact that nowadays the spectrum of californium spontaneous fission neutrons is known to an accuracy that is in general superior to the one of the energy differential cross sections for neutron induced reactions in the same energy range.

Therefore the use of a ^{252}Cf source in conjunction with a well calibrated detector set up makes possible integral tests of the energy differential cross section data. This comparison follows from the definition of the average cross section measured in a ^{252}Cf spectrum:

$$\langle \sigma \rangle = \frac{\int_0^{\infty} N(E) \sigma(E) dE}{\int_0^{\infty} N(E) dE} \quad (1)$$

From the relation (1) it is obvious that if the spectrum shape $N(E)$ is accurately known, the comparison between the measured $\langle \sigma \rangle$ and the one calculated via (1) shall bear on the knowledge of $\sigma(E)$. The measurement of $\langle \sigma \rangle$ is a relatively simple measurement many small laboratories can afford but surprisingly the data are still rather scarce and scattered in results.

To be able to study the actual situation in the nuclear data for fission neutrons we have measured the average cross sections for the reactions shown in Table 1.

2. Experiment

Source

The source of ^{252}Cf of 20 μg ^{252}Cf located at the laboratoire de Physique Nucléaire in Rabat has been used. The source strength was declared by the manufacturer (Amersham) to be $4.6 \times 10^7 \text{ s}^{-1} \pm 1.5\%$ on 31 December 1975. To determine the strength of the source at the time of irradiation a half life of 2.638 years⁵⁾ was used.

The ^{252}Cf under the chemical form of oxide is doubly encapsulated in welded stainless steel capsules. The thicknesses of the walls of each capsule is 0.8 mm. The outside dimensions of the source cylinder is 7.8 mm in diameter and 10 mm in height.

Irradiation

For irradiations a special set up was constructed out of thin aluminium, allowing for irradiations at a distance of 4.3 ± 0.15 mm from the vertical axis running through the source in the median plane of the cylinder. The targets were discs of 10 mm in diameter ranging in thickness from 0.1 to 0.7 mm.

To determine the neutron fluence at the target position, the yield of the reaction $^{115}\text{In}(n, n')$ was measured in function of the source-target distance. It was found that the neutron fluence could be satisfactorily calculated assuming a point source and $1/r_e$ dependence, where r_e was found to be 5.2 ± 0.12 mm for sample thickness 0.1 mm.

During irradiation the set up was hung in air 6 meters above ground and from any wall, minimizing thus the scattered neutron background.

The thermal neutron flux was measured in the following way — the $^{115}\text{In}(n, \gamma)$ reaction was measured with and without a cadmium foil. The two yields, after corrections for distance and attenuation of the neutrons in the cadmium foil, were equal within the statistical error. We therefore did not introduce a correction for the thermal neutrons in the measured (n, γ) reactions.

Activity measurement

The activity was measured with a 67 cm^3 GeLi detector. The samples have been placed at 6.5 mm from the detector window. Special corrections were introduced to take care of the counting geometry which is different from the one the efficiency was measured with. The detector efficiency has been determined with an uncertainty of $\pm 1.5\%$.

The gamma ray absorption in the samples was taken care off using measured attenuation coefficients. The dead time corrections were negligibly small in our case since we are working with a low intensity neutron source.

3. Reactions studied

The list of the reactions studied and the nuclear parameters used is presented in Table 1.

The parameters given in Table 1 have been taken from the Table of Isotopes⁶⁾.

4. Results

The average cross sections measured in the present experiment are shown in Table 2 together with available literature data on the same reactions.

The uncertainties quoted for our results are the ones due to counting statistics and the error in the r_e . The uncertainty in the source strength $\approx 1.5\%$ and in the detector efficiency $\approx 1.5\%$ should be added to the errors in Table 2 to obtain

TABLE 1.

