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**INTERNATIONAL NUCLEAR DATA COMMITTEE**

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THE INDC/NEANDC JOINT DISCREPANCY FILE

1990

Compiled by

B.H. Patrick  
Harwell Laboratory, Didcot, OXON  
United Kingdom

N.P. Kocherov  
IAEA Nuclear Data Section  
Vienna, Austria

June 1990

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IAEA NUCLEAR DATA SECTION, WAGRAMERSTRASSE 5, A-1400 VIENNA



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## FOREWORD

The International Nuclear Data Committee (INDC) and the Nuclear Energy Agency Nuclear Data Committee (NEANDC) maintain, among their activities, a close interest in nuclear data which exhibit discrepancies and, by making known the details of the disagreements, encouragement is given to the undertaking of new measurements as a means of eliminating the ambiguities. From time to time therefore, the Committees publish the current Discrepancy File, the last one being produced by the NEANDC as the NEANDC-INDC Joint Discrepancy File, OECD, Paris 1984.

The committees agreed a few years ago that it was time to publish the latest version of the file and the INDC accepted responsibility on this occasion.

The INDC Sub-Committee on Discrepancies, which met in Beijing during the 16th INDC meeting in October 1987, considered the following topics on the discrepancy list:

- Li-7(n,n't) $\alpha$
- Li-6 production of tritium
- Fe, Cr and Ni capture and total cross-sections
- Nb-93(n,n')
- U-235 resolved resonance parameters
- Pu-239 resolved resonance parameters
- Np-237(n,2n)
- Np-237 nubar
- U-238 capture cross-section
- U-238(n,n')
- Pu-239 fission above the resolved resonances
- Pu-241 resolved resonance parameters
- U-238 and Pu-239 delayed neutron fractions
- Fe-54 radiative width

As a result of the discussions, it was decided to drop the following items from the list:

- Li-7(n,n't) $\alpha$
- Li-6 production of tritium
- U-238(n,n')

Following the meeting, requests were made to responsible reviewers for status reports on the discrepancies still considered to be significant.

The status of reports on discrepancies was once more considered at the 17th meeting of the INDC in Vienna in 1989 and it was decided to allow additional time for the production of reports not yet received.

In the meantime the status of  $^{54}\text{Fe}$  radiative width was analysed by V.G. Pronyaev with the recommendation that it should be excluded from the discrepancy list. Also, H.K. Vonach reported results on a recent  $^{93}\text{Nb}(n,n')$  evaluation at the IRK (Report NEANDC-259/U, p. 309), concluding that the discrepancy no longer existed and the item should be removed from the list.

At the present time, contributions on most of the existing discrepancies have been received, the exceptions being:

Pu-239 fission above resolved resonances  
U-238 and Pu-239 delayed neutron fraction

It was decided to proceed with publication of the discrepancy file in this form to make evaluators and measurers aware of the unsatisfactory status of these items of nuclear data in the hope that this will generate more work to eliminate the differences. Included in the reports are two items which are not in the list of topics above. The first of these is a new item on the half-lives of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{252}\text{Cf}$ , while the second ( $^{239}\text{Pu}$  decay power) was included in the previous publication of the discrepancy file.

The status reports received are published below.

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## The Capture Cross-Section of U-238

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U-238 is the major actinide in power reactor fuel and the capture cross-section is an important quantity for both thermal and fast reactors. The various measured values of shape and the magnitude of the U-238 capture cross-section show discrepancies that are larger than the quoted errors and the evaluations have attempted to minimise these discrepancies.

The neutron energy range can be divided into three regions:

(i) The region below the 6.7 eV resonance

The value of the capture cross-section at thermal energies has been well determined by various techniques. The value given by the ENDF/B Standards Committee is 2.708 barns and the evaluation technique used was described by Peelle and Condé [1] at the Mito Conference. The known resonances can account for 2.353 barns, the remainder has to come from bound levels. Although the magnitude at thermal is known there are very few measurements of its energy dependence. The only accurate shape measurement was carried out at Geel by Corvi et al. [2] and shows a  $1/v$  dependence in the energy region from 0.002 to 0.1 eV. In the energy region from 0.1 to just below the 6.7 eV resonance there are no high quality data on the capture cross-section.

(ii) The resolved resonance region

The discrepancies in the resonance parameters and the capture cross-section in the energy region up to 10 keV have been reduced in the evaluation carried out by Moxon and Sowerby [3] by the adjustment of the normalization and the background. In the energy region below 1 keV it was possible to determine the radiation and neutron widths of most of the s-wave levels. The average radiation width is 23.0 meV and this value was used to determine the neutron widths of all the resonances above 1 keV and the small mainly p-wave resonances below 1 keV which were observed in the high resolution capture cross-section data obtained at Oak Ridge by Macklin et al. [4] but were not seen in the transmission data. The process identified many additional resonances particularly at high energies as many peaks in the observed structure in both the transmission and capture data contain more than one resonance. The analysis leads to an average capture cross-section in broad agreement with the evaluated average capture cross-sections obtained by Poenitz [5] as part of the ENDF/B-VI standards evaluation procedure.

