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THERMAL NEUTRON ACTINIDE DATA

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INTRODUCTION

During the 70's, the physicists involved in the cross section measurements for the low energy neutrons were almost exclusively interested in the resonance energy range. The thermal range was consedered as sufficiently known. In the begining of the 80's, reactor physists had again to deal with the delicate problem of the power reactor temperature coefficient, essentially for the light water reactors. The measured value of the reactivity temperature coefficient does not agree with the computed one. The later is too negative. For obvious safety reasons, it is an important problem which must be solved. Several causes were suggested to explain this discrepancy. Among all these causes, the spectral shift in the thermal energy range seems to be very important. Sensibility calculations shown that this spectral shift is very sensitive to the shape of the neutron cross sections of the actinides for energies below one electron-volt. Consequently, reactor physisists require new and accurate measurements in the thermal and subthermal energy ranges [1,2]. A part of these new measurement results were recently released and reviewed [3]. The purpose of this study is to complete the preceding review with the new informations which are now available. In reactor physics the major actinides are the fertile nuclei, uranium 238, thorium 232 and plutonium 240 and the fissile nuclei, uranium 233, uranium 235 and plutonium 239. For the fertile nuclei the main datum is the capture cross section, for the fissile nuclei the data of interest are nu-bar, the fission and capture cross sections or a combination of these data such as η or α. In the following sections, we will review the neutron data of the major actinides for the energy below 1 eV.

URANIUM 238

At low energy, the cross section shapes of uranium 238 are given by the 4.4 eV "p" wave and the 6.67 eV "s" wave resonances. These two resonances induce for capture cross section a 1/v behaviour in the thermal range. It is the shape which is universally adopted by all the evaluated files. But a part of the temperature coefficient descrepancy could be explain by a non 1/v dependence of the uranium 238 capture cross section for the low energy neutron. The cross section must decrease with the energy faster that the 1/v shape. This effect can be obtained with the assumption of a weak bound level near the zero energy. L. Erradi proposed a small resonance at - 0.005 ev [4]. This hypothesis was supported by the measurement of the fission cross section at 0.025 ev which was performed in Grenoble [5]. The experimental value cannot be exclusively explained by the contribution of the nearby resonances. An extra resonance is needed. If this resonance is close to the zero energy as in the Erradi's assumption, it must have an impact on the cross section shape in the thermal range. The non 1/v shape which is obtained with the - 0.005 eV resonance is not incompatible with the Harwell measurement of the uranium 238 capture cross section [6] but only because the experimental incertainties of this measurement are rather large. New and more accurate measurements of the uranium 238 capture cross section were required. These measurements were performed at the Geel laboratory [7]. As it can be seen on figure 1 which represents the experimental variation of $\sigma\gamma \sqrt{E}$ as a function of the neutron energy, and a comparison with the JEF evaluation, these results confirm without ambiguity a 1/v behaviour for the capture cross section. The assumption of the resonance in the immediate vicinity of the zero energy and the explanation of a part of the temperature coefficient discrepancy by such a resonance must be dropped. Reasonably, we must consider that the uranium 238 capture cross section problem is solved in so far as the thermal energy range is concerned.

THORIUM 232

Nowadays, the use of thorium fuel cycle in thermal neutron reactors is no more a high priority and the physicist interest for thorium nuclear data is less important in the case of low energy neutron than in the fast range. Consequently the situation is fondamuntally different from the one of uranium 238. That is why no request recently appeared for the thermal neutron energy range. In this energy domain it exists only two recent differential measurements which give access to the shape of the thorium 282 capture cross section. They are the measurement performed in Brookhaven for the energies between 35 and 1000 meV [8] and the experiment of RPI which covered the neutron energies above 10 meV [9]. These two sets of experimental data are compared on figure 2 which displays the variation of $\sigma_\gamma \sqrt{E}$ versus the neutron energy. If we take into account the experimental uncertainties, the agreement between the two measurements is good enough above 50 meV. They are also in good agreement with the recommended value of the ENDF/B6 evaluation. In the very low energy domain, between 10 and 25 meV, it exist only one measurement which significantly deviates from the evaluated recommendation. The experimental cross section decreases less than a 1/v shape. We know that the temperature coefficient of a multiplying lattice is very sensitive to the shape of the fertile nucleus capture cross section below 25 meV. In the case of a thorium cycle revival, the observed discrepancy between the measurement and the evaluation must be clarified as it was recently done for uranium 238. New and accurate measurement of the thorium 282 captur cross section would be needful in the thermal and subthermal energy range.

PLUTONIUM 240

For this isotope, the cross section behaviour in the thermal energy range is mainly governed by the 1.056 eV resonance. Consequently it is necessary to have a very accurate knowledge of this resonance parameters. Only two measurements of the 1.056 eV resonance parameters were recently carried out, the experiment of Brookhaven [10] and the one of Oak Ridge [11]. In the Brookhaven experiment, total and capture cross sections with room temperature and cooled samples were used. In the Oak Ridge measurement, transmission measurements with seven thicknesses of sample were performed. Thus it was expected that the results could be very satisfactory. Unfortunately, as it can be seen in table 1, the two sets of results are significantly discrepant.

