# THERMAL FISSION CROSS SECTION MEASUREMENTS OF $^{243}$CM AND $^{245}$CM

L. POPESCU, J. HEYSE, J. WAGEMANS,

*SCK•CEN, Boeretang 200, B-2400 Mol, Belgium*

C. WAGEMANS

*University of Ghent, Proeftuinstraat 86, B-9000 Ghent, Belgium*

A new measurement program was set up at SCK•CEN to determine the thermal neutron-induced fission cross section of a number of Cm isotopes. The experiments are performed at a thermal neutron beam from the graphite moderated reactor BR1 at SCK•CEN. This paper presents preliminary results of our $^{243}$Cm(n,f) and $^{245}$Cm(n,f) cross-section measurements.

## 1. Introduction

Neutron-induced fission cross-section data are of interest for basic nuclear physics and astrophysics studies and are a crucial input in reactor physics calculations. Spent nuclear fuel contains important quantities of Cm isotopes. Hence Cm(n,f) cross-section data are needed for nuclear waste transmutation calculations. They play an important role in the core design of Accelerator Driven Systems and Generation IV reactors.

The currently available evaluated databases show important gaps and discrepancies. Some of these evaluations are based on old measurements using poorly characterized samples with low enrichment. Therefore, new measurements are required to improve the evaluated nuclear data libraries.

In the framework of the Interuniversity Attraction Poles project financed by the Belgian Science Policy Office of the Belgian State, a measurement program was set up at SCK•CEN to determine the thermal neutron-induced fission cross section of a number of Cm isotopes.

## 2. Experimental setup

The measurements were performed at the Z59 irradiation channel of the graphite moderated BR1 reactor [1]. The neutron beam has been collimated to a diameter of 20 mm by a system of three boron carbide collimators. The samples and the detectors have been installed in a vacuum chamber, with the samples positioned perpendicular to the beam, at 60 cm from the reactor wall. At the sample position the thermal neutron flux was of the order of $6{\cdot}10^5$ $s^{-1}cm^{-2}$.

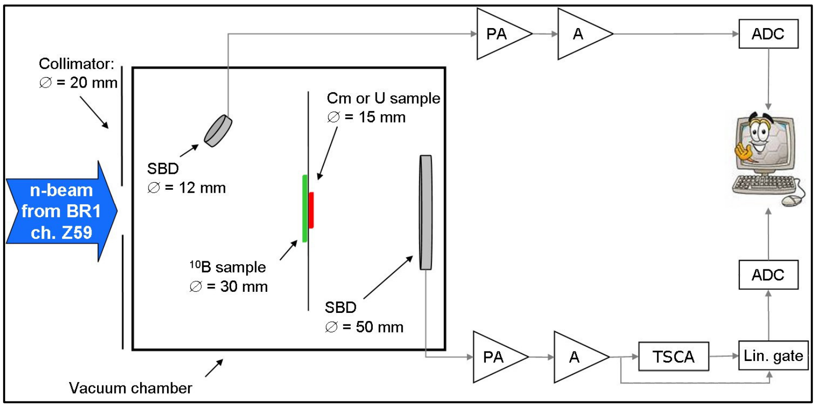


Figure 1. Schematic view of the detection system. The samples and the two surface barrier detectors (SBD) have been mounted in a vacuum chamber. The data acquisition system is sketched on the right-hand side of the picture.

The Cm(n,f) measurements have been performed relative to $^{235}$U(n,f). Two samples have been mounted in the center of a vacuum chamber. They were positioned back-to-back on a support plate perpendicular to the beam (see Fig. 1). One of these samples was either $^{235}$U, or the fissile material under investigation; the other was a $^{10}$B sample, used for monitoring the neutron flux. The $^{10}$B nuclei in the monitoring sample are subject to (n,$\alpha$) reactions with a high cross section; therefore, the sample ensures an accurate determination of the neutron flux variations.

The detection of the emitted alphas and fission fragments was done via surface barrier detectors (SBD) positioned as shown in Fig. 1. The collected signals were pre-amplified, amplified, passed through an ADC module and then collected by a mixing unit before being sent to the acquisition PC. A linear gate was inserted after the amplifier in the Cm chain, to cut most of the pile-up signals caused by $\alpha$ decay of nuclei in the Cm sample.