In the region of the resonance peaks the capture cross-section calculated from the evaluated parameters is in good agreement with the data. The shape of the capture cross-section between the resonance is poorly determined from the measurements. The error in the calculated cross-section in these regions due to the omission or interference terms in the calculation of the capture cross-section, and the missing of a few small resonances will add very little to the uncertainty in the infinite dilute cross-section but may contribute to errors in the effective capture cross-section for sample of finite thickness.

(iii) Average cross-section region

The region above 10 keV has recently been evaluated by Poenitz, Fröhner [6] and Kanda et al. [7]. The evaluations of Poenitz and Kanda et al. are simultaneous evaluations which consider a number of cross-sections of different materials. Fröhner's evaluation on the other hand considers all the U-238 partial cross-sections of importance up to 500 keV. Between 10 keV and 300 keV the evaluations of Fröhner and Poenitz are in good agreement but the evaluation of Kanda et al. is significantly higher above 45 keV. The disagreement between the evaluations of Poenitz and Kanda et al. extends as far as the upper limit of the Poenitz data (2.2 MeV) but is less significant above 1 MeV. The evaluations of Fröhner and Poenitz tend to lie below the mean values of the measured cross-sections and the reasons for this need to be understood and justified. The average resonance parameters derived in Fröhner's work differ in detail with those extracted by others from the resolved resonance region and this requires further investigation. Above 2.2 MeV there are few measured data and the evaluations tend to be inaccurate.

The errors in the cross-section are difficult to summarise in a document of this type. The following figures give approximate values:

<u>Energy</u>	<u>Error (%)</u>
0.0253 eV	±0.5
Resonance integral	±1.0
Average capture in resonance region	±3.0
10-1000 keV	±3-5
2 MeV	±3
3 MeV	±6
5 MeV	±20
10-20 MeV	±30

References

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## Status Report

### The $^{10}\text{B}(n,\alpha)$ Cross Section

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Natural boron or  $^{10}\text{B}$ -enriched samples are often used for neutron induced reaction rate measurements or for neutron flux determination. A large variety of detectors is available and the reaction underlying the detection systems is either  $^{10}\text{B}(n,\alpha\gamma)^7\text{Li}$  or  $^{10}\text{B}(n,\alpha)^7\text{Li}$ . Different symbols are used:  $(n,\alpha\gamma)$  is identical with  $(n,\alpha_1)$ ; and  $(n,\alpha)$ , also called total  $(n,\alpha)$ , is equal to the sum of  $(n,\alpha_0)$  and  $(n,\alpha_1)$ . The  $\alpha_0$  refers to an  $\alpha$ -emission with energy  $E_{\alpha} = 1.7891$  MeV, leaving the residual nucleus  $^7\text{Li}$  in its ground state. The  $\alpha_1$  refers to an  $\alpha$ -emission with energy of  $E_{\alpha}=1.4832$  MeV, leaving the residual nucleus  $^7\text{Li}$  in its first excited state, which decays by prompt emission of a gamma ray of 478.5 keV with isotropic angular distribution in the center of mass system. The branching ratio is defined as  $R=\sigma(n,\alpha_1):(\sigma(n,\alpha_0) + \sigma(n,\alpha_1))$ . In the ENDF/B-VI standard file the  $^{10}\text{B}(n,\alpha_0)$  and  $^{10}\text{B}(n,\alpha_1)$  cross sections are considered as standard reference data in the energy range from thermal energy to 250 keV.

In WREND A 83/84 there are seven requests pending for  $^{10}\text{B}(n,\alpha)$  absolute cross section measurements below 250 keV, with accuracies ranging from 3% down to as low as 1%. There are no requests below 250 keV for total- or elastic scattering-cross section data, which is surprising considering the statements made by W.P. Poenitz (4). Requests for cross section measurements of  $^{10}\text{B}(n,\alpha)$ ,  $^{10}\text{B}(n,\alpha_1)$  and for the branching ratio as well, are also made in the high priority request list (1) and (2).

A review of measured cross section data for the neutron interactions with  $^{10}\text{B}$  was made by A.D. Carlson (3) in 1983, and also by W.P. Poenitz (4) in 1984. It is shown that the data base of the  $^{10}\text{B}$  neutron interaction is poor. No absolute measurements of the  $^{10}\text{B}(n,\alpha)$  and the  $^{10}\text{B}(n,\alpha_1)$  cross section were made in the last 15 to 20 years, and even the total cross section needs to be known better. In 1984 W.P. Poenitz concludes and recommends, see ref. (4) p.126, "... that the  $^6\text{Li}(n,\alpha)$  cross section should be used as a reference cross section below 100-150 keV until a reasonable data base for the  $^{10}\text{B} + \text{neutron}$  interaction becomes available".

At the IAEA conference on Nuclear Standard Reference Data, held in Geel, November 1984, W.P. Poenitz presented a paper on: "The simultaneous evaluation of interrelated cross sections by generalized least squares and related data file requirements" (5). The cross sections involved are:  $^6\text{Li}(n,\alpha)$ ,  $^6\text{Li}(n,n)$ ,  $^{10}\text{B}(n,\alpha_0)$ ,  $^{10}\text{B}(n,\alpha_1)$ ,  $^{10}\text{B}(n,n)$ ,  $^{197}\text{Au}(n,g)$ ,  $^{238}\text{U}(n,g)$ ,  $^{235}\text{U}(n,g)$ ,  $^{238}\text{U}(n,f)$  and  $^{239}\text{Pu}(n,f)$ . The experimental data included are:

- \* Absolute measurements or measurements of shapes of cross sections, of sums or of ratios of cross sections,
- \* Absolute measurements or measurements of shapes of ratios of a cross section versus the sum of cross sections,
- \* The integral of the fission cross section of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  over the fission neutron spectrum.

