International Evaluation Co-operation

Volume 5

PLUTONIUM-239 FISSION CROSS-SECTION BETWEEN 1 AND 100 keV

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PLUTONIUM-239 FISSION CROSS-SECTION BETWEEN 1 AND 100 keV

A report by the Working Party
on International Evaluation Co-operation
of the NEA Nuclear Science Committee

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FOREWORD

A Working Party on International Evaluation Co-operation was established under the sponsorship of the OECD/NEA Nuclear Science Committee (NSC) to promote the exchange of information on nuclear data evaluations, validation, and related topics. Its aim is also to provide a framework for co-operative activities between members of the major nuclear data evaluation projects. This includes the possible exchange of scientists in order to encourage co-operation. Requirements for experimental data resulting from this activity are compiled. The Working Party determines common criteria for evaluated nuclear data files with a view to assessing and improving the quality and completeness of evaluated data.

The Parties to the project are: ENDF (United States), JEF/EFF (NEA Data Bank Member countries), and JENDL (Japan). Co-operation with evaluation projects of non-OECD countries are organised through the Nuclear Data Section of the International Atomic Energy Agency (IAEA).

The following report was issued by a Subgroup investigating the fission cross-section of Plutonium-239 in the energy range 1 to 100 keV. This cross section is of particular importance for fast reactor applications, such as k_{eff} , sodium void reactivity coefficient and control rod worth. An analysis of recent experimental data by L. Weston et.al. give significantly lower cross-section values that the simultaneous evaluation performed by W. Poenitz for the ENDF/B-VI library. The objective of the subgroup was to resolve this discrepancy.

The opinions expressed in this report are those of the authors only and do not represent the position of any Member country or international organisation. This report is published on the responsibility of the Secretary-General of the OECD.

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SUMMARY

In order to understand the discrepancies between Weston's data – *NSE 88, 567, 1984* – and the major file evaluations, one experimental program and one evaluation one have been agreed upon:

The experimental program which essentially aims at normalisation checking has been performed in GEEL and Oak Ridge. It supports an upward re-normalisation by ~3.1%. The evaluation program has not been completed and even, as a consequence of the experimental results, loses a part of its justification. But some acquired results are important and can be used for future ^{239}Pu evaluations. The JEF-2.2 benchmarking performed in another context supports the results of the experimental program although it suggests a slightly larger re-normalisation.

PLUTONIUM-239 FISSION CROSS-SECTION BETWEEN 1 AND 100 keV

1. Introduction

The fission cross-section measured by Weston – *NSE 88, 567, 1984* – in excellent resolution conditions is about 5% lower than almost all recent measurements and all major evaluations in the range 1–100 keV. Expressed in terms of critical mass, control rod worth, and void coefficient, this 5%-difference is of importance for fast reactor calculations.

In order to solve this problem Subgroup.5 of the NEA/NSC Working Party on International Evaluation Co-operation adopted the following two-part plan:

1. *Critical examination of Weston's experiment with possibly additional experiments to check questionable points*;

2. *Examination of other sources of information relating to fission cross-section*.

2. Experimental work

2.1. Critical examination of Weston's and Todd's experiment

This is a T.O.F. experiment performed at ORELA using a multiparallel plate fission chamber; the shape of the neutron flux was measured relative to a $^{10}BF_3$ chamber up to a neutron energy of 1 keV, and relative to a ^{6}Li glass scintillator at higher energies. The flux inter-normalisation was made in the energy interval between 100 and 1000 eV with a statistical uncertainty of less than 0.1%. Furthermore the σ_f-curve was normalised at thermal energy; this method of normalisation is unsuitable for experiments aimed at obtaining data at high energy, because of sample thickness self-absorption, and dead-time effects. The energy range of flux intercomparison in Weston's experiment has been chosen to intercompare the various sets of data through fission integral values:

$$I_f = \int_{100\mathrm{eV}}^{1000\mathrm{eV}} \sigma_f(E)\,dE$$

For the Weston's 84 data the fission integral is $I_f = 8996\ b{\bullet}eV$, which is known to have rather significant uncertainties: 1.9% for normalisation, 1% systematic, 0.15% statistical. When considering Gwin's measurements performed in similar conditions, we obtain $I_f = 9268\ b{\bullet}eV$ [1] and $I_f = 9286\ b{\bullet}eV$ [2]. The 4%-difference suggested the possibility of a normalisation error in Weston's experiment. But since Gwin's experiments are affected with a large uncertainty (11%) in the ^{10}B-content of the neutron flux counter, they have not been considered as absolutely reliable references.

