

^{239}Pu FISSION CROSS-SECTION BETWEEN 1 KeV and 100 KeV**Subgroup 5 report***CE Cadarache - BP N° 1 - Saint Paul lez Durance - France*E. FORT**ABSTRACT**

To understand the discrepancies between WESTON's data (NSE 88, 567, 1984) and major files evaluations, experimental and evaluation programs have been agreed upon.

The experimental program essentially aimed at normalization checking has been performed in GEEL and OAK-RIDGE. It supports a renormalization upwards by - 3.1 %.

The evaluation program is not 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 JEF2.2 benchmarking performed in another context supports the results of the experimental program, although it suggests a slightly larger renormalization.

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 KeV - 100 KeV. Expressed in terms of critical mass, control rod worth, void coefficient this 5 % difference is of importance for Fast Reactor Calculations.

To solve this problem a subgroup of NEA, subgroup 5, adopted the following two part plan :

1. Critical examination of WESTON's experiment reserving the possibility of additional experiments to check questionable points.
2. Examination of all other sources of information relating to the fission cross-section.

1.1. Critical examination of the Weston and Todd experiment

This is a T.O.F. experiment performed with ORELA, using a multiparallel plate fission chamber ; the shape of the neutron flux was measured relative to a $^{10}\text{BF}_3$ chamber up to a neutron energy a 1 KeV and to a ^6Li glass scintillator at higher energies. The flux internormalization was made in the energy interval 100 eV - 1000 eV with a statistical uncertainty less than 0.1 %. Finally the σ_f curve was normalised to the thermal value. For this experiment conceived for a measurement at high energy, the following aspects were considered to detract from such way of normalization : sample thickness self absorption, dead time. The energy range of flux intercomparison in WESTON'S experiment has been chosen to intercompare the various sets of data through fission integral values $\left(I_f = \int_{100\text{ eV}}^{1000\text{ eV}} \sigma_f(E) dE \right)$. For the

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

In addition when looking at the figure 1 which describes the situation and considering the POENITZ standard (16) as a good average picture of the major evaluations, one observes a $\sim 3\%$ difference between 0.1 KeV and 10 KeV and of $\sim 5\%$ between 10 KeV and 100 KeV. This situation of an energy dependent discrepancy raises the questions of a normalization error and of a possible energy dependent effect in WESTON's experiment.