Table 1 : Parameters of the ^{240}Pu 1.056 eV resonance

Γ_n (meV)	Γ_γ (meV)	Laboratory
2.32 ± 0.06	32.4 ± 0.6	Brookhaven (81) [10]
2.45 ± 0.02	30.3 ± 0.3	Oak Ridge (87) [11]

A detailed analysis of the two experiments leads to be more confident in the second set of results. But the interpretation of spent fuel isotopic composition suggests tendencies closer to the high value of radiative capture width. The différence between the two series of resonance parameters induces a change of 1.2 percent on the contribution of the 1.056 eV resonance to the capture cross section at 0.025 eV and a change of 4.5 percent on the resonance integral value. These modifications become very important every time that the plutonium 240 is significantly involved. It is main by case for irradiated fuel analysis or for plutonium recycling is light water reactor. The discrepancy between the two differential measurements and the tendency deduced from the integral experiments must be clarified.

URANIUM 235

In the case of a fissile nucleus, the problem is more complex than for a fertile nucleus. In addition to the capture cross section, other fundamental nuclear data are involved, the number ν of neutron which are emitted in a fission and the fission probability. These three quantities or a combination of them such as η or α must be investigated. As it has been shown that the temperature coefficient is very sensitive to the shape of the various nuclear parameters versus the neutron energy, several differential measurements were performed during the last years. They are relative to the fission cross section, nu-bar and eta in the very low energy range.

a) **Nu-bar**

One measurement of nu-bar was recently performed in the energy range which we are interested in. It is the Oak Ridge experiment which gives the ratio of the uranium 235 prompt nu-bar over the Californium ν_{sp} of spontaneous fission for the neutron energies between 5 meV and 1 eV [12]. According to these results, which are displayed on figure 3, nothing appears in the vicinity of the 0.29 eV resonance and we can reasonably keep the assumption of a constant value of nu-bar below 1 eV. This flat shape, which is adopted in all the evaluated files, has important consequences for the capture cross section behaviour at low energy.

b) **Fission cross section**

Since 1984, very accurate measurements was performed in the low energy neutron range [13, 14, 15]. As it can be seen on figure 4, above 20 meV and in particular in the 0.290 eV resonance all these results are in agreement between them and with the most recent evaluated files ENDF/B5 and JEF2. In the subthermal energy range, below 5 meV, only the Geel experiment gives informations. According to this measurement the uranium 235 fission cross section reaches an 1/v shape for energies higher than it was assumed in ENDF/B5. On the contrary, JEF2 which was released after the Geel experiment takes into account its results below 10 meV and adopts a fission cross section shape closer to a 1/v behaviour.

c) **Eta measurements**

To explain the temperature coefficient discrepancy of the uranium fuel reactor, it is $\eta = \nu \frac{\sigma_f}{\sigma_\alpha}$ which is the most sensitive parameter. In all the previous evaluated files, including ENDF/B5 and JEF1 eta was assumed to have a constant value below 0.1 eV. As the reactor physicists proposed to increase eta between 5 and 100 meV, measurements of this neutron parameter were necessary to validate the modification of shape. Four measurements of eta were recently performed in the range of interest.

The first one is the Geel experiment [16]. This experiment was performed with a linac and a liquid methane moderator to inhance the importance of the low energy neutrons. It covered the neutron energy between 2 and 450 meV. As it is shown in figure 5 the results suggest an increase of eta between 2 and 80 meV. It is in the good direction for the reactor physics.

A second measurement was carried out with the Harwell linac [17]. Unfortunately the number of low energy neutron was not very high. Consequently the accuracy was not good enough. Nevertheless this experiment did not show a significant shape of eta versus the neutron energy as it is displayed on figure 6. It is contradictory with the Geel results.

A third experiment was performed in Grenoble [18]. Instead of a linear accelerator, like in Geel, the neutron source was constituted by a cold neutron beam of the high flux reactor and more neutron of low energy were obtained. The accuracy was then expected to be better because the background would be lower. In the energy range between 2 and 150 meV. this experiment perfectly confirms the Geel results and the shape of eta versus energy as it is shown on figure 5.

Finally a fourth experiment was carried out with the Oak Ridge linac [19]. The preliminary results of the last experiment are compared with the Harwell results on figure 6. These two series of results seem more or less in agreement and do not show a significant shape of eta.

These four experimental results can be splitted in two sets, a first set which indicates a shape of eta (the Geel and Grenoble data) and the second set which does not (the Harwell an Oak Ridge data). As the low neutron flux was the highest in the Grenoble experiments, it is possible to given a more important weight to these results and to propose a slope for the eta shape below 100 meV. This attitude was adopted for the preliminary version of JEF2. But for the physical point of view, the desagreement between the two sets of results is not acceptable. On behalf on the NEA Nuclear Science Comittee, a working group carefully studied the various corrections (count loss, background substraction, absorption...) which were applied to the raw data of the four measurements. The final recommendations of the working group are not established yet but the preliminary results are encouraging. It seems that it may be quite possible to define a curve of eta with an energy dependent shape. Eta would increase of about 1,3 % between 3 and 80 meV and this energy dependence would be compatible with the four experimental data [20]. This shape would be close to the reactor physicist suggestion.

d) **Alpha measurement**

A complementary and important information upon the uranium 235 cross section in the thermal energy range is given by the recent Geel measurement of α [21]. It is a very interesting result because it constitutes an independent way to obtain information about $\eta = \frac{\nu}{1 + \alpha}$. As it can be seen on figure 7, the experimental values of α are not reproduced by the ENDF/B5 evaluation. As all the previous files, ENDF/B5 recommend a flat shape of α below 100 meV. As ν is energy independent this shape corresponds to a constant value of η. On the contrary, it was adopted in JEF2 a slope for η and this file, which was released before the experimental values of α, is in good agreement with the measurement results. It is an important fact, because now, we have a coherent set of experimental data for ν, σ_f, η and α, which confirm the slope of η, as it was suggested by the integral experiment.

