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Measurement of the ^{236}U(n,f) cross section as a function of the neutron energy

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Abstract. The ^{236}U(n,f) cross section has been measured in the neutron energy region from 0.5 eV to 25 keV at the GELINA neutron facility of the IRMM in Geel, Belgium. A highly enriched ^{236}U sample was mounted back-to-back with a ^{10}B sample in the centre of a Frisch-gridded ionisation chamber; a control measurement was performed with a ^{235}U sample in the same configuration. Besides a dominant resonance at 5.45 eV, for which a resonance analysis was performed, the next resonance (cluster) only occurs at about 1.3 keV. It is demonstrated that the fission resonance integral and the thermal fission cross section adopted in all commonly used evaluated data libraries are too large by two orders of magnitude.

1 Introduction

The experimental data base for the ^{236}U(n,f) cross section in the thermal and resonance energy region is very poor: with thermal neutrons, only one direct measurement is available [1], whilst with resonance neutrons only one σ_f(E) data set has been released [2] which has however been contradicted by more recent measurements [3,4]. Moreover, the older measurements [2,3] are hampered by a strong ^{235}U(n,f) background, due to a non-negligible amount of ^{235}U in the sample. In view of this situation, a new measurement in the neutron resonance region was performed at the GELINA neutron facility of the Institute for Reference Materials and Measurements in Geel (Belgium), using a highly enriched ^{236}U sample.

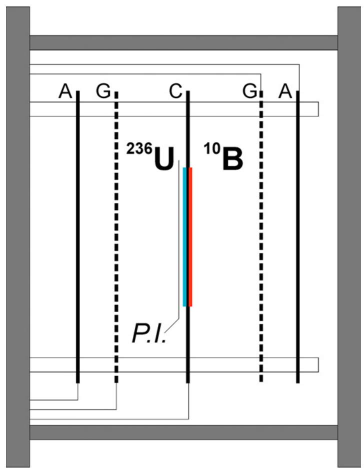

Fig. 1. Details of the double Frisch-gridded ionisation chamber.

2 Measurements

2.1 Experimental conditions

The measurements were performed at an 8.3 m long flight path, GELINA being operated at a repetition frequency of 800 Hz and an electron burst width of 1 ns. A double Frisch-gridded ionisation chamber was used with ultra-pure methane as detector gas (fig. 1). The uranium sample (covered with a very thin poly-imide foil to avoid contamination of the chamber) was mounted in the centre of this chamber, back-to-back with a ^{10}B sample. Two consecutive measurements were performed, one with a ^{236}U sample, a second one with a ^{235}U sample. The sample characteristics are given in table 1.

In all cases, a permanent cadmium neutron filter was placed in the beam to remove overlap neutrons. The background was determined using black resonance neutron filters. The α-pile-up due to the radioactive decay of the uranium isotopes was checked in measurements without neutron beam.

Table 1. Sample characteristics.

Sample	Thickness μg/cm^2	Isotopic composition (at.%) ^{233}U	^{234}U	^{235}U	^{236}U	^{238}U
^{10}B	8.05 ± 0.10					
^{235}U	110.9 ± 0.7	0.0056	0.0408	99.040	0.2632	0.6502
^{236}U	209.9 ± 1.3			0.0043	99.9732	0.0225

For the U(n,f) and ^{10}B(n,α)^{7}Li reactions, coincident pulse-height and time-of-flight (TOF) events were recorded with a Labview-based data acquisition system.

2.2 Results

The data recording method used permits a good selection of the fission fragments and the ^{10}B(n,α) particles, as illustrated in figure 2. The perturbing ^{235}U(n,f) background in the ^{236}U(n,f) TOF measurement is strongly reduced, but still needs to be corrected for, as illustrated in figure 3. Thanks to the measurement with the ^{235}U sample, this can easily be done.