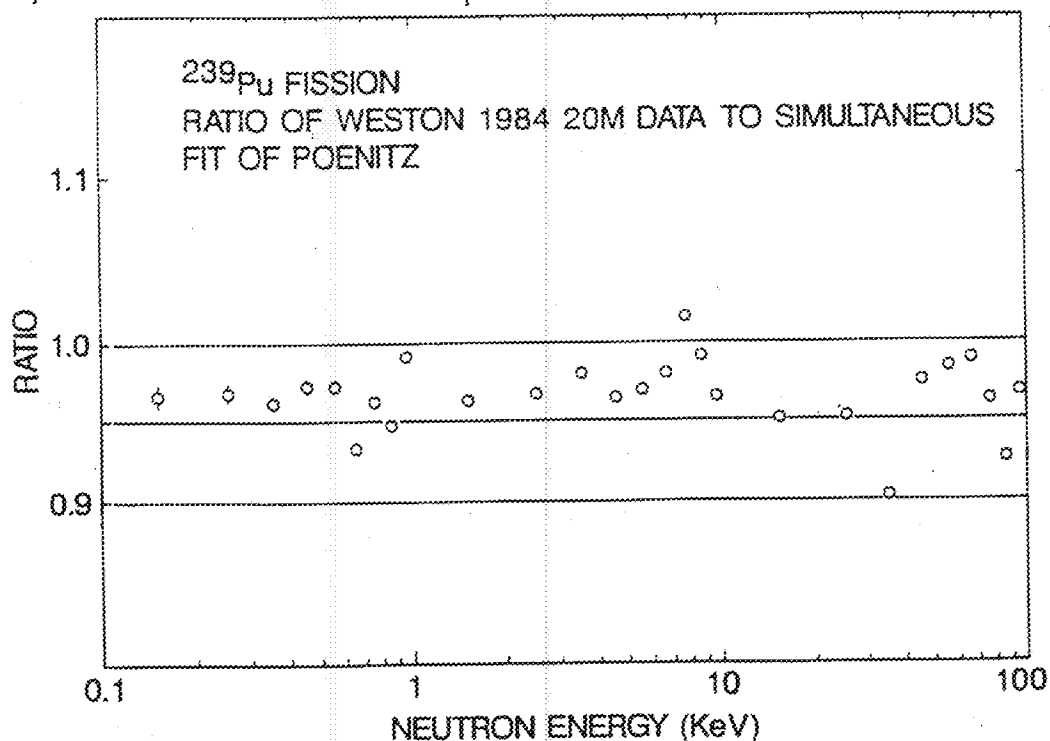


Fig. 1 - Ratio of WESTON's data (1984) to simultaneous fit of POENITZ

Therefore it has been decided to plan experimental programs both at OAK-RIDGE and GEEL to check this normalization point.

1.2. Experimental work performed in the framework of the subgroup 5

In GEEL (1992) two different geometries have been used, by C. WAGEMANS [3]:

- ① Measurement with a double ionisation chamber in a "2 π geometry".
The characteristics were as follows :
 ^{239}Pu sample : $186 \mu\text{g}/\text{cm}^2$,
 ^{10}B sample : $10 \mu\text{g}/\text{cm}^2$.

Flight path length : 8.5 m.

The normalization 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 meV and 60 meV,

$$\int_{20 \text{ meV}}^{60 \text{ meV}} \sigma_f dE = 25.36 \text{ b.eV.}$$

from which an average value of 634 b is obtained, to be compared with 631.4 b that is an average of several measurements selected by H. DERRIEN in his most recent resonance parameter evaluation [4] ;

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

With these conditions of normalization WAGEMANS derived $I_f = 9190 \pm 110 \text{ b.eV}$. 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 normalization.

② Measurement in "low geometry" using surface barrier detectors.

The ^{239}Pu sample was $106 \mu\text{g}/\text{cm}^2$ and the ^{10}B sample was $10 \mu\text{g}/\text{cm}^2$.

The neutron flight path length was : 8.3 m.

For this measurement, the fission integral value is $(9450 \pm 200) \text{ b.eV}$. In the final uncertainty the components are 0.8 %, 0.5 % and 1 % respectively, due to counting statistics, background and normalization corrections.

Considering both data a weighted average value is obtained for I_f : ${}^{\text{Wa}}I_f = 9250 \pm 96 \text{ b.eV}$.

In OAK-RIDGE a new fission cross section measurement has been performed by WESTON and coworkers [9]. The experimental technique was similar as previously, but the detectors were different : a parallel plate ^{10}B ionization chamber with solid coating of Boron on the center plate was used rather than $^{10}\text{BF}_3$ gas. The fission chamber was also different in that sense that the active plates were loaded with ^{239}Pu only, so that better counting statistics is obtained for the same irradiation time. The flight path was 19 m and the measurement was continuous from below 0.025 eV to 10 keV. Careful attention was paid to the normalization that is more precise than in the previous measurement (0.5 % against 1.9 % [8]) while the overall systematic uncertainty is estimated to be 0.6 % (to be compared with 1 %). The normalization was done on the integral of $\sigma_f \sqrt{E}$ over the interval 0.02 eV to 0.03 eV referring to ENDF B6 evaluation ($\int_{0.02 \text{ eV}}^{0.03 \text{ eV}} \sigma_f \sqrt{E} dE = 118.98 \text{ b.eV}^{3/2}$). The I_f value obtained in these conditions is : ${}^{\text{We}}I_f = 9302 \pm 102 \text{ b.eV}$ (the total uncertainty on this value is estimated from the values just above mentioned about normalization and systematic uncertainty and including a 0.1 % component due to the statistical uncertainty).

This normalization operation is consistent with the set of normalization 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 normalized on the ENDF B6 thermal value of the fission cross section of ^{239}Pu as a primary standard and adapting the ENDF B6 values for the $^{10}\text{B}(n,\alpha)^7\text{Li}$ cross-section.

The final I_f value that results from the experimental work for the purpose of normalization is obtained from a weighted averaging of WAGEMANS and WESTON's data displayed in the Table 1.