Uncertainties for each experiment, correlations in that experiment, as well as correlations with other experiments are part of the input, and a full covariance analysis is performed.

In 1985 A.D. Carlson et al. (6) presented the initial phase in the development of the new ENDF/B-VI file. This standards evaluation is basically different from earlier ENDF/B evaluations and has three separate parts:

- \* A simultaneous evaluation using generalized least squares program, see W.P. Poenitz (5),
- \* A R-matrix evaluation, by G.M. Hale (7), relying on a large number of reaction channels of the  $^{11}\text{B}$  system including inverse reactions,
- \* A procedure for combining the simultaneous and the R-matrix evaluation (8).

Cross section values for  $^{10}\text{B}(n,\alpha_1)$  and  $^{10}\text{B}(n,\alpha)$  from the simultaneous evaluation are higher than the corresponding values from the R-matrix fit for neutron energies above 50 keV, and these discrepancies are believed to be explained by the poor boron data base as discussed in (3) and (4).

In October 1987 the ENDF/B-VI  $^{10}\text{B}(n,\alpha_1)$  and  $^{10}\text{B}(n,\alpha_0)$  standards cross section data were released. These values are listed in Annex 1. A selection of data points with preliminary uncertainties is also given in Table 1 and 2 for comparison with ENDF/B-V.

At thermal energy only a small change is observed compared to ENDF/B-V, but the value underlying the recent evaluation of thermal constants (9) is neither the ENDF/B-V nor the ENDF/B-VI value. Changes are within quoted uncertainties in previous evaluations and such differences may be expected as the data bases are slightly different. However, the recommendation of even slightly different values for the same quantity is confusing for the data user and should be avoided.

The uncertainty on ENDF/B-VI cross sections of  $^{10}\text{B}(n,\alpha_1)$  is reduced by about a factor of two compared to the values claimed in ENDF/B-V, and sensibly smaller than the changes of the cross section values. For instance at 100 keV the change amounts to 2.8%, whereas the uncertainty is claimed to be 0.7% in ENDF/B-V and as small as 0.44% in ENDF/B-VI. Changes for  $^{10}\text{B}(n,\alpha_0)$  from ENDF/B-V to VI are even larger. The branching ratio remains almost unaltered as expected from the accuracy of the underlying measurements. In column six of Table 1 and 2 are listed the uncertainties in barn, thus illustrating in absolute terms the degree of difficulty to perform a significant new measurement, especially difficult above 10 keV.

Integral measurements of helium production by the  $^{10}\text{B}(n,\alpha)$  reaction in benchmark fields were performed by J.A. Grundl (10) at the NBS and by B.M. Oliver et al. (11) in ZPPR at ANL. S.M. Qaim et al. (12) report measurements made at KFA Jülich to determine the cross section for  $^{10}\text{B}(n,t)2\alpha$  at thermal energy. The most recent measurements of  $^{10}\text{B}(n,\alpha)$  to  $^6\text{Li}(n,\alpha)$  cross section ratios are those of A.D. Carlson (13) at NBS, and those of C. Bastian et al. at the CBNM (14). It is unlikely that the recent measurements mentioned above would change the recommended values of the simultaneous evaluation, but since a simultaneous evaluation starts from a very large data base, all neutron reaction measurements with  $^6\text{Li}$ ,  $^{235}\text{U}$ ,  $^{197}\text{Au}$  and  $^{239}\text{Pu}$  should be screened. This is beyond the scope of this status report on  $^{10}\text{B}(n,\alpha)$ , but other sections will mention some recent related accurate cross section measurements.

The new evaluation procedure underlying the ENDF/B-VI standards file is a very large and successfully coordinated undertaking, that takes profit of all available and related information. It reduces the arbitrariness in evaluating the quality of a specific measurement and it provides a complete covariance matrix. The preliminary uncertainty values are very small and more insight in

the procedure is needed to make them acceptable. Differences between simultaneous evaluations and R-matrix fits should be explained. The new evaluation procedure might be a tool to define those measurements best suited to further reduce uncertainties, possibly total or elastic scattering cross sections of  $^{10}\text{B}$  (4). The forthcoming issue of WREND A 1989 should reflect this.

Table 1: Comparison of  $\sigma(n,\alpha_1)$  data of ENDF/B-V and ENDF/B-VI. Uncertainty of ENDF/B-VI values are preliminary.

Energy	Uncertainty of V (%)	Change V to VI (%)	Value VI (barn)	Uncertainty of VI (%)	Uncertainty of VI (barn)
25.3 meV	0.3	+0.17	3598.23	0.16	5.700
10 eV	0.3	+0.22	180.74	0.16	0.289
100 eV	0.3	+0.33	56.98	0.16	0.091
1 keV	0.3	+0.68	17.875	0.17	0.030
10 keV	0.3	+1.05	5.562	0.18	0.010
50 keV	0.7	+1.10	2.500	0.31	0.008
100 keV	0.7	+2.78	1.842	0.44	0.008
150 keV	0.8	+3.85	1.533	0.55	0.008
200 keV	1.2	+1.97	1.288	0.60	0.008

Table 2: Comparison of  $\sigma(n,\alpha_0)$  data of ENDF/B-V and ENDF/B-VI. Last column is the branching ratio R implicit in the ENDF/B-VI data.