URANIUM 233

For the same reasons as for thorium, the uranium 233 nuclear data in the low energy range was not systematically studied these last years. Nevertheless it exist some scarce results, mainly for nu-bar and the fission cross section. These measurement were generally performed in the same campaign as uranium 235 when the physicists have uranium 233 samples at their disposal. For η and α, nothing new is available.

a) **Nu-bar**

The only result about nu-bar below 1 eV is the one of Oak Ridge [12] which gives the ratio of uranium 283 prompt nu-bar to the spontaneous fission nu-bar of californium 252. No significant structure was observed in this energy range, as it is shown as figure 8 and we can reasonally admit the flat shape which is adopted in ENDF/B6. But don't forget that the ENDF/B6 absolute value of nu-bar is not in agreement with the tendency which is deduced from the buckling measurements [22].

b) **Fission cross section**

At the opportunity of the campaign of measurements on fissile nuclei, an accurate determination of the shape of the uranium 233 cross section shape was carried out with the Geel linac [23]. In order to inhance the low energy neutron flux and obtain a good accuracy in the thermal and subthermal energy range, a liquid nitrogen cooled moderator was used. In these experimental conditions we can be very confident in the results which are displayed on figure 9. They are also in fair agreement with the ENDF/B6 recommendation. Reasonably we can admit that the shape of the uranium 233 fission cross section is well known below 1 eV.

PLUTONIUM 239

All the old evaluations of the plutonium 239 neutron data, including ENDF/B5, are considered as not satisfactory by the reactor physicists. As a matter of fact, in all these files it was adopted a flat behaviour of nu-bar in the low energy range, the spin of the resonances was not considered and it was used a Breit and Wigner formalism to compute the cross sections. With the high burn up fuels and the recycling, the plutonium became more and more important in the thermal neutron reactors. An updating of the plutonium 239 neutron data was strongly required. This updating was performed by Derrien et al. [24] who took into account new experimental results, concerning nu-bar [12], the fission cross section [13] and the total cross section [11], and used a Reich and Moore formalism which is more convenient for the fissile nuclei.

a) **Nu-bar**

On the contrary to the uranium 233 and uranium 235 cases, the recent measurement of plutonium 239 nu-bar [12] shows an important decrease in the vicinity of the low energy resonance at 0.3 eV. This strong structure is well reproduced by the Fort's theoritical calculation which takes into account the spin effect and the (n, γf) effect of the J = 1 resonances of plutonium 239 [25]. Figure 10 represents a comparison between the experimental values of nu-bar, normalized to the spontaneous fission nu-bar of californium 252, and the evaluation of Fort et al. As the agreement is very good this shape of nu-bar was included in JEF2.

b) **Fission cross section**

All the new evaluated files use the resonance parameter set which was deduced from the Derrien's analysis. The behaviour of the plutonium 239 cross sections in the thermal range is well reproduced by the contributions of the low energy resonances and of the bound level. After the release of the initial version of the recent evaluated files a new measurement of the plutonium 239 fission cross section became available [23]. The figure 11 shows the comparison of these new experimental values with the recommended values of JEF2 below 1 eV. The agreement is quite satisfactory and the new fission cross section measurement constitute a confirmation of the recommended values. Today, no request upon the plutonium 239 fission cross section seems needful as far as the low energy is concerned.

CONCLUSION

Since a few years, the status of the thermal neutron data for the major actinides was greatly improved. The recent measurements of the microscopic data led to a better knowledge of the cross section shapes in the low energy domain. Several problems which were of great importance for reactor physics were solved. Let us mention the 1/v dependance of the uranium 238 capture cross section and the behaviour of the uranium 235 fission cross section below 20 meV. The structure of the plutonium 239 nu-bar, which was not taken into account in the past, was well established and theoritically explained. We can reasonably expect that the uranium 235 eta discrepancy will be solved in the next future. Nevertheless it remains some problems which have to be further investigated. The most important, today, is the discrepancy between the two sets of parameters of the 1.056 eV plutonium

240 resonance. This desagreement has an important impact in high burn up fuels or plutonium recycle studies. The difference between the evaluated values and the measured values of the thorium 232 capture cross section below 20 meV and the problem of the absolute value of the uranium 233 nu - bar have a lower priority. But in the case of new interest for the thorium cycle, requests upon these two actinides will be certainly needful.

REFERENCES

[1] - J.L. Rowlands : "Neutron physics and Nuclear Data - Harwell", 7 (1978).

[2] - J. Bouchard, C. Golinelli et H. Tellier : "Neutron data for Science and Technology - Anvers", 21 (1982).

[3] - H. Tellier : "Neutron Data for Science and Technology - Jülich" (1991).

[4] - L. Erradi : These doctorat es sciences. Paris Orsay (1982).

[5] - P. D'Hont et al. : Annals of Nuclear Energy, 11, 485 (1984).

[6] - M.C. Moxon and J.E. Jolly : "Uranium and Plutonium resonance parameters - Vienna" 124, INDC-129 (1981).

[7] - F. Corvi and G. Froni : "Nuclear Data for Science and Technology-Mito", 127 (1988).

[8] - R. E. Chrien et al. : Nuclear Science and Engineering, 72, 202 (1979).

[9] - R.C. Little et al. : Nuclear Science and Engineering, 79, 175 (1981).

[10] - H.J. Liou and R.E. Chrien : "Uranium and Plutonium resonance parameters - Vienna" 438, INDC-129 (1981).