In addition when looking at Figure 1, and considering the Poenitz standard [16] as a good average representation of major evaluations, we observe a difference of ~3% between 0.1 and 10 keV and ~5% between 10 and 100 keV. This situation of energy-dependent discrepancy raises the questions of a normalisation error and of a possible energy-dependent effect in Weston's experiment.

Therefore it has been decided to plan experimental programs both at Oak Ridge and Geel in order to check this normalisation point.

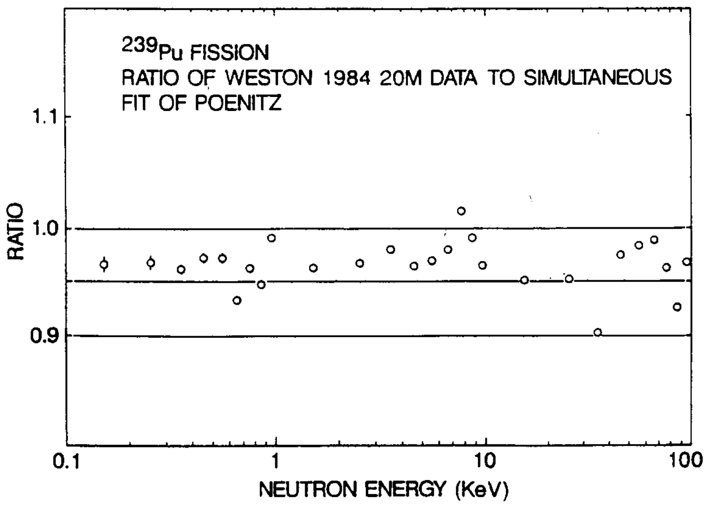

Figure 1 ***Ratio of Weston's data (1984) to simultaneous fit of Poenitz***

2.2. Experimental work performed within the framework of Subgroup 5

2.2.1. Geel experiment

In Geel (1992) two different geometries have been used by C. Wagemans [3]:

1. *Measurement with double ionisation chamber in a '2-π geometry'.* The characteristics were as follows:

   - ^{239}Pu sample: 186 μg/cm^2
   - ^{10}B sample: 10 μg/cm^2
   - Flight path length: 8.5 cm

     The normalisation has been made in two ways, all based on the thermal value of the ^{239}Pu fission cross-section.

     i. *via the fission integral between 20 and 60 meV,*

     $\int_{20\,\mathrm{meV}}^{60\,\mathrm{meV}} \sigma_f \, dE = 25.36\,\mathrm{b} \bullet \mathrm{eV}$, from which an average value of 634 b is obtained and is to be compared with 631.4 b, which is the average of several measurements selected by H. Derrien from his most recent resonance parameter evaluation [4];

     ii. *via a linear least square fit of the* $\sigma\sqrt{E}$ *data between 20 and 30 meV*, from which a value of 784.25 b was derived for the thermal cross-section.

   Within these conditions of normalisation Wagemans derived $I_f = 9190 \pm 110$ *b•eV*. The 1.2%-uncertainty results from 0.2% of counting statistics, 0.5% of uncertainty on the background correction, and 0.6% due to the normalisation.

2. *Measurement in 'low geometry' using surface barrier detectors*, with:

   - ^{239}Pu sample: 106 μg/cm^2
   - ^{10}B sample: 10 μg/cm^2
   - Neutron flight path length: 8.3 cm

   For this measurement the fission integral value is *(9450±200) b•eV*. The components of the total uncertainty are: 0.8%, 0.5% and 1%

respectively, due to counting statistics, background and normalisation corrections.