## 3. $^{243}$Cm(n,f) measurement and results

A Cm-oxide sample containing (2.320±0.025) μg of $^{243}$Cm was used for the $^{243}$Cm(n,f) measurement. The material was deposited on a 30 μm thick Al backing. The sample diameter was 15 mm and its isotopic composition at the moment of the experiment (April 2010) is given in Table 1.

Table 1. Isotopic composition of the $^{243}$Cm sample.

| Isotope | Number of Atoms | At.% |
|---|---|---|
| $^{243}$Cm | $5.70 \times 10^{15}$ | 92,503 |
| $^{244}$Cm | $2.57 \times 10^{14}$ | 4,173 |
| $^{245}$Cm | $4.01 \times 10^{12}$ | 0,065 |
| $^{246}$Cm | $5.72 \times 10^{13}$ | 0,928 |
| $^{247}$Cm | $1.34 \times 10^{12}$ | 0,022 |
| $^{248}$Cm | $1.70 \times 10^{12}$ | 0,028 |
| $^{239}$Pu | $1.41 \times 10^{14}$ | 2,282 |

The measured $^{243}$Cm(n,f) pulse height spectrum is plotted in Fig. 2. As the sample is a strong α-emitter, the pile-up of several α-signals can produce a signal with amplitude close to the one of the fission fragments. This undesirable background (represented by the large peak at the left side of the spectrum) could be well separated from the $^{243}$Cm(n,f) region.

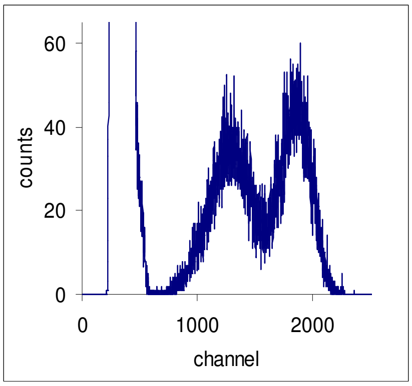


Figure 2. Pulse height spectrum measured with the $^{243}$Cm sample. The signals produced by the fission fragments could be well separated from the α pile-up (the large peak on the left side of the figure).

As the even-A isotopes in the sample decay also via spontaneous fission (SF), we determined the SF contributions to the $^{243}$Cm(n,f) spectrum by closing the neutron beam and repeating the measurement. The result indicates almost 16% contributions from SF to the spectrum shown in Fig. 2. This spectrum contains also a 9.5% contribution from the epithermal component of the neutron beam, which was determined by repeating the $^{243}$Cm(n,f) measurements with a

Cd filter (1 mm thick) positioned perpendicular to the beam, in front of the vacuum chamber.

For determining the neutron flux and the detection efficiency, a $^{235}U(n,f)$ measurement was performed keeping the same configuration as for $^{243}Cm$. A 99.94% enriched $^{235}U$ sample, with a diameter of 15 mm and a mass of (193.29±1.33) μg was used. This sample was deposited on a 30 μm thick Al backing. For the $^{235}U(n,f)$ cross section we used the $\sigma_f(E_n = 25.3\ \text{meV}) = 582.60$ b value as given in Ref. [2] .

The ratio of the counting rates for the $^{10}B(n,\alpha)$ measurement performed in parallel with the $^{235}U(n,f)$ and $^{243}Cm(n,f)$ measurements allowed an accurate normalization of the beam flux. In the present analysis, the Westcott $g_f$-factors, at $E_n = 25.3$ meV, $g_f(^{235}U) = (0.9771 \pm 0.0008)$ and $g_f\ (^{243}Cm) = 1.0054$ as given in Ref. [2] have been used to correct for the deviation from the 1/v shape of these fission cross sections.