[11] - R.R. Spencer et al. : Nuclear Science and Engineering, 96, 318 (1987).

[12] - R. Gwin et al. : Nuclear Science and Engineering, 87, 381 (1984).

[13] - R. Gwin et al. : Nuclear Science and Engineering, 88, 37 (1984).

[14] - C. Wagemans and A.J. Deruytter : "Nuclear Data for Basic and Applied Science - Santa Fe" I, 499 (1985).

[15] - R.A. Schrack : "Nuclear Data for Science and Technology - Mito", 101 (1988).

[16] - J.A. Wartena et al. : Report IAEA Tecdoc 491, 123 (1987).

[17] - M.C. Moxon et al. : "Nuclear Data for Science and Technology - Mito", 75 (1988).

[18] - J.A. Wartena et al. : Report NEANDC (E) 312, III (1989).

[19] - M.C. Moxon et al. : Private communication to the NEA Data Bank (1990).

[20] - H. Weigmann : Private Communication (199[illegible]).

[21] - H. Weigmann et al. : "Nuclear Data for Science and Technology - Jülich" (1991).

[22] - H. Tellier and J. Vanuxeem : (This meeting).

[23] - C. Wagemans et al. : "Nuclear Data for Science and Technology - Mito", 91 (1988).

[24] - H. Derrien et al. : Nuclear Science and Engineering, 106, 434 (1990).

[25] - E. Fort et al. : Nuclear Science and Engineering, 99, 375 (1988).

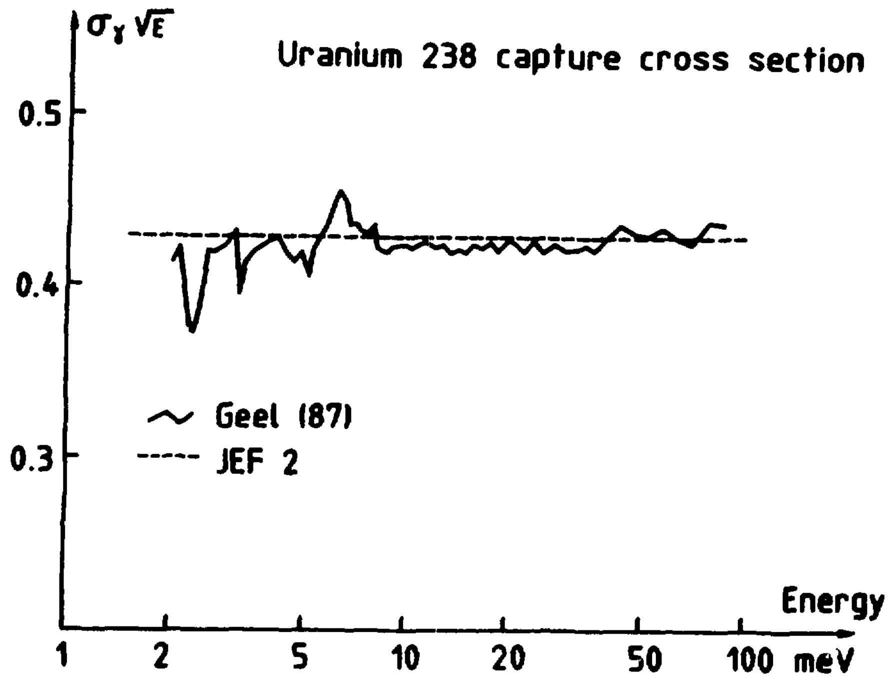

Figure 1 - Comparison between the uranium 238 capture cross section measurement of Geel, for a metal sample, and the evaluated values of JEF2.

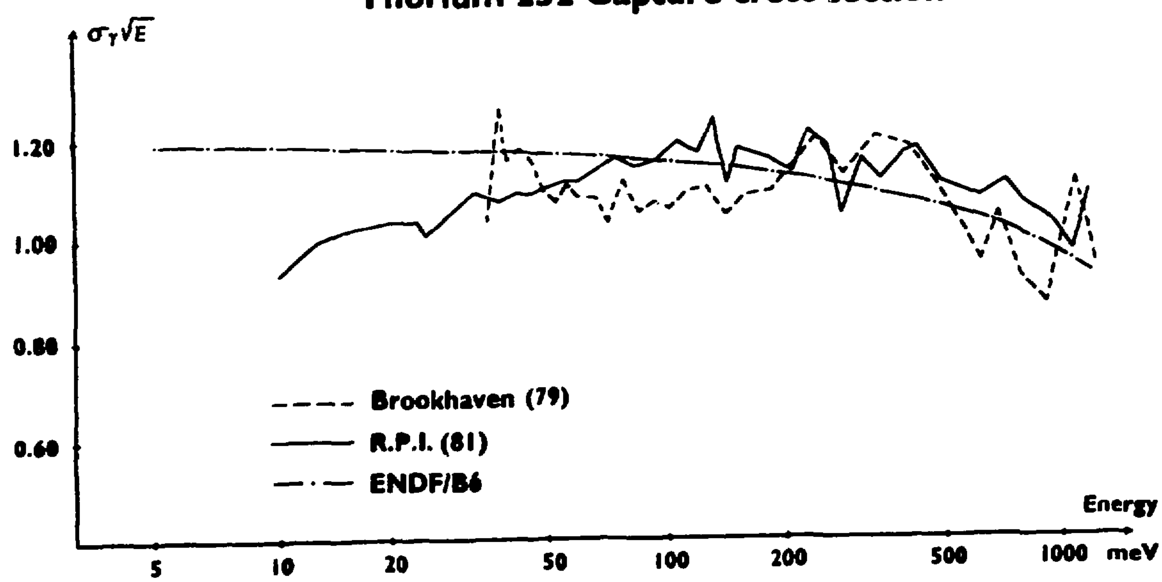

Figure 2 - Comparison of the recent measurements of the thorium 232 capture cross section and the evaluated values of ENDF/B6.