Considering both data, a weighted average value is obtained for I_f:

$$^{Wa}I_f = 9250 \pm 96 \text{ b} \bullet \text{eV}.$$

2.2.2. Oak Ridge experiment

In Oak Ridge a new fission cross-section measurement was performed by Weston et al. [9]. The experimental technique was similar as previously but the detectors were different: A parallel plate ^{10}B ionisation chamber with solid coating of Boron on the centre plate was used rather than the $^{10}BF_3$ gas. The fission chamber was also different as the active plates were loaded with ^{239}Pu only, so that better counting statistics is obtained for the same irradiation time. The flight path length was 19 m, and the measurement was continuous from below 0.025 eV up to 10 keV. Careful attention was paid to the normalisation which was more precise than in the previous measurement *–0.5% against 1.9%* [8] –, while the overall systematic uncertainty was estimated to be 0.6% *– to be compared with 1%*. The normalisation was done on the integral $\sigma_f = \sqrt{E}$ over the interval 0.02 to 0.03 eV referring to the ENDF/B-VI evaluation:

$$\int_{0.02\,\text{eV}}^{0.03\,\text{eV}} \sigma_f \sqrt{E}\, dE = 118.98\, \text{b} \bullet \text{eV}^{3/2}$$

The I_f value obtained in these conditions is:

$$^{We}I_f = 9302 \pm 102 \text{ b} \bullet \text{eV}$$

The total uncertainty of this value is estimated from the just above-mentioned values on normalisation and systematic uncertainty, including a 0.1%-component due to statistical uncertainty.

This normalisation operation is consistent with the set of normalisation operations used by Wagemans et al.

Although obtained in somewhat different conditions of detection, Wagemans' and Weston's I_f values based on thermal fission data are in excellent agreement. All these values have been obtained from data normalised on the ENDF/B-VI thermal value of the ^{239}Pu fission cross-section as a primary standard, and adapting the ENDF/B-VI values for the ^{10}B(n,α) ^{7}Li cross-section.

The final I_f value which results from the experimental work on normalisation is obtained from a weighted average of Wagemans' and Weston's data, which are shown in Table 1.

Table 1 ***Wagemans' and Weston's data***

WAGEMANS$_1$ [92]	WAGEMANS$_2$ [92]	WESTON [92]
9190±110	9450±200	9302±102
STANDARD I_f = 9275±85 b•eV		

This average value which results from particularly careful measurements should be recommended as a secondary standard in addition to the thermal value, to be used in the case of two-step measurements or any measurement subject to energy-dependent effects. In the following, we will refer to it as the Standard I_f.

The <u>*N*</u>ormalisation <u>*F*</u>actor to be applied to Weston's data (1984) is derived as:

$$NF = \frac{9275}{8996} = 1.031 \pm 0.009$$

The I_f values obtained from the few measurements performed in the thermal range up to 1000 eV (see Table 2), are consistent with the Standard I_f, since a mean value of 9278 b•eV is derived, affected with a 61 b•eV standard deviation.

Table 2 ***I_f value in b•eV related to experimental data***

GWIN [71]	GWIN [76]	WAGEMANS [80]
9268	9286	9280 *

* This re-calculated using the ENDF/B-VI values for the ^{10}B(n,α) ^{7}Li cross-section.

Note that all above-mentioned fission integrals are normalised on σ_f = 748.0 b at thermal energy.

3. Examination of other sources of information

Below we describe the work performed within the framework of Subgroup 5 in order to explore other sources of information than the experimental one. It brings an indirect support to the normalisation and shows that many of the major evaluations could be improved, although they perform satisfactory in several applications. The following provides data, information or tools which should be used for improving these evaluations over large energy ranges.

3.1. Japanese evaluation work

A specific study, made in Japan (Nakagawa, Nikai,...), has shown that other types of data such as α data or competitive reaction data like capture, non elastic, inelastic,.etc., are too inaccurate to be used as references.

In the resolved range, H. Derrien in CEA-France and Nakagawa in JAERI-Japan have produced a new resonance parameter set extended up to 2.5 keV.

This evaluation is a simultaneous fit [4] of various experimental data sets according to a Bayesian method using the SAMMY code allowing background and normalisation coefficient adjustment. The following experimental data base was used:

- Absorption and fission data from Gwin et al. [1,2],
- Fission data from Gwin et al. [5,6], Blons [7], Weston and Todd [8,9];
- Transmission data from Spencer [10], Harvey [11].

These experimental data sets are characterised by high-energy resolution and/or low background. With respect to the data sets from which previous resolved resonance parameters – included in JEF-2.2 and ENDF/B-VI – have been derived, additional sets are included, in particular the high-resolution transmission data by Harvey et al. [11] and the high-resolution cross-section measurement by Weston and Todd (1988) [14]. If we use this Japanese evaluation, we obtain for the range 100-1000 eV a fission integral equal to

9304 b•eV, which is to be compared with the I_f values obtained from major evaluations as quoted in Table 3.