TAR-GET	REACTION	$T_{1/2}$	ATOMIC MASS	ISOTOP. ABUND. (%)	γ -RAY DETECT. (keV)	INTENS. OF THE γ (%)
In	$^{115}\text{In} (n,n')^{115m}\text{In}$	4.486h	114.82	95.7	336.0	45.9
	$^{115}\text{In} (n,\gamma)^{116m}\text{In}$	54.3 mn	"	"	1293.4	84.6
	$^{113}\text{In} (n,n')^{113}\text{In}$	99.47mn	"	4.3	391.72	64
Au	$^{197}\text{Au} (n,\gamma)^{198}\text{Au}$	2.697d	196.967	100	411.794	95.5
Cd	$^{111}\text{Cd} (n,n')^{111m}\text{Cd}$	48.6mn	112.4	12.8	245.35	94.2
	$^{110}\text{Cd} (n,\gamma)^{111m}\text{Cd}$	"	"	12.4	"	"
Al	$^{27}\text{Al} (n,p)^{27}\text{Mg}$	9.462mn	26.98154	100	843.7	73
Zn	$^{64}\text{Zn} (n,p)^{64}\text{Cu}$	12.699h	65.38	48.9	511.006	38.6
	$^{68}\text{Zn} (n,\gamma)^{69m}\text{Zn}$	13.76h	"	18.6	438.7	94.8
BaO	$^{138}\text{Ba} (n,\gamma)^{139}\text{Ba}$	82.9mn	137.34	71.9	165.85	22
	$^{134}\text{Ba} (n,\gamma)^{135m}\text{Ba}$	28.7h	"	2.4	268.1	16
	$^{135}\text{Ba} (n,n')^{135m}\text{Ba}$	"	"	6.5	"	"
Ni	$^{58}\text{Ni} (n,p)^{58}\text{Co}$	70.78d	58.7	67.76	810.74	99.44
Fe	$^{56}\text{Fe} (n,p)^{56}\text{Mn}$	2.58h	55.847	91.7	846.74	98.87
[Sr (OH) ₂ , 8H ₂ O]	$^{87}\text{Sr} (n,n')^{87m}\text{Sr}$	2.805h	87.62	7.0	388.4	82
	$^{86}\text{Sr} (n,\gamma)^{87m}\text{Sr}$	"	"	9.9	"	"

Parameters of the reactions studied.

the total uncertainty. However, since the statistical error quoted is much larger than the latter two, the overall uncertainty shall not be much larger than the one presented in Table 2

$$\langle \sigma \rangle = \frac{n_1 \langle \sigma_1 \rangle + n_2 \langle \sigma_2 \rangle}{n_1 + n_2}$$

where n_1 and n_2 are the respective number of nuclei of each isotope and $\langle \sigma_1 \rangle$ and $\langle \sigma_2 \rangle$ are the average cross section for each reaction.

TABLE 2

REACTIONS	PRESENT WORK	Ref. 7	Ref. 9	Ref. 8	Ref. 11	Ref. 12	Ref. 13
$^{115}\text{In} (n,n')^{115m}\text{In}$	196 ± 8	198 ± 5	202.2 ± 12.0	—	199.4 ± 10.5*	188 ± 8	195 ± 5
$^{115}\text{In} (n,\gamma)^{116m}\text{In}$	115.6 ± 5	—	—	—	131.6 ± 5.7	125.0 ± 4.3	123.8 ± 3.6*
$^{113}\text{In} (n,n')^{113m}\text{In}$	168 ± 9	—	—	—	178.3 ± 7.6	—	162.3 ± 4.1*
$^{197}\text{Au} (n,\gamma)^{198}\text{Au}$	78 ± 3	—	—	79.9 ± 2.9	119.1 ± 5.2	95.5 ± 2.3	76.2 ± 1.8
$^{111}\text{Cd} (n,n')^{111m}\text{Cd}$	110.6 ± 4	—	—	—	204 ± 7	—	—
$^{100}\text{Cd} (n,\gamma)^{111m}\text{Cd}$	4.7 ± 0.37	—	—	—	5.04 ± 0.42*	—	—
$^{27}\text{Al} (n,p)^{27}\text{Mg}$	36.2 ± 1.5	40 ± 1	—	—	46.4 ± 2.3	—	—
$^{64}\text{Zn} (n,p)^{64}\text{Cu}$	1.85 ± 0.12	—	—	—	—	—	—
$^{13}\text{Ba} (n,\gamma)^{139}\text{Ba}$	1.30 ± 0.26	—	—	—	3.8 ± 0.4	—	—
$^{135}\text{Ba} (n,n')^{135m}\text{Ba}$	180.9 ± 11	—	—	—	255 ± 28	—	—
$^{134}\text{Ba} (n,\gamma)^{135}\text{Ba}$	95.0 ± 4.5	118 ± 3	105 ± 5	—	110.7 ± 4.8*	—	—
$^{56}\text{Ni} (n,p)^{56}\text{Co}$	1.15 ± 0.08	1.450 ± 0.035	11.8 ± 0.08	—	—	—	—
$^{56}\text{Fe} (n,p)^{56}\text{Mn}$	130 ± 6	—	—	—	182 ± 22	—	—
$^{87}\text{Sr} (n,n')^{87m}\text{Sr}$							
$^{86}\text{Sr} (n,\gamma)^{87m}\text{Sr}$							

Average cross sections $\langle\sigma\rangle$ in 10^{-31}m^2 .