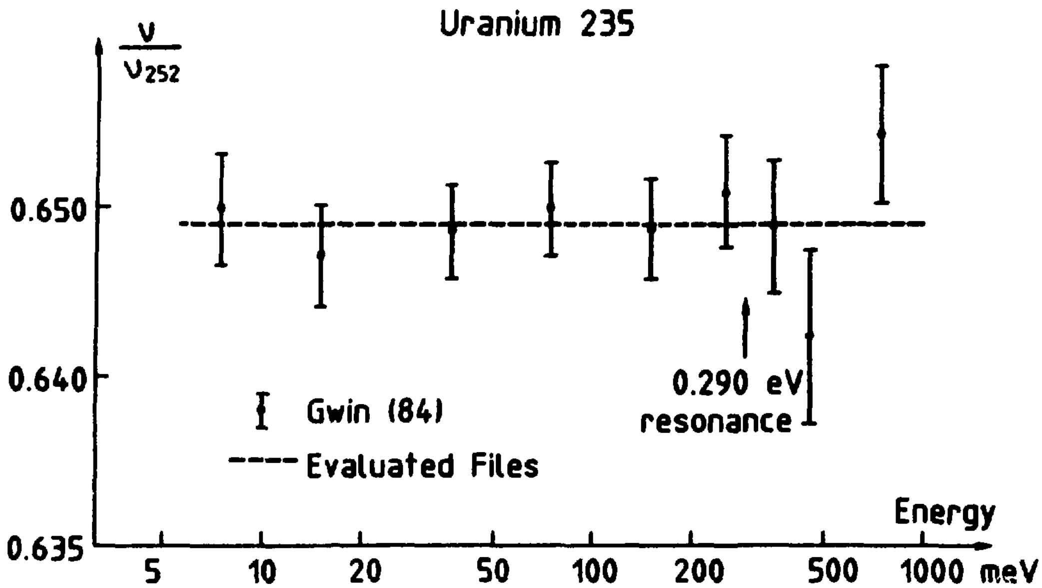

Figure 3 - Ratio of uranium 235 nu-bar to the one of californium 252 in the thermal energy range and comparison with the evaluated values.

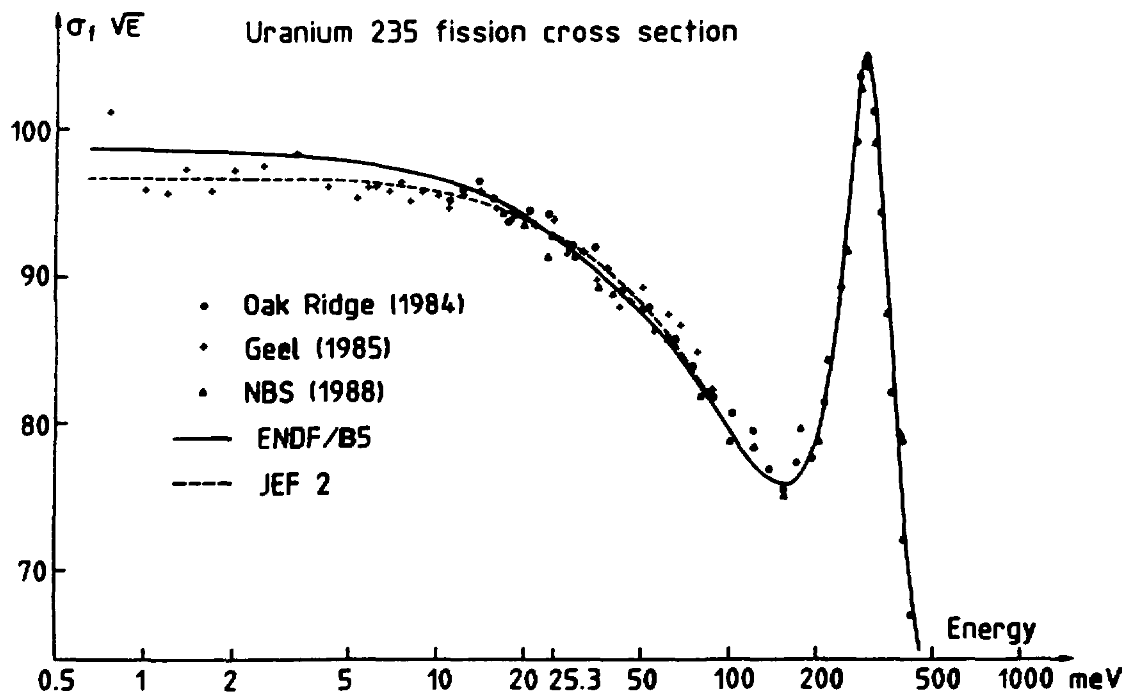

Figure 4 - Recent measurements of the uranium 235 fission cross section and comparison with ENDF/B5 and JEF2.