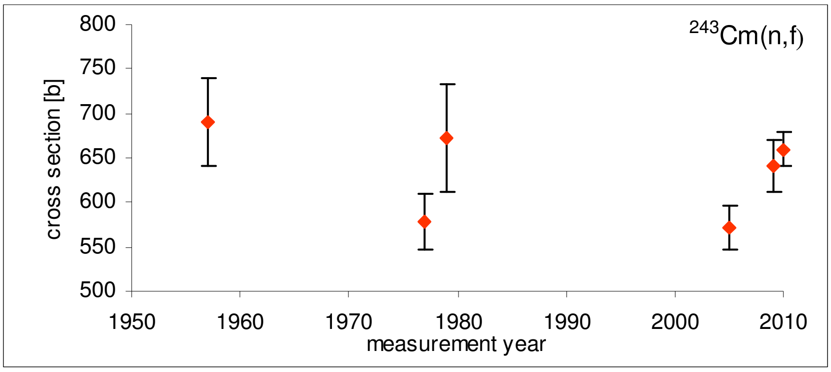


Figure 3. Comparison of the present result (2010) with literature data for the $^{243}Cm(n,f)$ cross section at thermal energy [3-7].

Figure 3 compares our preliminary value (660±18) b with different $^{243}Cm(n,f)$ thermal cross section values presented in literature [3-7]. Some of these authors did not consider the Westcott factor when extrapolating the cross section to $E_n = 25.3$ meV [3,6]. Moreover, we did not renormalize the old values using the latest cross sections for the calibration measurement (e.g. $^{235}U(n,f)$).

The thermal fission cross section adopted in the European (JEFF-3.1.1), Japanese (JENDL-AC-2008) and American (ENDF/B-VII.0) neutron libraries are 617.4 b, 587.36 b and 613.32 b, respectively. Our result overlaps within the error bars with the results of Hulet [3], Zhuravlev [5] and Alekseev [7] and it shows a deviation of 7% from the value adopted by JEFF-3.1.1 and 12% from the value adopted by JENDL nuclear data library.

The statistical uncertainty of the present result is less than 1%, while the systematic uncertainty is almost 2%. The main systematic uncertainty is induced by the uncertainty on the mass of the sample.

## 4. $^{245}$Cm(n,f) measurement and results

For the $^{245}$Cm(n,f) measurement, we used a Cm-oxide sample containing (141±3) μg of $^{245}$Cm, which was deposited on a 30 μm thick Al backing. The sample diameter was 15 mm and its isotopic composition at the date of the experiment (November 2008) is given in Table 2.

Table 2. Isotopic composition of the $^{245}$Cm sample.

| Isotope | Number of Atoms | At.% |
|---|---|---|
| $^{244}$Cm | $3.16\text{x}10^{15}$ | 0.898 |
| $^{245}$Cm | $3.48\text{x}10^{17}$ | 98.687 |
| $^{246}$Cm | $1.43\text{x}10^{15}$ | 0.406 |
| $^{247}$Cm | $2.82\text{x}10^{13}$ | 0.008 |
| $^{248}$Cm | $7.06\text{x}10^{12}$ | 0.002 |

The measured $^{245}$Cm(n,f) pulse height spectrum is plotted in Fig. 4. Similarly to the case of $^{243}$Cm, the sample is a strong α-emitter, the pile-up of several α-signals producing a signal with amplitude close to the one of the fission fragments. This background could be well separated from the $^{245}$Cm(n,f) region.

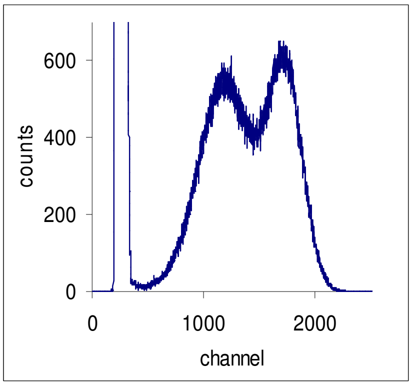


Figure 4. Pulse height spectrum measured with the $^{245}$Cm sample. The signals produced by the fission fragments could be well separated from the α pile-up (the large peak on the left side of the figure).