Energy	Uncertainty of V (%)	Change V to VI (%)	Value VI (barn)	Uncertainty of VI (%)	Uncertainty of VI (barn)	Change R (%)
25.3 meV	2.2	-1.20	241.23	0.21	0.506	+0.10
10 eV	2.2	-1.01	12.12	0.21	0.025	+0.10
100 eV	2.2	-1.11	3.819	0.21	0.008	+0.10
1 keV	2.2	-0.98	1.197	0.21	0.003	+0.11
10 keV	2.2	-2.48	0.371	0.30	0.001	+0.22
50 keV	2.0	-3.89	0.1730	0.83	0.001	+0.33
100 keV	1.2	+3.25	0.1431	0.86	0.001	-0.05
150 keV	1.2	+12.3	0.1417	0.84	0.001	-0.61
200 keV	1.6	+15.6	0.1438	0.76	0.001	-1.01

#### List of References

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Annex 1

5-B - 10 NBS EVAL-JUL87 CARLSON, HALE, PELLE, POENITZ  
 LTR TO DUNFORD 8/87 DIST-OCT87 871028  
 ----ENDF/B-VI MATERIAL 525  
 ----INCIDENT NEUTRON DATA  
 ----ENDF-6 FORMAT

\*\*\*\*\*  
 THE STANDARDS SUBCOMMITTEE OF CSEWG RECOMMENDS USING THE  
 (B-10 (N,ALPHA 0) + B-10 (N,ALPHA 1)) AND THE B-10 (N,ALPHA 1)  
 CROSS SECTIONS AS STANDARDS FROM THERMAL ENERGY TO 250 KEV.  
 \*\*\*\*\*

10B(n, a0)

E(ev) s(barn)

2.530000-2	2.412677+2	9.400000+0	1.250020+1	1.500000+2	3.116900+0
2.500000+2	2.410500+0	3.500000+2	2.034700+0	4.500000+2	1.792800+0
5.500000+2	1.619900+0	6.500000+2	1.488900+0	7.500000+2	1.385000+0
8.500000+2	1.300000+0	9.500000+2	1.229000+0	1.500000+3	9.748000-1
2.500000+3	7.519000-1	3.500000+3	6.333000-1	4.500000+3	5.571000-1
5.500000+3	5.028000-1	6.500000+3	4.617000-1	7.500000+3	4.291000-1
8.500000+3	4.025000-1	9.500000+3	3.804000-1	1.500000+4	3.017000-1
2.000000+4	2.613000-1	2.400000+4	2.390000-1	3.000000+4	2.148000-1
4.500000+4	1.796000-1	5.500000+4	1.662000-1	6.492999+4	1.572000-1
7.500000+4	1.510000-1	8.500000+4	1.467000-1	9.500000+4	1.440000-1
1.000000+5	1.431000-1	1.200000+5	1.412000-1	1.500000+5	1.417000-1
1.700000+5	1.426000-1	1.800000+5	1.431000-1	1.900000+5	1.434000-1
2.000000+5	1.438000-1	2.100000+5	1.439000-1	2.200000+5	1.439000-1
2.300000+5	1.439000-1	2.350000+5	1.438000-1	2.400000+5	1.438000-1
2.450000+5	1.437000-1	2.500000+5	1.436000-1		

10E(n, a1)

2.530000-2	3.598228+3	9.400000+0	1.864350+2	1.500000+2	4.650030+1
2.500000+2	3.596710+1	3.500000+2	3.036590+1	4.500000+2	2.675160+1
5.500000+2	2.417560+1	6.500000+2	2.222870+1	7.500000+2	2.068110+1
8.500000+2	1.941300+1	9.500000+2	1.834460+1	1.500000+3	1.456030+1
2.500000+3	1.123520+1	3.500000+3	9.471700+0	4.500000+3	8.335300+0
5.500000+3	7.525400+0	6.500000+3	6.912300+0	7.500000+3	6.426000+0
8.500000+3	6.029600+0	9.500000+3	5.698100+0	1.500000+4	4.519400+0
2.000000+4	3.909000+0	2.400000+4	3.568200+0	3.000000+4	3.195900+0
4.500000+4	2.628500+0	5.500000+4	2.394300+0	6.499799+4	2.220300+0
7.500000+4	2.084400+0	8.500000+4	1.974500+0	9.500000+4	1.883500+0
1.000000+5	1.842000+0	1.200000+5	1.703100+0	1.500000+5	1.533000+0
1.700000+5	1.430300+0	1.800000+5	1.381700+0	1.900000+5	1.334000+0
2.000000+5	1.287600+0	2.100000+5	1.241700+0	2.200000+5	1.197200+0
2.300000+5	1.154300+0	2.350000+5	1.133800+0	2.400000+5	1.112700+0
2.450000+5	1.092800+0	2.500000+5	1.073200+0		





## Capture Cross Sections for Cr, Fe and Ni

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Commission of the European Communities

Joint Research Centre

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### Significance

Since stainless steel represents about 25% of the volume of a fast power reactor, its constituent elements strongly influence its two main neutronic parameters: critical enrichment and breeding gain. Also, capture in the narrow p and d-wave resonances of Cr, Fe and Ni contributes as much as 10 to 15% of the Doppler coefficient of reactivity. Following sensitivity calculations, typical accuracy requirements<sup>1</sup> in the energy range 0.1-100 keV are 5-10% for capture in Fe and 10-20% for capture in Ni and Cr.