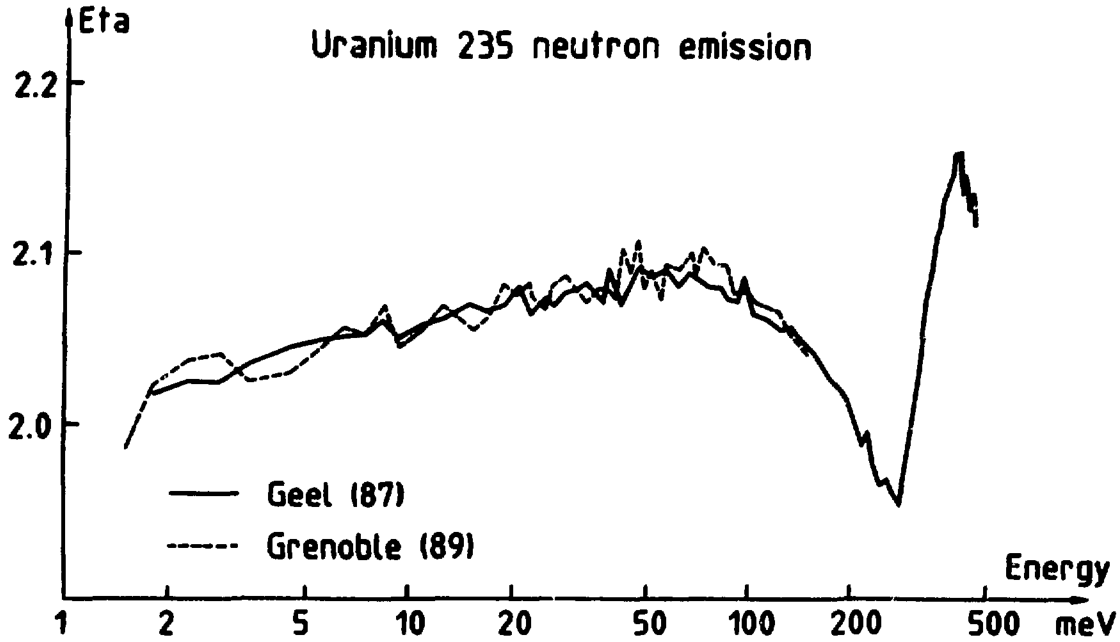

Figure 5 - Experimental results of uranium 235 eta measurements of Geel and Grenoble in the low energy neutron range.

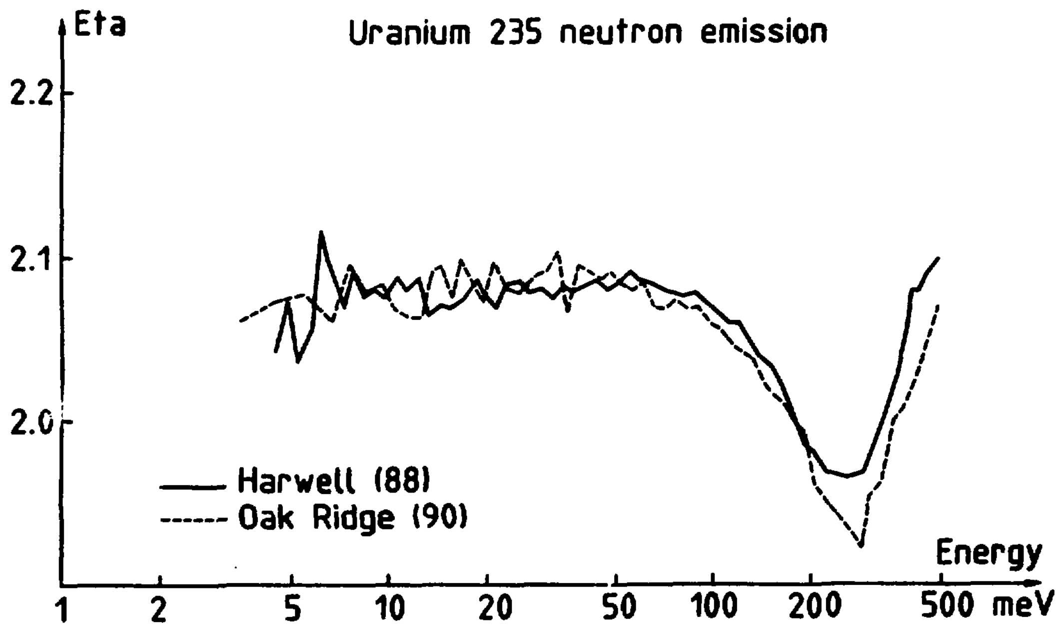

Figure 6 - Experimental results of the uranium 235 eta measurements of Harwell and Oak Ridge below 500 meV.