Table 3 ***I_f value in b•eV related to evaluated data***

ENDF/B-VI	JEF-2.2	JENDL-3	POENITZ *standard*	DERRIEN [92]
9017	9040	9040	9377	9304

The I_f values have been calculated from averaged fission cross-sections according to the energy scheme given in Table 4 below.

Table 4 ***Averaged fission cross-section***

ENERGY *eV*	DERRIEN NAKAGAWA *1992*	JENDL-3	JEF-2	WESTON *1984 -renorm.*	POENITZ *standard*
0.010-10	80.12		80.25		
9-20	94.74	91.87	91.87		
20-40	17.52	16.98	17.09	17.97	
40-60	50.64	49.1	49.16	50.87	
60-100	54.42	53.23	52.94	54.33	
100-200	18.63	18.14	18.14	18.56	18.66
200-300	17.85	13.31	17.32	17.89	17.88
300-400	8.31	8.083	8.083	8.34	8.43
400-500	9.59	9.391	9.391	9.58	9.57
200-500	11.92	11.59	11.60	11.93	11.96
500-600	15.39	15.06	15.06	15.57	15.86
600-700	4.37	4.131	4.131	4.30	4.46
700-800	5.51	5.324	5.324	5.53	5.63
800-900	4.84	4.730	4.730	4.89	4.98
900-1000	8.33	8.230	8.230	8.38	8.30
500-1000	7.69	7.496	7.496	7.73	7.79

3.2. *Theoretical and validation work*

To start with, it was decided to perform two model calculations in order to describe the fission data, the first one based on Weston's '84 ❶, and the second on ENDF/B-VI ❷. Both would be based on the same neutron channel description and validated against clean integral experiments.

3.2.1. Theoretical work

High resolution transmission data have been obtained at ORELA by J. Harvey et al. [11] using three samples, cooled at liquid-nitrogen temperature, the thicknesses of which were chosen so as to be a good compromise between getting accurate data and moderate self-screening effects. From these transmission data, a total cross-section has been derived. It is in agreement with the experimental data of Poenitz et al. [12] but significantly lower – by 3 to 4% – than JEF-2 or ENDF/B-VI. In our opinion the difference results from the self-screening correction and also from the better quality of raw transmission data.

The data base formed by Poenitz' and Derrien's data have been considered as total cross-section reference data in the range 1 to 500 keV.

The scattering radius and $S^{l=0}$ and $S^{l=1}$ neutron strength function values in the resonance range were extracted from the simultaneous fit by H. Derrien to derive the JEF-2 resonance parameter sets.

These were all ingredients needed to derive an optical model parametrization that was obtained by Ch. Lagrange in 1990 (OMP-90). Compared with the previous OMP used in JEF-2 (OMP-86) [13], real and imaginary parts of the potential were re-normalised; the ranges of the effective interactions and the spin-orbit potential remained unchanged. With this new optical model parametrization, an overall improved fit to the experimental data is obtained: neutron scattering angular distributions (see Figure 2) and the total cross-section above 0.5 MeV, which is an energy range rich in data (see Figure 3).

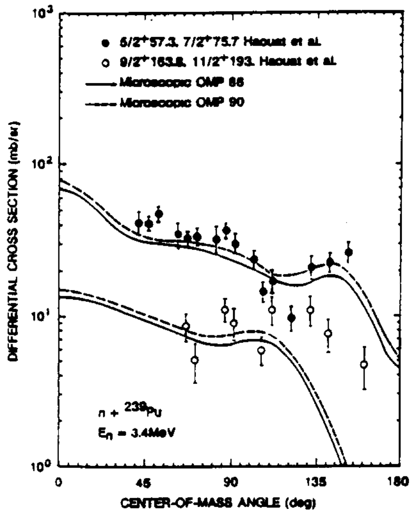

Figure 2

Comparison of experimental inelastic scattering cross-sections at 3.4 MeV with data calculated with OMP-86 and OMP-90