### The findings of the 1.15 keV $^{56}\text{Fe}$ task force

Since a number of years, a systematic difference of about 20% was noticed<sup>2,3</sup> between the results of capture and those of transmission for the neutron width of the important 1.15 keV resonance of  $^{56}\text{Fe}$ . Such a discrepancy has become the object of a task force set up by NEANDC after the Antwerp Conference. Although this task force has not yet completed his works, a major progress has been obtained recently and will be summarized in the following.

The origin of the discrepancy lies in the fact that the neutron capture measurement techniques used in the past and based on the pulse height weighting method were unable to deal correctly with such large differences in spectrum shape as those met when comparing capture in  $^{56}\text{Fe}$  to capture in the elements used for normalization such as Ag and Au. Basically, the method failed because the weighting function was calculated with ad hoc Monte Carlo codes which were entirely inadequate since they lacked an accurate description of the electron transport and of the influence of the environment. On the contrary, improved weighting functions have been obtained recently both with an original experimental method<sup>4</sup> and with a state-of-the-art electron-gamma transport code<sup>5</sup>: although some differences still persist between these two approaches, the use of such functions allows to obtain parameters of the 1.15 keV resonance in agreement with the transmission results, therefore eliminating the discrepancy.

### Impact on structural materials

The results reported in the previous section are not limited to  $^{56}\text{Fe}$  alone but put in question all past measurements performed with the pulse height weighting method, every time different capture spectrum shapes were compared. This applies to structural materials in general as it may be seen in Table 1 : here the sums of the intensities of thermal neutron capture  $\gamma$ -rays with energies higher than 6 MeV are listed for the three main isotopes of Cr, Fe and Ni and compared to similar values for the normalizing nuclides  $^{109}\text{Ag}$  and  $^{197}\text{Au}$ . One may notice that, for all structural isotopes, this quantity is comparable to  $^{56}\text{Fe}$  capture while being definitely larger than in Ag or Au. The same picture should hold also for p-wave capture since, in this mass region, E1 and M1 average strengths are comparable<sup>6</sup>.

The systematic error due to a wrong weighting and associated to a given resonance can be seen as made up of two parts : the first is related to the difference between the  $\gamma$ -spectrum of the normalizing element (Ag or Au) and the average spectrum of the given isotope. The second is due to variations in the spectrum shape amongst the resonances of this isotope, due to Porter-Thomas fluctuations of the intensities of primary transitions. The first error, which is very likely the predominant one, was avoided when capture data were normalized to a low energy resonance belonging to the same isotope, whose parameters were obtained from transmission <sup>2,7,8</sup>. In fact, besides the 1.15 keV resonance, there are other cases, listed in Table 2, in which the capture area can be determined from transmission data. As far as the second effect is concerned it certainly plays an important role <sup>2,10</sup> particularly in  $^{54}\text{Fe}$  and  $^{56}\text{Fe}$ .

### Recommandations and plans for the future

Although it is clear that the whole field of structural material capture should be revisited, this does not necessarily lead to the repetition of all measurements. First of all, it is possible to re-analyse past experiments with the correct weighting as long as the amplitude information has been preserved : this is for example the case for the Geel  $^{56}\text{Fe}$  data<sup>2</sup>. Secondly, there are cases (the  $E_0 = 1.63$  keV resonances of  $^{52}\text{Cr}$  and  $^{57}\text{Fe}$ ) in which no capture-to-transmission discrepancy has been detected within the limits of the experimental uncertainties <sup>7,8</sup>. To check the data for these isotopes it is suggested to repeat the measurements with the correct weighting only for a few outstanding low energy resonances. In parallel, some transmission measurements of the resonances

of Table 2 should be performed in order to check and possibly improve the precision of the parameters. Only after comparison of the new with the old results one should decide whether it is worthwhile to repeat the whole measurements.

Finally, as far as Ni is concerned, a complete programme of capture measurements of the three main even isotopes is due to start afresh in Geel in the following months.