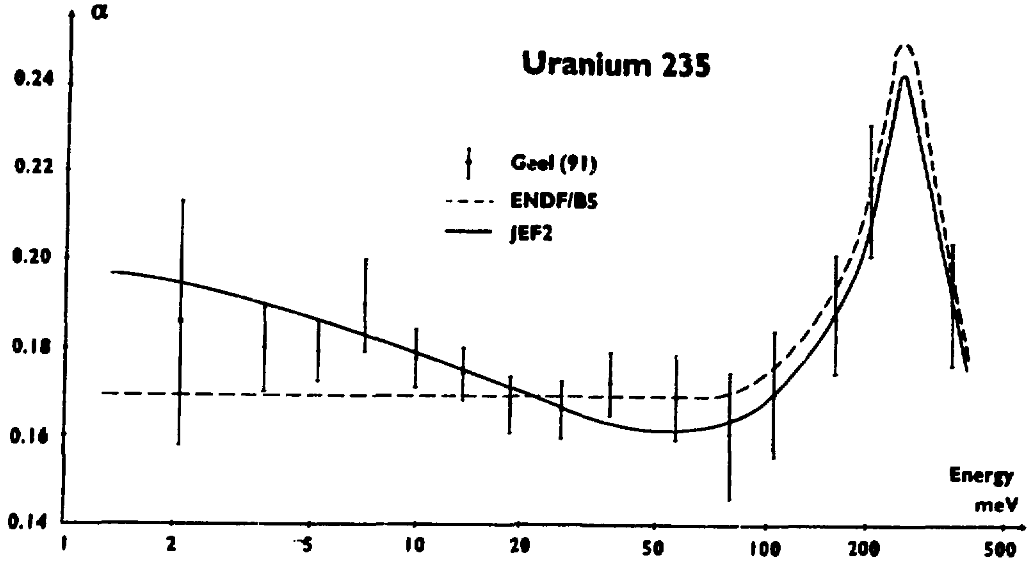

Figure 7 - Comparison of the measurement of the uranium 235 α parameter and the recommended values of ENDF/B5 and JEF2.

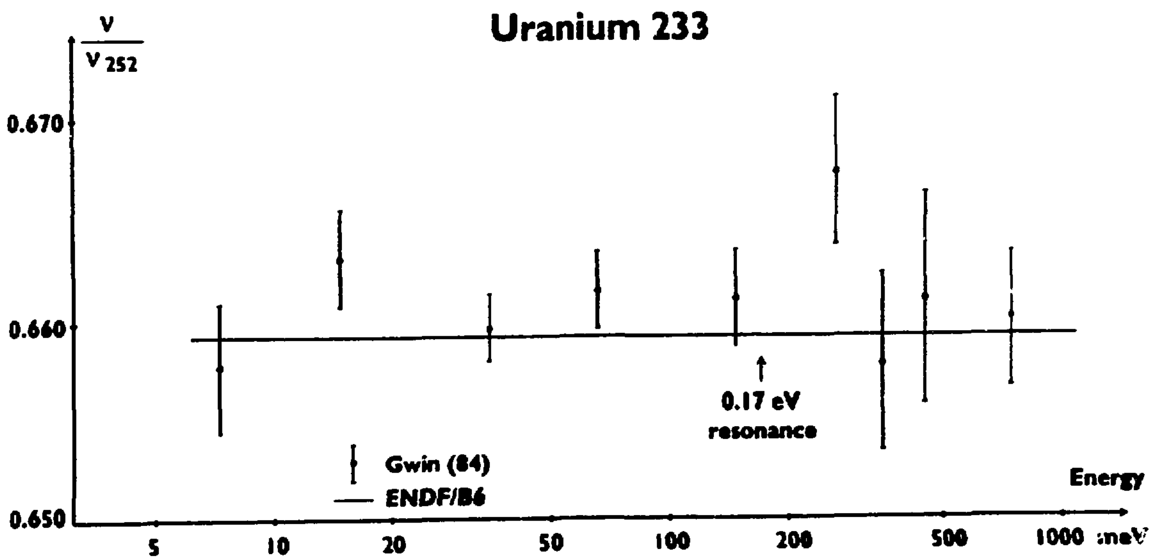

Figure 8 - Ratio of the uranium 233 nu-bar to the one of californium 252 and comparison with the evaluated values below 1 eV.

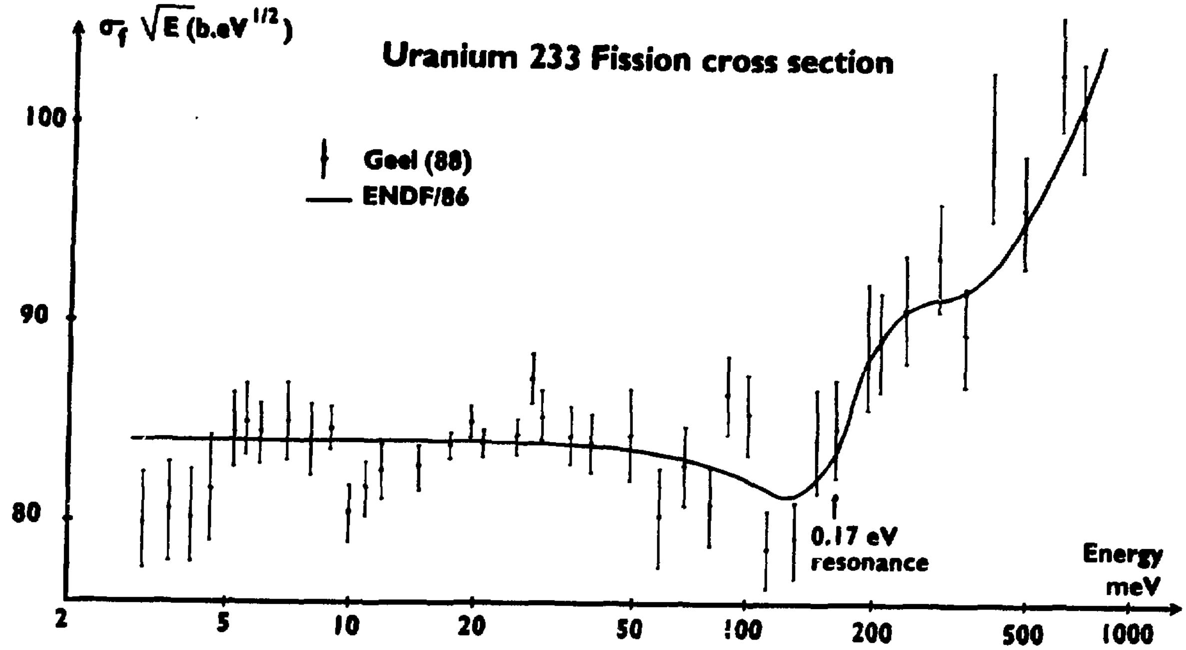

Figure 9 - Experimental fission cross section of uranium 233 and comparison with ENDF/B6.