* renormalized in accordance with data from Table 1.

5. Discussion

Comparison of the present results with previous data found in the literature

Table 2 shows our results compared to available data. It is apparent that even for very useful dosimetric reactions like the one on indium we do not have a very large number of data. Actually for most of the reactions from Table 2 there is only one other measurement and only in one case there existed already 5 measurements.

The present results are in general agreement with literature data except for some of the Debrecen data^{10,11}.

It is gratifying to see that in the case of the reactions on indium and gold the data are coherent enough to warrant the calculation of the mean value of the data from different laboratories.

We have done that, rejecting two data for the Au (n, γ) reaction, namely the results of the Debrecen group^{10,11} and the results of Pauw et al.¹². We have rejected those data on the ground that they are coherent neither among themselves nor with the rest of the data as a whole. The mean values obtained in this way are given in Table 3.

TABLE 3.

REACTION	$\langle\sigma\rangle 10^{-31}\text{m}^2$
$^{115}\text{In} (n,n')^{115\text{m}}\text{In}$	196.4 ± 5.0
$^{115}\text{In} (n,\gamma)^{116\text{m}}\text{In}$	124.0 ± 3.5
$^{113}\text{In} (n,n')^{113\text{m}}\text{In}$	169.5 ± 5.5
$^{197}\text{Au} (n,\gamma)^{198}\text{Au}$	78.0 ± 2.0

Mean values of $\langle\sigma\rangle$.

Comparing the mean values in Table 3 with our values from Table 2, we find that only in the case of the $^{115}\text{In} (n, \gamma)$ reaction is our value outside the 68% error quoted.

Comparison between measured and calculated average cross sections

We have calculated the values of $\langle\sigma\rangle$ according to the relation (1) using the $\sigma(E)$ from the evaluated data files ENDF/BIV and ENDF/BV as well as the results of Smith and Meadows¹⁴. The average cross sections were calculated assuming $N(E)$ to be a Maxwellian with the temperature parameter $T = 1.42$ MeV.

The results for the calculated values $\langle\sigma\rangle_{\text{calc}}$ compared with the measured values $\langle\sigma\rangle_{\text{meas}}$ are shown in Table 4 for those reactions where $\sigma(E)$ data were available to us. When two sources for $\sigma(E)$ have been used both values of $\langle\sigma\rangle_{\text{calc}}$ are shown. In the last column the ratio $\langle\sigma\rangle_{\text{meas}}/\langle\sigma\rangle_{\text{calc}}$ has been shown. This ratio should give a measure of the accuracy of the agreement between the integral measurement and the data on $\sigma(E)$. It was pointed out by Smith¹⁵ that the ave-

rage cross sections may be insensitive to some variations in the shape of the excitation functions and that therefore the requisite data base for nuclear energy applications cannot be established by means of integral measurements alone. However if the agreement between $\langle\sigma\rangle_{calc}$ and $\langle\sigma\rangle_{meas}$ may not be always significant, the disagreement most certainly is indicative.

TABLE 4.