Table 1 : Thermal neutron capture data

Target	Abund. %	$\sum I_{\gamma}^a)$ ( $E_{\gamma} > 6$ MeV)	Target	Abund. %	$\sum I_{\gamma}^a)$ ( $E_{\gamma} > 6$ MeV)
50Cr	4.35	71	58Ni	68.27	62
52Cr	83.79	70	60Ni	26.10	62
53Cr	9.50	83	62Ni	3.59	92
54Fe	5.8	88	109Ag	48.17	5.4
56Fe	91.72	72	197Au	100	18.9
57Fe	2.15	52			

a) Intensities in photons per 100 captures from Nuclear Data Sheets

Table 2 : Resonances suitable for normalization to transmission data

Target	$E_0$ (keV)	J	l	$g \Gamma_n$ (eV)	$\Gamma_{\gamma}$ (eV)	Ref.
52Cr	1.63	3/2	1	$0.0624 \pm 0.0020$	$0.67 \pm 0.14$	8
56Fe	1.15	1/2	1	$0.0617 \pm 0.0009$	$0.574 \pm 0.040$	9
57Fe	1.63	2	1	$0.0533 \pm 0.0022$	$1.00 \pm 0.24$	7
60Ni	2.25		1	$0.053 \pm 0.001$	$1.20 \pm 0.08$	11

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## CHROMIUM AND NICKEL INELASTIC SCATTERING "DISCREPANCIES"

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As far as we know, there are no "integral measurements" to check the quality of inelastic scattering data for nickel and chromium. (For iron, such integral data exist in terms of iron spheres). Stainless steel integral data are dominated by effects of iron, so are not very useful for the purpose at hand. The Livermore 14 MeV pulsed sphere measurements are sensitive to the energy and angular distributions of both discrete and continuum inelastic scattering, so would be ideal tests if the data existed. In lieu of integral information, the problem must be addressed by looking at the quality of available differential data. With most evaluated libraries going to isotopic evaluations, the importance of isotopic differential data has increased.

For  $^{58}\text{Ni}$ , data is available mainly for energies below 5 MeV, with a few data sets recently available (TUNL) from 8-17 MeV. The situation for  $^{60}\text{Ni}$  is similar. Guidance for the total inelastic scattering cross section is available from gamma-ray production measurements to 40 MeV (ORNL). For both of these isotopes, discrepancies larger than the uncertainties often exist among the various measurements. For the minor isotopes ( $^{61}, ^{62}, ^{64}\text{Ni}$ ) differential inelastic scattering data is generally sparse. Differential data for inelastic scattering from the natural materials is more plentiful, but also suffers from disagreements larger than the quoted uncertainties. There are several neutron emission cross section measurements for natural Ni, but all are at an incident energy of 14 MeV. Some data sets provide detailed angular distributions for each outgoing energy spectra.

For  $^{52}\text{Cr}$ , the two lowest lying levels have data to 8 MeV, but data for higher lying levels generally only goes to about 4 MeV. There is one measurement at 14 MeV for the two low lying levels. The total inelastic scattering cross section is available to 40 MeV, as for  $^{58}, ^{60}\text{Ni}$ . Data for the other isotopes ( $^{50}, ^{53}, ^{54}\text{Cr}$ ) is available only up to about 4 MeV. As for natural Ni, neutron emission cross section data is available only at 14 MeV. Comparison of the various inelastic scattering data sets for chromium isotopes shows that they often disagree by more than their quoted uncertainties.

For the region from 8-14 MeV, important to fusion energy technologies, the data base for the isotopes of nickel and chromium is inadequate. More differential data (including neutron emission data) is needed in this region to improve the evaluations and reduce the uncertainties for these materials. For fast reactor studies, high resolution inelastic scattering data in the resonance region (and just above, as exists for iron) would remove the unphysical, smooth cross sections.



## STATUS OF INTEGRAL AND DIFFERENTIAL CONSISTENCY FOR $Fe(n,n')$

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In June 1986 a test file for iron, based on ENDF/B-V Revision 2, was generated at Oak Ridge National Laboratory. The purpose was to remove several outstanding discrepancies between integral data and calculations using ENDF/B-V iron:

1. For a 14-MeV source, it was known for a long time that inelastically scattered neutrons are forward-peaked in the continuum (threshold = 4.5 MeV). Forward-peaked neutrons are more penetrating, hence the isotropic assumption for the continuum for ENDF/B-V iron is no doubt a source of error for thick-shield analysis involving fusion sources (UI pulse sphere, Rich Johnson, Washington Conference, 1975).

2. For a fission source, three analyses of integral measurements consistently indicated that the  $Fe(n,n')$  cross section in ENDF/B-V was too large. The three problems all involve ORNL personnel - (1) Dick Maerker: pressure vessel surveillance for several existing power reactors, (2) Joe Pace: source terms for the Hiroshima and Nagasaki dosimetry reevaluation, and (3) Charles Slater: Tower Shielding Facility deep penetration measurements. Maerker specifically suggested that the ENDF/B-V  $Fe(n,n')$  was 8% too large between 3 and 8 MeV.

New  $Fe(n,n'\gamma)$  data for the 846-keV level (Duane Larson) and the improved TNG code (probably the most reliable code for angular distributions in the 5- to 15-MeV range) were timely for addressing the above problems and generating the test file. This file was processed into the Vitamin-E 174-neutron and 38-gamma group structures and reapplied to all three problems in item (2) above, all showing large improvements (Maerker: ORNL/TM-10389, 1987; Pace: private communication, 1987; Slater: Jasper workshop, 1987). The UI pulse sphere problem was also reanalyzed with much improved consistency (Fu, 1986 ANS winter conference). Smaller discrepancies remain, but they are more difficult to resolve. For example, Maerker suggested that the  $Fe(n,n')$  cross section below 3 MeV is still a few percent too large but we don't have any new experimental data or theoretical means to do anything about it. Perhaps a remeasurement of Kinney and Perey's high resolution  $Fe(n,n')$  cross section below 2.2 MeV is worthwhile, preferably extending the energy to 3.5 MeV.