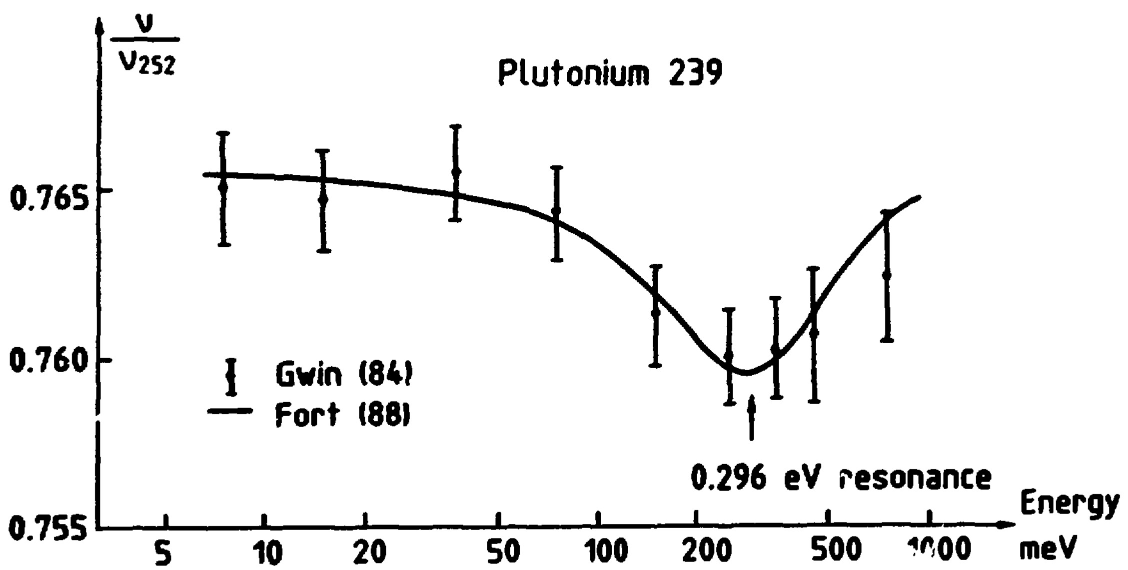

Figure 10 - Ratio of the plutonium 239 nu-bar to the one of californium 252 and comparison with the theoritical calculation of Fort.

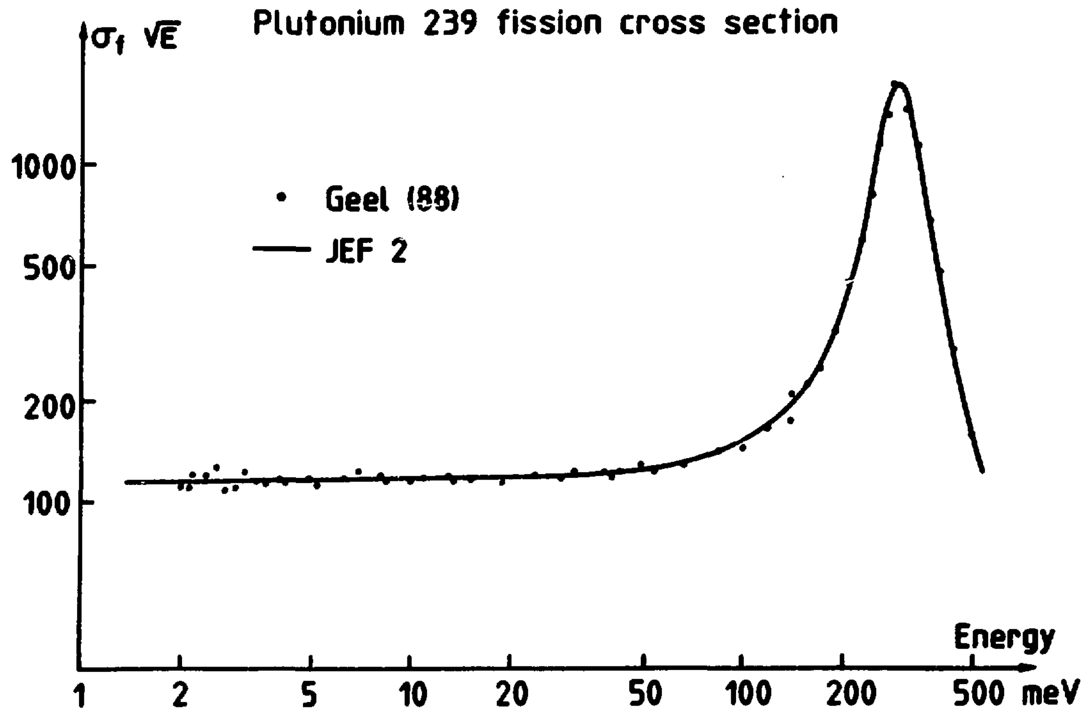

Figure 11 - Comparison of the last fission cross section of Geel and the recommended values of JEF2 for the plutonium 239 below 500 meV.