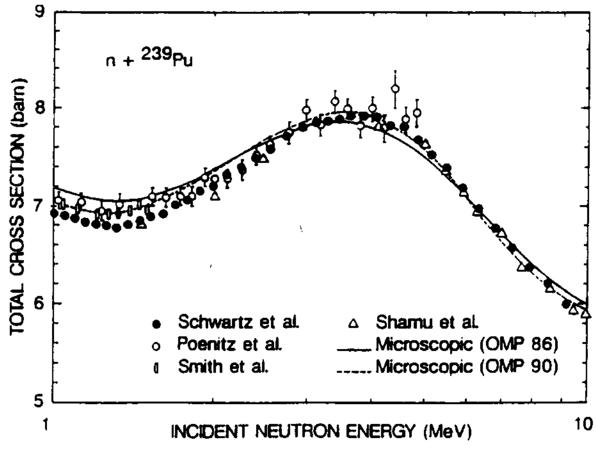

Figure 3

Experimental total cross-sections compared with data calculated with OMP-86 and OMP-90

In addition there is a significant decrease (10-15%) in the compound cross-section below 1.5 MeV (See Figure 4).

This is an important feature from which direct conclusions can be drawn.

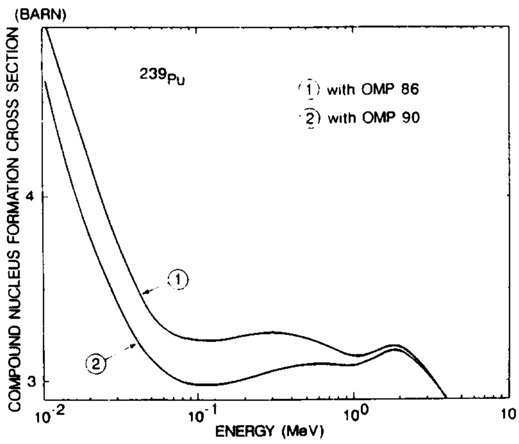

Figure 4
Comparison of the coumpound nucleus formation cross-section obtained by using OMP-86 (*curve 1*) and OMP-90 (*curve 2*)

The model evaluations ➊ is almost completed and shows that a fission cross-section based on Weston '84 is low with respect to all other evaluations up to 1 MeV, i.e., in the full sensitivity range of fast reactors (see Figure 5). We also observe, with respect to the JEF-2.2 evaluation, a 10%-lowering of the inelastic cross-section. This suggests that a model based on data from Poenitz, for example, would result in even lower values for the inelastic cross-section. Concerning the impact on integral data of k-eff, we note that a lowering of $\sigma_{n,n'}$ compensates to some extent the effect of a lowering of $\sigma_{n,f}$ because of the η-curve shape. Figure 5 also shows the amplitude of discrepancies between major values concerning this cross-section.

3.2.2. Validation of JEF-2.2 against integral data

The model evaluation ➋ has not been performed. The model evaluation ➊ has not been tested yet against chosen clean integral data, but the JEF-2.2

benchmarking, which is being performed allows some conclusions to be drawn. The conditions of this benchmarking are fully described in reference [15]. They can be summarised as follows:

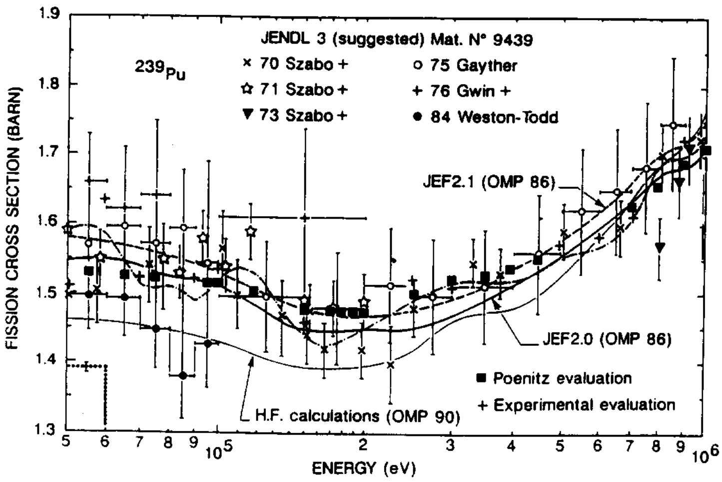

Figure 5
Experimental fission cross-section data compared with the model calculations (OMP-90) based on Weston's data

The integral data base was made of a large number of data (~200) of different types – *critical masses, Bucklings, spectral indices, neutron deep penetration data, ...* – for a large range of spectral hardness.