Table 1

WAGEMANS ₁ ⁹²	WAGEMANS ₂ ⁹²	WESTON ⁹²
9190±110	9450±200	9302±102

Standard $I_f = 9275 \pm 85$ b.eV.

This 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 steps measurements or any measurement subject to energy dependent effects. In the following we will refer to it as the standard I_f .

The Normalization Factor 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 rare measurements performed in the range thermal 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(*)

One has to note that all the fission integrals here above cited are normalized on $\sigma_f = 748.0$ b at thermal energy.

(*) This recalculated using the ENDF B6 values for the $^{10}\text{B}(n,\alpha)^7\text{Li}$ cross-section.

2. Examination of all other sources of information

In what follows we describe works which have been developed in the framework of the subgroup 5 to explore an other way than the experimental way. They bring an indirect support to the normalization but they show that the major evaluations which were implied in the discrepancy should be improved, although they are satisfying in several aspects. These works provide data, pieces of information or tools which should be used for evaluation improved in large energy ranges.

2.1. A specific study made in JAPAN (NAKAGAWA, KAWAI, ...) has shown that other types of data such as alpha data or competitive reaction (capture, non elastic, inelastic, ...) data are too inaccurate to be used as references.

In the resolved range, H. DERRIEN and NAKAGAWA in JAERI 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, with allowance for background and normalisation coefficient adjustment. The following experimental data base has been 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 ensemble from which the previous resolved resonance parameter set (included in JEF2.2 and ENDF-B6) has been derived, additional sets are included, in particular the high resolution transmission data by HARVEY et al [11] and the high resolution fission cross-section measurement by WESTON and TODD (1988) [14]. If one uses this evaluation one obtains for the range 100 eV - 1000 eV a fission integral equal to 9304 b.eV to be compared to I_f values obtained from major evaluations as quoted in Table 3.

Table 3 - I_f values (in b.eV) related to evaluated data

ENDF-B6	JEF2.2	JENDL3	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 appearing in the Table 4 below.

Table 4 - Averaged fission cross-section

Energy (eV)	DERRIEN NAKAGAWA 1992	JENDL3	JEF2	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

2.2. Evaluation and validation work

At the beginning it was decided to perform two model calculations to describe the fission data, one based on WESTON's 84 eV , another one based on ENDF-B6 eV . Both would be based on the same neutron channel description and validated against clean integral experiments.

Neutron channel description :

High resolution transmission data have been obtained at ORELA by J. HARVEY et al [11] using 3 samples cooled at liquid nitrogen temperature whose thicknesses were chosen so as to be a good compromise between opposite conditions to get accurate data and moderate self screening effects. From these transmission data a total cross-section has been derived that is in agreement with POENITZ et al [12] experimental data but significantly lower (3 % - 4 %) than JEF2 or ENDF-B6. The difference results, in our judgement, from the self screening correction and also from the better quality of the raw transmission data.

The data base formed by POENITZ's and DERRIEN's data have been considered as total cross-section reference data in the range 1 KeV - 500 KeV.

The scattering radius and $S^l=0$ and $S^l=1$ neutron strength functions values in the resonance range, were extracted from the simultaneous fit by H. DERRIEN to derive the JEF2 resonance parameter sets.

These were all ingredients needed to derive an optical model parametrization that has been obtained by C. LAGRANGE in 1990 (OMP 90). Compared to the previous OMP used in JEF2 (OMP 86) [13] real and imaginary parts of the potential were renormalized, the ranges of the effective interactions and the spin-orbit potential remained unchanged. With this new optical model parametrization one obtains an overall improved fit to the experimental data : the neutron scattering angular

distributions (see fig. 2 below) but essentially the total cross-section above 0.5 MeV in an energy range rich in data (see fig. 3).