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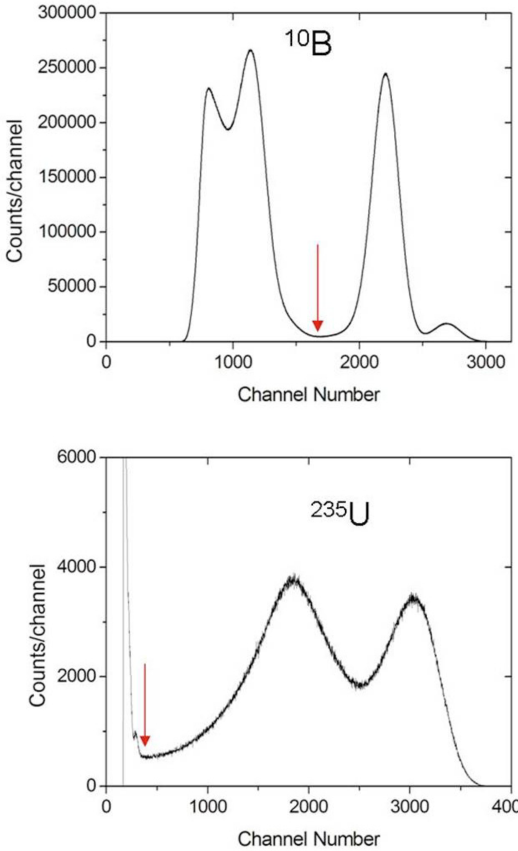

Fig. 2. Pulse-height spectrum of the ^{10}B(n,α) particles (top) and of the ^{235}U(n,f) fragments (bottom). The (software) discriminator setting is indicated with an arrow.

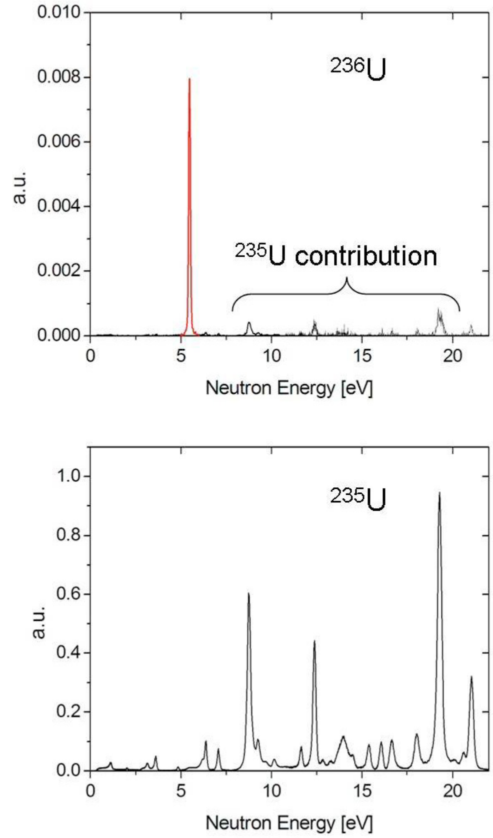

Fig. 3. Raw ^{236}U(n,f) data uncorrected for the ^{235}U(n,f) contribution yielding the small peaks (top); the corresponding ^{235}U(n,f) spectrum is shown below.

The time-dependent background in the U(n,f) and ^{10}B(n,α)^{7}Li measurements has been determined as a function of the time-of-flight t by fitting a function $at^b + c$ through the counting rates in the black resonances. After transformation into energy, this background $Y^{BGR}(E_n)$ has been subtracted from the raw fission and ^{10}B(n,α) counting rates $Y_f(E_n)$ and $Y_B(E_n)$ calculated from the experimental data. Finally, the neutron induced fission cross section of ^{236}U can be calculated as follows (see J. Heyse et al. [5] for details):

$$\sigma_{6u}(E_n) = \frac{N_B}{N_{6u}} \sigma_B^{ENDF}(E_n) \frac{Y_f(E_n) - Y_f^{BGR}(E_n)}{Y_B(E_n) - Y_B^{BGR}(E_n)} - \frac{N_{5u}}{N_{6u}} \sigma_{5u}(E_n).$$

Here N stands for the number of atoms in the B or U sample considered; for the ^{10}B(n,α)^{7}Li reaction cross section $\sigma_B(E_n)$ the ENDFB-VII.0 evaluated data have been adopted. All these operations are implemented in the computer code AGS [6], which takes into account all sources of uncertainty and takes care of the error propagation, producing an experimental variance-covariance matrix.