The SF contributions and the contributions from the epithermal component of the neutron beam to the $^{245}$Cm(n,f) spectrum shown in Fig. 4 were determined in the same way as for the case of $^{243}$Cm(n,f) measurement. The SF contributions were of the order of 1.6%, while the contributions from the epithermal component were of the order of 2.6%.

A value of $g_f$ ($^{245}$Cm) = (0.954 ± 0.033) [2] has been used in the data analysis to correct for the deviation from the 1/v shape of the fission cross section. The large uncertainty in the Westcott $g_f$-factor for $^{245}$Cm induces a comparatively large systematic uncertainty in the obtained $^{245}$Cm(n,f) thermal cross section.

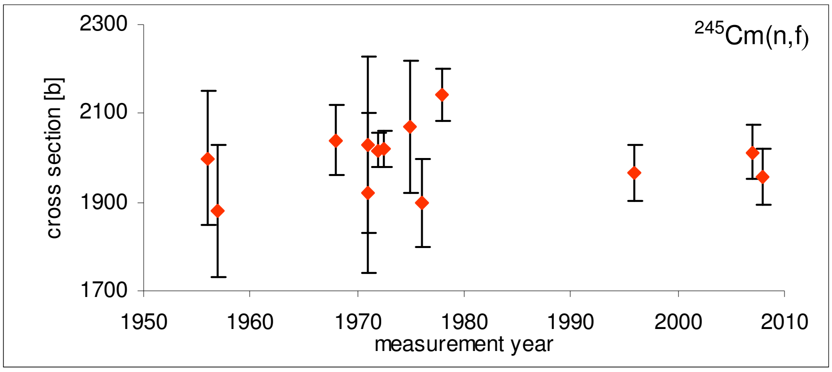


Figure 5. Comparison of the present result (2008) with literature data for the $^{245}$Cm(n,f) cross section at thermal energy [8-18].

Figure 5 compares our preliminary value (1927±86) b with different $^{245}$Cm(n,f) thermal cross section values presented in literature [8-18]. Most of these values are not including the $g_f$-factor when extrapolating the cross section to $E_n$ = 25.3 meV and their uncertainties are underestimated. Similarly to the case of $^{243}$Cm, the old values from the plot shown in Fig. 5 have not been renormalized by using the latest cross sections for the calibration measurements.

The thermal values adopted in the nuclear data libraries are 2054.1 b (JENDL-AC-2008) and 2142.4 b (ENDF/B-VII.0 and JEFF-3.1.1). However, apart from the result of Browne et al. [16], all other experimental data point to a smaller cross section.

Our result overlaps within the error bars with the previous results and it shows a deviation of about 10% from the value adopted by the ENDF/B-VII.0 and JEFF-3.1.1 nuclear data libraries and 6.2% from the value adopted by JENDL-AC-2008.

The statistical uncertainty of the present result is of the order of 0.4% while the systematic uncertainty is 4.1%. As already mentioned, the main systematic uncertainty is induced by the large uncertainty on the $g_f$-factor for $^{245}$Cm. Recent measurements at the GELINA facility of IRMM, Geel, lead to a more accurate value: $g_f$ ($^{245}$Cm) = (0.939±0.019) [19]. This new result leads to a value of (1957±65) b for the thermal cross section of $^{245}$Cm(n,f), which has a better overlap with previous results. It shows a deviation of 8.6% from the value adopted by the ENDF/B-VII.0 and JEFF-3.1.1 nuclear data libraries and 4.7% from the value adopted by JENDL-AC-2008.

## 5. Conclusions

The $^{243}$Cm(n,f) and $^{245}$Cm(n,f) thermal cross sections at 25.3 meV have been measured at the BR1 reactor of SCK•CEN. Our preliminary analysis indicate a thermal cross-section value of (660±18) b for $^{243}$Cm and (1927±86) b for $^{245}$Cm. Although these preliminary results deviate up to 12% from the values adopted by different nuclear data libraries, they overlap within error-bars with most of the previous experimental values [3-18]. A better agreement is obtained for $^{245}$Cm, when using the recent $g_f$ value of Serot et al.[19], which leads to a thermal cross-section value of (1957±65) b.

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