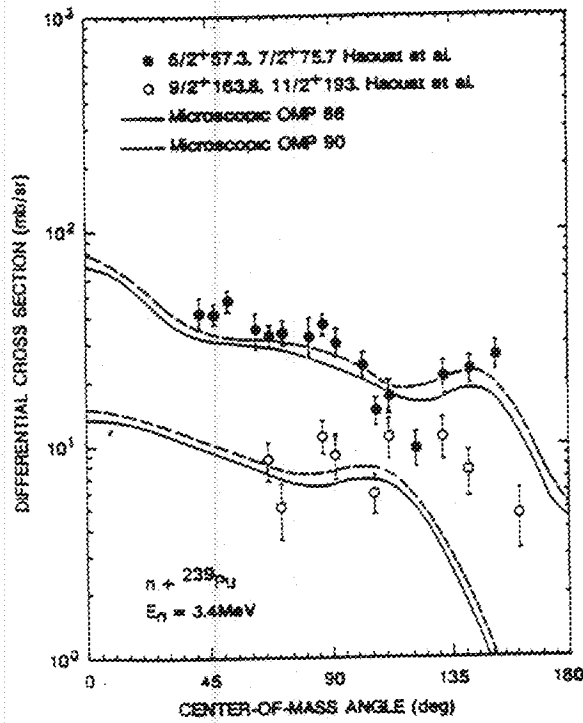


Fig. 2 - Comparison of experimental inelastic scattering cross-sections at 3.4 MeV with data calculated with OMP86 and OMP90

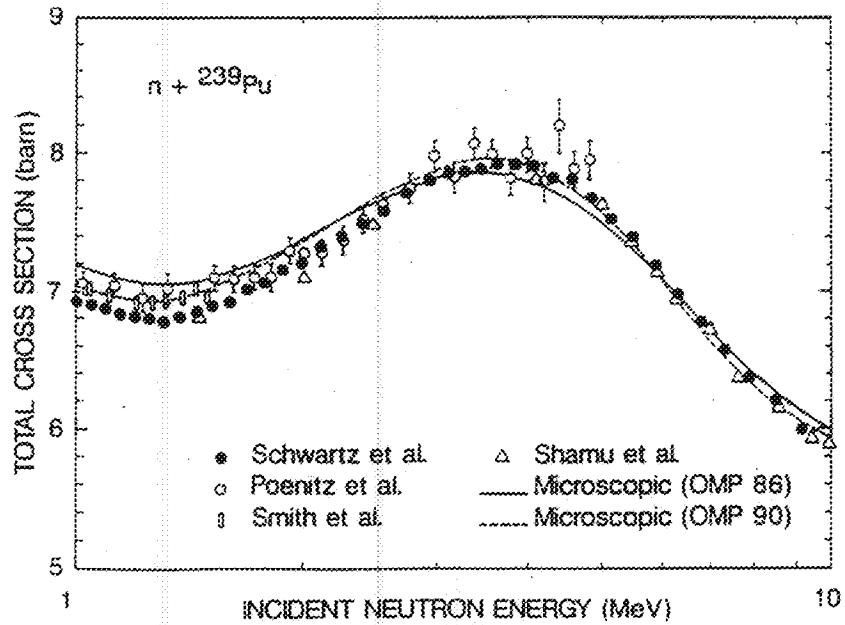


Fig. 3 - Experimental total cross-sections compared with data calculated using OMP86 and OMP90

In addition there is a significant lowering (10 % - 15 %) of the compound cross-section below 1.5 MeV (see figure 4).