REACTIONS	$\langle\sigma\rangle_{meas}/10^{-31} m^2$	$\langle\sigma\rangle_{calc}/10^{-31} m^2$	ORIGIN OF $\sigma(E)$	$\frac{\langle\sigma\rangle_{meas}}{\langle\sigma\rangle_{calc}}$
$^{115}\text{In}(n,n')$	196	183	ENDF/B-V ANL	1.07
$^{115}\text{In}(n,\gamma)$	116	130	ENDF/B-IV	0.89
$^{113}\text{In}(n,n')$	168	143.5	ANL	1.17
$^{197}\text{Au}(n,\gamma)$	78	76.5	ENDF/B-V	1.02
$^{58}\text{Ni}(n,p)$	95.1	116.5	ENDF/B-V	0.82
$^{58}\text{Ni}(n,p)$	95.1	102	ANL	0.93
$^{27}\text{Al}(n,p)$	4.7	5.18	ENDF/B-V	0.91
$^{27}\text{Al}(n,p)$	4.7	4.78	ANL	0.98
$^{64}\text{Zn}(n,p)$	36.2	37	ANL	0.98
$^{56}\text{Fe}(n,p)$	1.15	1.5	ENDF/B-V	0.77
$^{56}\text{Fe}(n,p)$	1.1	1.43	ANL	0.76
$^{138}\text{Ba}(n,\gamma)$	1.3	2.2	ENDF/B-V	0.59
$^{135}\text{Ba}(n,n')$ + $^{134}\text{Ba}(n,\gamma)$	181	230	ENDF/B-V	0.79

Comparison of measured average cross sections with calculated ones using $T = 1.42$ MeV.

The errors to the ratios in Table 4 is between 10 and 15 percents due to both uncertainties in $\langle\sigma\rangle_{meas}$ and $\sigma(E)$. Therefore on the basis of Table 4, the following conclusions can be drawn:

- i) When two sets of $\sigma(E)$ were used the ANL one gives a ratio closer to one;
- ii) According to all measured values of $\langle\sigma\rangle$ for the reaction $^{113}\text{In}(n,n')$ ^{113m}In the ratio indicates too low a value. The disagreement may suggest one or more resonances too narrow to be detected in the $\sigma(E)$ measurements.
- iii) The $\langle\sigma\rangle_{calc}$ values for $^{56}\text{Fe}(n,p)$ seem to be too high compared with our value as well as with the one from Ref. 9. On the contrary the value of Alberts⁷⁾ is in agreement with $\langle\sigma\rangle_{calc}$ comparing data from Table 2 and 4.
- iv) The data for reactions on ^{138}Ba , ^{134}Ba and ^{135}Ba yield $\langle\sigma\rangle_{calc}$ values which are much higher than these measured in our experiment. The lack of more integral data on the some reaction precludes a definite statement about the $\sigma(E)$ in these cases.

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MJERENJE USREDNJENIH UDARNIH PRESJEKA ZA AKTIVACIJU
NEUTRONIMA IZ SPONTANE FISIJE ^{252}Cf

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Usrednjene vrijednosti udarnih presjeka za reakcije $^{115}\text{In}(n, n')^{115m}\text{In}$, $^{115}\text{In}(n, \gamma)^{116m}\text{In}$, $^{113}\text{In}(n, n')^{113m}\text{In}$, $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$, $^{111}\text{Cd}(n, n')^{111m}\text{Cd} + ^{110}\text{Cd}(n, \gamma)^{111m}\text{Cd}$, $^{27}\text{Al}(n, p)^{27}\text{Mg}$, $^{64}\text{Zn}(n, p)^{64}\text{Zn}$, $^{68}\text{Zn}(n, \gamma)^{69m}\text{Zn}$, $^{138}\text{Ba}(n, \gamma)^{139}\text{Ba}$, $^{135}\text{Ba}(n, n')^{135m}\text{Ba} + ^{134}\text{Ba}(n, \gamma)^{135m}\text{Ba}$, $^{58}\text{Ni}(p, n)^{58}\text{Co}$ i $^{87}\text{Sr}(n, n')^{87m}\text{Sr} + ^{86}\text{Sr}(n, \gamma)^{87m}\text{Sr}$ izmjerene su u fizijskom spektru neutrona jednog malog izvora ^{252}Cf . Rezultati su općenito uzevši u slaganju s podacima iz literature. U slučajevima gdje su diferencijalni udarni presjeci u ovisnosti o energiji bili dostupni, usrednjene vrijednosti udarnih presjeka bile su izračunate pretpostavljajući neutronske energijski spektar Maxwellovog oblika s parametrom $T = 1,42$ MeV što odgovara srednjoj energiji spektra od 2,13 MeV. Usporedba izmjerenih i izračunatih vrijednosti pokazuje velika neslaganja za slučajeve reakcije $^{56}\text{Fe}(n, p)$, $^{138}\text{Ba}(n, \gamma)$ te $^{135}\text{Ba}(n, n') + ^{134}\text{Ba}(n, \gamma)$.