Figure 4 shows the ^{236}U(n,f) cross section obtained in the neutron energy region from 0.5 eV to 25 keV. Besides the dominant resonance at 5.45 eV, four resonance clusters with a spacing of 2–3 keV can be observed, as reported by Parker et al. [3]. Due to the short flight path used in our measurements, the resonances inside the clusters are not very well resolved.

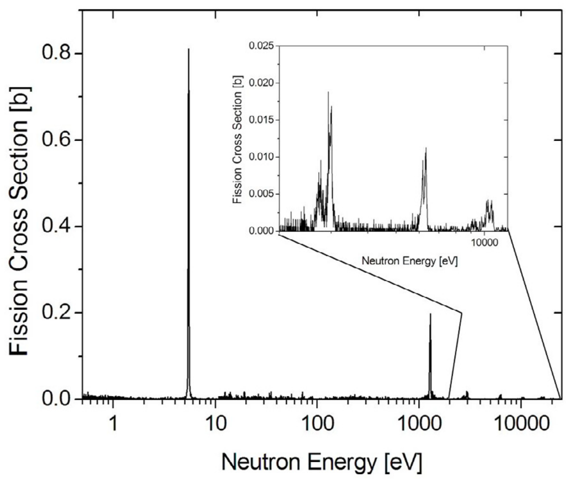

Fig. 4. ^{236}U(n,f) cross section from 0.5 eV to 25 keV.

3 Discussion

3.1 Intermediate resonance structure

The ^{236}U(n,f) cross section results given in figure 4 do not show the large number of resonances reported by Theobald

Table 2. Experimental and evaluated values of the fission resonance integral and the thermal fission cross section.

	This work	Wagemans [1]	JEFF3.1	JENDL3.3	ENDFB-VII.0
$I_f(25)$ [b]	0.032		4.334	4.359	4.452
$I_f(100)$ [b]			4.336	4.363	4.460
σ_f(th) [mb]	0.22	0.3 ± 1	61.3	61.3	47.1
	BW extrapol.	experiment			

et al. [2]. These resonances are probably due to a small sensitivity for radiative capture events of the liquid scintillator used in their measurements. The present picture of fairly distant resonance clusters is a clear signature of intermediate structure, which is not surprising seen the sub-barrier character of the neutron induced fission of ^{236}U.

3.2 The 5.45 eV resonance and the thermal region

Figure 5 shows the result of a fit to the cross section data in the region of the 5.45 eV resonance, using the R-matrix code SAMMY [7]. With Γ_γ = 24.5 meV and Γ_n = 2.16 meV (taken from JEFF3.1), a fission width Γ_f = 1.71 μeV was obtained. This result is quite comparable with the value of (1.3 ± 0.1) μeV reported by Parker et al. [2], but two orders of magnitude lower than the Γ_f = 290 μeV adopted in JEFF3.1. A Breit-Wigner extrapolation of the 5.45 eV resonance tail towards 0.0253 eV using the resonance parameters obtained in the present work yields a thermal contribution of 0.22 mb (fig. 6), in agreement with the experimental value (0.3 ± 1) mb of Wagemans et al. [1]. To the thermal contribution of the 5.45 eV resonance, a small contribution of a bound state could possibly be added. The lower curve in figure 6 is a result of the present measurements, which yield σ_f(E) values about two orders of magnitude smaller than the corresponding JEFF3.1 values (the same is true for JENDL3.3 and ENDFB-VII.0).

3.3 The fission resonance integral

In table 2 the values for the fission resonance integral

$$I_f = \int_{0.5eV}^{100keV} \sigma_f(E) \frac{dE}{E}$$

denoted by $I_f(100)$ in the table – calculated from the JEFF3.1, JENDL3.3 and ENDFB-VII.0 evaluated data libraries are reported. Since the ^{236}U(n,f) cross section data obtained in the present measurements only cover the neutron energy region from 0.5 eV to 25 keV, also the integral $I_f(25)$ being

$$I_f = \int_{0.5eV}^{25keV} \sigma_f(E) \frac{dE}{E}$$

has been calculated, which is almost equal to $I_f(100)$. The comparison clearly demonstrates that the fission resonance integral calculated from all evaluated data libraries is too large by two orders of magnitude.