The ENDF/B-VI iron evaluation retains the basic features of the above test file, with some improvements. For instance, the new File-6 format for energy-angle correlated distributions and the separated isotope files allow more accurate representation of the evaluated results.





# Discrepancies in the Half-Lives of $^{90}\text{Sr}$ , $^{137}\text{Cs}$ and $^{252}\text{Cf}$

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## INTRODUCTION

Recent reports [1,2,3,4] have pointed out that significant discrepancies exist in published half-life data for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{252}\text{Cf}$ , amongst others. These discrepancies make the estimation of evaluated half-lives for these isotopes difficult and contentious.

This paper reviews the current situation, takes into account recently available data and attempts to derive recommended half-lives and associated uncertainties for the three isotopes.

### $^{90}\text{Sr}$

1. Data: Measurements of half-lives of  $^{90}\text{Sr}$  reported since 1955 are listed in Table 1. Under the heading of technique, SA refers to a specific activity measurement and DD refers to a direct decay measurement, the number being the number of half-lives over which the decay was measured.
2. Evaluation: The weighted mean of the nine values shown in table 1 is  $28.48 \pm 0.03\text{y}$  with a total chi-squared of 238 for eight degrees of freedom indicating a high degree of inconsistency. Two values in particular (Lagoutine et al. [9] and Martin [12]) contribute together 91% of the total chi-squared. One can only conclude that one or both of these measurements have associated uncertainties that have been seriously under-estimated. In order to arrive at an evaluated half-life that is not dominated by these two discrepant values their associated uncertainties have each been increased to 0.2 years. The new weighted mean value then becomes  $28.67 \pm 0.10\text{y}$  with a chi-squared of 26, of which now only 32% is contributed by the two measurements discussed above. The adjusted data set still remains inconsistent, and to reduce chi-squared to its expected value, 8, it is necessary to inflate

all uncertainties by a factor of 1.82 to obtain the final recommended value:-

$$28.67 \pm 0.18 \text{ y}$$
$$\text{or } 10473 \pm 66 \text{ d}$$

This value is almost identical to that ( $28.7 \pm 0.2\text{y}$ ) in the 1985 evaluation [1] despite the slightly different data set.

### $^{137}\text{Cs}$

1. Data Measurements of the half-life of  $^{137}\text{Cs}$  reported since 1955 are listed in Table 2. The designations SA and DD in the 'Technique' column have the same meanings as in Table 1. BI indicates a technique based on the measurement of the growth of  $^{137}\text{Ba}$  by mass spectrometry.
2. Evaluation The weighted mean of the eighteen values shown in Table 2 is  $30.24 \pm 0.01$  with a chi-squared of 783 for seventeen degrees of freedom indicating, as in the  $^{90}\text{Sr}$  case, a very high degree of inconsistency. 62% of the total chi-squared arises from one value (Hoppes & Schima 1982) but there are also major contributions to chi-squared from other values with quoted uncertainties less than 0.1 year. It is again clear that, in many cases, uncertainties have been seriously underestimated. In order to arrive at a recommended value, all uncertainties quoted as less than 0.1 year have been increased to 0.1 year. As shown in Table 2 this leads to a more even chi-squared distribution and to a mean value of  $30.11 \pm 0.03\text{y}$  with a total chi-squared of 114 for seventeen degrees of freedom. To remove the remaining inconsistency and to reduce chi-squared to its expected value it is necessary to inflate all the uncertainties by a factor of 2.6 leading to the final recommended value

$$30.11 \pm 0.08 \text{ y}$$
$$\text{or } 10,999 \pm 31 \text{ d}$$

Which differs only marginally from the previous evaluation ( $30.13 \pm 0.09$  years) [2], but is slightly lower than the IAEA-CRP

recommended value ( $11050 \pm 40$  days) [4], which ignores data more than 20 years old.

### $^{252}\text{Cf}$

Half-life data for  $^{252}\text{Cf}$  have recently been reviewed by Smith [3,29] who gave a recommended value of  $2.645 \pm 0.008$  years based on a subjective analysis of the available data.

1. Data The data shown in Table 3 are taken from Smith [3].
2. Evaluation The weighted mean of the data shown in Table 3 is  $2.6504 \pm 0.0008$  with a chi-squared of 49 for eight degrees of freedom. Half of the total chi-squared arises from one value (De Volpi & Porges [31]) which can be regarded as an outlier. Rejection of this value leads to a new weighted mean of  $2.6510 \pm 0.0008$  with a chi-squared of 25 for seven degrees of freedom. To reduce chi-squared to its expected value it is necessary to inflate all the uncertainties by a factor of 1.9, leading to a final recommended value of:-