The JEF-2.2 library has been processed into a 1968-energy-group library containing infinite dilute cross-section and probability tables, which have been produced by using the NJOY-THEMIS and CALENDF codes, respectively. A great effort has been devoted to Quality Assurance, in order to ensure that data processing would preserve the information quality and integrity. The integral data have been re-calculated with this fine group library. Cross-section modifications, to minimise discrepancies between experimental and re-calculated integral data, have been obtained by a statistical adjustment according to the "*general least squares*" method. Since there is no uncertainty information in JEF-2, a previous systematic study had defined the covariance data needed for obtaining the best conditions for this adjustment procedure.

The quality of the data adjustment has been measured by the a posteriori χ^2 value which according to the theory should be equal to $N \pm \sqrt{2N}$, N being the number of degree of freedom – *number of integral data in the present case*. A technique based on the comparison of the practical χ^2 distribution with the theoretical one was used to identify in the integral data those data (~10%) giving spurions information in order to obtain $\chi^2/N = 0.997$.

The conclusions of this adjustment, published at the Gatlinburg Conference [18] have been carefully compared nucleus by nucleus with external independent information – experimental, critical analysis – obtained in the meantime and related to ^{235}U, ^{56}Fe, ^{58}Ni and ^{23}Na.

The agreement is excellent. The conclusions of this global adjustment can therefore be considered as perfectly reliable for ^{239}Pu. They are as follows:

- For $\sigma_{n,n}$: decrease by about 10%,

  This conclusion is perfectly consistent with the above-mentioned model calculation.

- For $\sigma_{n,f}$: if the following modifications are made, namely:

  +3.1% renormalisation of Weston's '84 data, on the one hand, and JEF2 σ_f-adjustment, on the other hand,

  then, the discrepancies – expressed by $\left(\frac{\text{JEF2}}{\text{Weston}} - 1\right)$ in %

  –are significantly reduced except for energy intervals of 3.355 – 9.119 keV and 9.119 – 24.79 keV located in the unresolved range.

This is explained as follows: The calculations for evaluation purposes are performed in Cadarache, CEA-France, with the FISINGA code [17] with a cross-section calculation formalism which is different from the one recommended by the ENDF/B prescriptions and used in the data processing code NJOY. *In particular this last one uses a single level formalism for the unresolved range and integer values for the degree of freedom for the* χ^2 *distribution for fission widths*. So, the evaluated average parameters have been translated into NJOY-type data in such a way that they respect the infinite dilute values but not the self-shielding factors, which are calculated with the CALENDF code from generated resonance ladders. Finally all effects result in too low effective fission cross-sections and consequently in anomalously high adjustment corrections.

On a qualitative level the benchmarking confirms first the need for a re-normalisation upwards by 3 to 4% but also the “general” shape of the fission cross-section as measured by Weston which comes out to be lower than most evaluations in the range 10 to 100 keV. More refined conclusions would have required a benchmark testing of the model evaluation ❶, as previously planned.

Table 5 ***Discrepancies between JEF-2 and Weston's data***

ENERGY GROUP STRUCTURE *eV*	JEF-2 *barn*	WESTON-84 *barn*	BEFORE MODIFICATION	AFTER MODIFICATION
4.5400E+02 - 1.2340E+03	7.091	6.99	1.44	-1±1.7
1.2340E+03 - 3.3550E+03	3.653	3.50	4.38	1.4±1.3
3.3550E+03 - 9.1190E+03	2.246	2.18	3.05	5.6±3.1
9.1190E+03 - 2.4790E+04	1.722	1.67	3.1	3.5±2.2
2.4790E+04 - 6.7380E+04	1.578	1.48	6.6	1.7±1.4
6.7380E+04 - 1.1110E+05	1.544	1.42	8.7	0.5±1.3

4. Conclusion

The experimental program performed in Geel and Oak Ridge has solved the problem of the normalisation in Weston’s experiment. It ended up in the definition of a standard value for $\mathbf{I_f}$ – *the fission integral between 100 and 1000 eV*, equal to **9275±85 b•eV**. Concerning the possible energy-dependent experimental effect, the exhaustive JEF-2 benchmarking tends to prove its non-existence although the demonstration requires more accuracy.

Due to low background and good energy resolution of the experiment, Weston’s ‘84 re-normalised cross-section values are proposed as reference data for any future ^{239}Pu evaluation, which should also take benefit from some other conclusions derived in the course of this work, such as the competitive data or the best presently avaible OMP for ^{239}Pu.

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