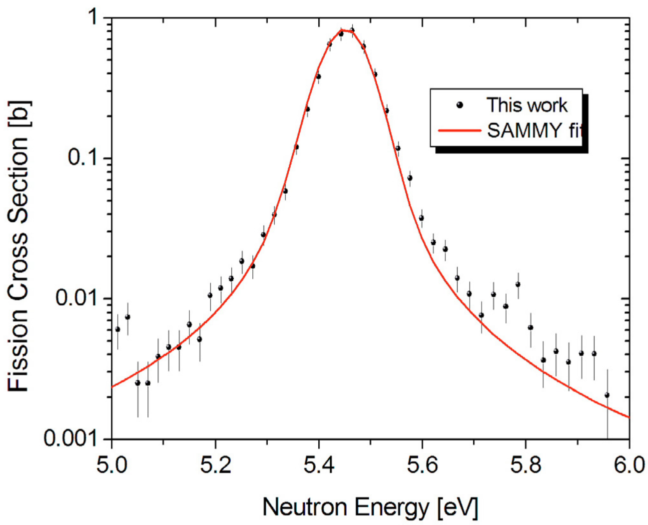

Fig. 5. Sammy fit to the 5.45 eV resonance data points.

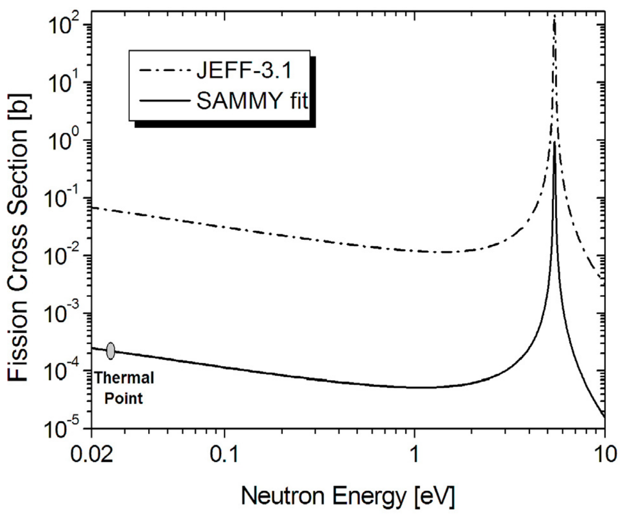

Fig. 6. Comparison of the JEFF3.1 evaluation below 10 eV with the present results extrapolated to the thermal energy region.

In the same table 2, also the thermal neutron induced fission cross sections adopted in the evaluations are reported and compared with the experimental value of Wagemans et al. [1] and with the value deduced from a Breit-Wigner extrapolation of the 5.45 eV resonance. Also here all evaluated data files are too large by two orders of magnitude.

These comparisons clearly demonstrate that all three evaluated data libraries need to be corrected, since all of them are still relying on the wrong fission cross section values of Theobald et al. [2].

3.4 Impact on the isotopic inventory of spent fuel

The MALIBU international program investigates the isotopic inventory of spent fuel [8]. Experimental data for the actinide composition are compared with calculated values based on the JEFF3.1 evaluated data library. The experimental /calculated actinide content is slightly improved for several isotopes after changing the ^{236}U(n,f) cross section data [9].

4 Conclusion

New experimental data for the ^{236}U(n,f) cross section in the resonance energy region are reported, in which the signature of intermediate structure is clearly present. These new data are consistent with the experimental value for the thermal ^{236}U(n,f) cross section obtained at the high flux reactor of the ILL [1]. Both the results in the thermal and resonance region are two orders of magnitude lower than the JEFF3.1, JENDL3.3 and ENDFB-VII.0 evaluated data libraries, which should be revised.

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