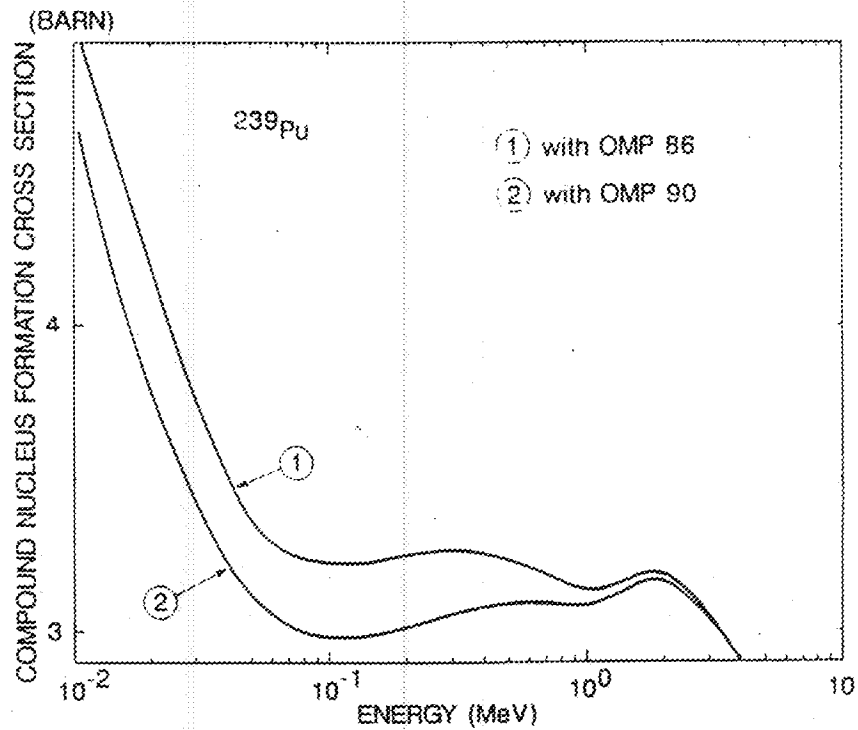


Fig. 4 - Comparison of the compound nucleus formation cross-section obtained by using OMP86 (curve 1) and OMP90 (curve 2)

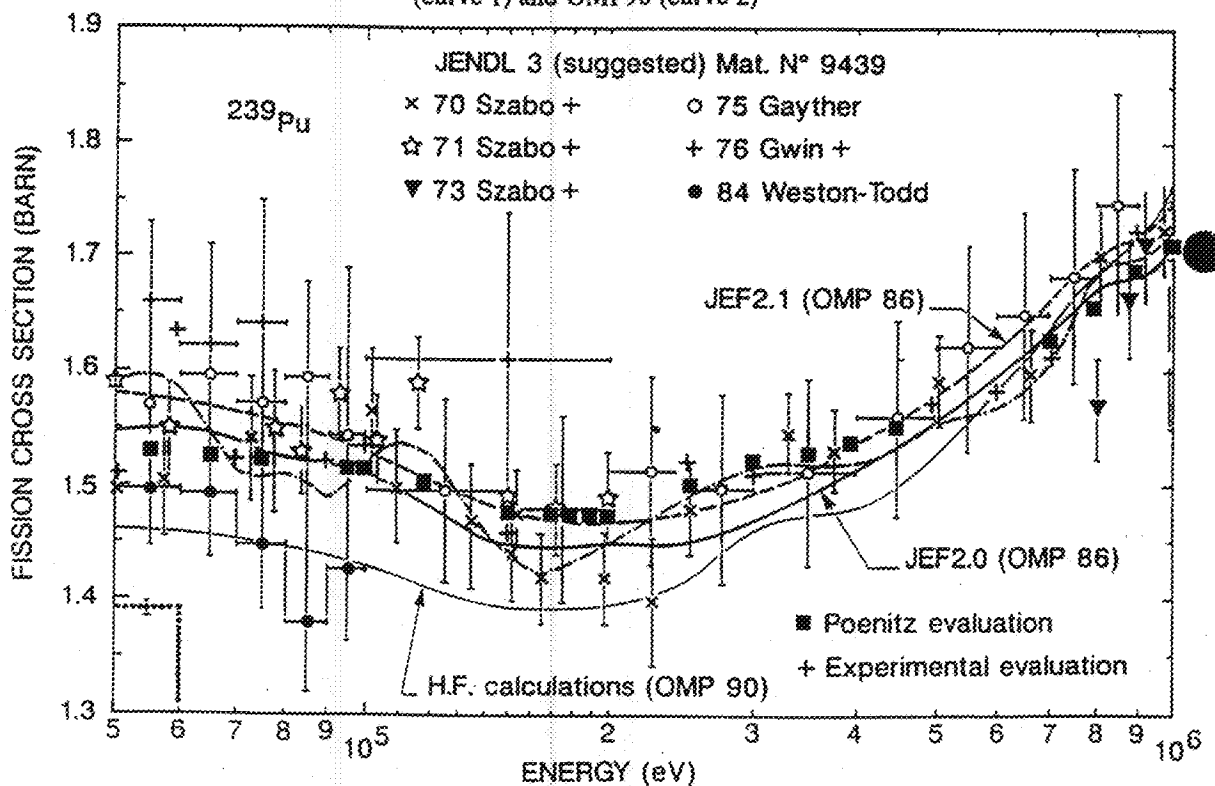


Fig. 5 - Experimental fission cross-section data compared with the model calculations (OMP90) based on WESTON's data

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

The model evaluations ① is almost completed and shows (see figure 5) that a fission cross-section based on WESTON 84 is low with respect to all the other evaluations up to 1 MeV, i.e., in the range of full sensitivity of Fast Reactors. One observes also with respect to the JEF2.2 evaluation a lowering of the inelastic cross-section by about 10 %. This suggests that a model calculation based on POENITZ's data for example would result in still lower values for the inelastic cross section. For what concerns the impact on integral data of K_{eff} one notes that a lowering of $\sigma_{n,n'}$ compensates in some extent the effect of a lowering of $\sigma_{n,f}$ because of the shape of the ETA curve. The figure 5 also exhibits the amplitude of the discrepancies between major evaluations concerning this cross section.

The model evaluation ② has not been performed. The model evaluation ① has not been yet tested against chosen clean integral data, but the JEF2.2 benchmarking that is being performed allows some conclusions to be transposed. The conditions of this benchmarking are fully described in the reference [15]. They can be summarised as follows :

The integral data base was made of a large number of data (~ 200) of different types (critical masses, Bücklings, Spectral Indices, neutron deep penetration data...) for a large range of spectral hardness.

The JEF2.2 library has been processed into a 1968 energy groups 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 to be sure that the data processing preserved the quality and the integrity of the information. The integral data have been recalculated with this fine group library. The cross-section modifications to minimise the discrepancies between the experimental and recalculated integral data have been obtained by a statistical adjustment according to the "general least squares" method. Since there is neither uncertainty information in JEF2, a previous systematic study had defined the covariance data needed to obtain the best conditions for this adjustment procedure.

The quality of the data adjustment has been measured by the a posteriori χ^2 value that according to the theory should equal $N \pm \sqrt{2N}$, N being the number of degree of freedom that is, in the present case, the number of integral data. A technique based on the comparison of the practical χ^2 distribution to the theoretical one was used to identify in the integral data base those data (~ 10 %) giving spurious information, so that it has been possible to obtain $\chi^2/N = 0,997$.

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

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

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

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

- For $\sigma_{n,f}$: if the following modifications are made : namely + 3.1 % renormalization of WESTON's 84 data on one hand, JEF2 σ_f adjustment as obtained in the benchmarking on the other hand, then the discrepancies (expressed by $\left(\frac{\text{JEF2}}{\text{WESTON}} - 1\right)$ in percent) are significantly reduced (see table 5) except for the energy intervals. 3.355 KeV - 9.119 KeV and 9.119 KeV and 24.79 KeV located in the unresolved range.

This is explained as follows : the calculations for evaluation purpose are performed in CADARACHE with the FISINGA Code [17] with a formalism for the cross section calculation that is different from the one recommended by the ENDF/B prescriptions and used in the data processing code NJOY. (In particular this 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 a way that respects the infinite dilute values but not the selfshielding factors that 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.

Table 5 - Discrepancies between JEF2 and WESTON's data

Energy group structure (eV)	JEF2-2 (barn)	WESTON84 (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

On a qualitative level the benchmarking confirms the need for a renormalization upwards by about 3 ÷ 4 percents but also the "general" shape of the fission cross-section as measured by WESTON which exhibits in the range 10 KeV - 100 KeV a level lower than in most evaluations. More refined conclusions would have required a benchmarking applied on the model evaluation, as previously planned.

Conclusion

The experimental program performed in GEEL and OAK-RIDGE has solved the problem of the normalization in WESTON's experiment. It ended in the definition of a standard value for I_f , the fission integral between 100 eV and 1000 eV, equal to 9275 ± 85 b.eV. Concerning the possible energy dependent experimental effect the exhaustive JEF2 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 renormalized cross section values are reference data for any future ^{239}Pu evaluation which should also take benefit of some conclusions derived in the course of this work, related to the competitive data or to the best presently available OMP for ^{239}Pu .

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