$$2.651 \pm 0.002 \text{ years}$$
$$\text{or } 968.3 \pm 0.6 \text{ days.}$$

TABLE 1

<sup>90</sup>Sr Half - Life Data

<u>Source</u>	<u>Technique</u>	<u>Half-Life (Years)</u>	<u>Uncertainty (years)</u>	<u>Chi-Squared Contribution</u>	<u>Adjusted Uncertainty</u>	<u>Adjusted Chi-Squared</u>
Wiles & Tomlinson 1955 [5]	SA	27.7	0.4	3.8		5.9
Anikina et al. 1958 [6]	SA	29.3	1.6	0.3		0.2
Flynn et al. 1965 [7]	DD(0.37)	28.0	0.4	1.5		2.8
Flynn et al. 1965 [7]	SA	28.5	0.9	0.0		0.0
Hoppes 1977 [8]	DD(0.58)	29.12	0.24	7.0		3.4
Lagoutine et al. 1978 [9]	DD(0.2)	28.15	0.034	96.7	0.2	6.9
Ramthun 1983 [10]	SA	28.99	0.25	4.1		1.6
Schrader, 1987 [11]	DD(0.17)	29.58	0.45	5.9		4.0
Martin, 1987 [12]	DD(0.75)	28.92	0.04	<u>118.6</u>	0.2	<u>1.5</u>
				<u>237.9</u>		<u>26.4</u>

TABLE 2

<sup>137</sup>Cs Half-Life Data

<u>Source</u>	<u>Technique</u>	<u>Half-Life (years)</u>	<u>Uncertainty (years)</u>	<u>Chi-squared Contribution</u>	<u>Adjusted Uncertainties</u>	<u>Adjusted Chi-squared</u>
Brown et al. 1955 [13]	SA	30.0	0.4	0.4		0.1
Farrar et al. 1961 [14]	BI	30.4	0.4	0.2		0.5
Cook 1962 [15]	SA	29.40	0.18	21.7		15.6
Fleishman et al. 1962 [16]	SA	30.1	0.7	0.0		0.0
Gorbics et al. 1963 [17]	DD(0.1)	29.68	0.05	124.9	0.1	18.6
Rider et al. 1963 [18]	BI	29.2	0.3	12.0		9.2
Lewis et al. 1965 [19]	SA	30.72	0.13	13.7		21.9
Flynn et al. 1965 [7]	DD(0.28)	29.9	0.5	0.5		0.2
Flynn et al. 1965 [7]	SA	30.9	0.7	0.9		1.3
Harbottle 1970 [20]	DD(0.33)	30.64	0.43	0.9		1.5
Walz & Weiss 1970 [21]	DD(0.11)	29.901	0.045	56.4	0.1	4.4
Emery et al. 1972 [22]	DD(0.35)	30.18	0.10	0.3		0.5
Dietz & Pachucki 1973 [23]	DD(0.36)	30.174	0.011	34.8	0.1	0.4
Corbett 1973 [24]	DD(0.22)	30.21	0.08	0.1	0.1	1.0
Gries & Steyn 1978 [25]	SA	29.86	0.09	17.7	0.1	6.3
Houtermans et al. 1980 [26]	DD(0.25)	30.142	0.030	10.4	0.1	0.1
Hoppes & Schima 1982 [27]	DD	30.68	0.02	486.5	0.1	32.3
Martin 1987 [28]	DD(0.20)	29.94	0.27	1.2		0.4
				<u>782.6</u>		<u>114.3</u>

Table 3  
 $^{252}\text{Cf}$  Half-Life Data

<u>Source</u>	<u>Half-Life</u> <u>(years)</u>	<u>Uncertainty</u> <u>(years)</u>	<u>Chi-squared</u> <u>Contribution</u>	<u>Chi-squared</u> <u>Contribution</u>
Metta et al. 1965 [30]	2.646	0.004	1.2	1.6
Devolpi & Porges 1969 [31]	2.621	0.006	24.1	rejected
Mijnheer & Vanden Houten-Zuidema 1973 [32]	2.659	0.010	0.7	0.6
Shchebolev et al. 1974 [33]	2.628	0.010	5.0	5.3
Mozhaev 1976 [34]	2.637	0.005	7.2	7.8
Alberts 1980 [35]	2.648	0.002	1.5	2.2
Spiegel 1980 [35]	2.653	0.001	6.5	4.0
Lagoutine & Legrand 1982 [36]	2.639	0.007	2.7	2.9
Smith 1984 [37]	2.651	0.003	0.0	0.0
			<u>49.0</u>	<u>24.5</u>

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# STATUS OF THE $^{235}\text{U}$ , $^{239}\text{Pu}$ and $^{241}\text{Pu}$ RESONANCE PARAMETERS

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## INTRODUCTION

Before 1978, the year of the introduction of the  $^{235}\text{U}$  and  $^{239}\text{Pu}$  resonance parameters in the INDC-NEANDC discrepancy file, the situation was indeed very bad in the most important evaluated data files. Due to data processing difficulties the evaluators were not able to use the best set of resonance parameters with the best formalism. The problems were clearly defined at the 1981 IAEA conference on U and Pu isotope resonance parameters <sup>(1)</sup>. Several requests were formulated for new measurements, new evaluations and use of the best formalism available for cross-section calculations in the data processing codes. During the years 1981 to 1988 most of these requests were fulfilled. New high resolution transmission measurements and fission measurements were performed at ORELA (2, 3, 4, 11) with very low experimental background. At the same time, improvements in the data processing codes were obtained (5, 6) for group cross-section calculations from the evaluated data files, allowing the use of the REICH-MOORE parameters with a reasonably fast DOPPLER broadening calculation. The REICH-MOORE codes for resonance analysis of the experimental data were also improved by using a generalized least square method based on the BAYES theorem, with correlation matrices for a consistent analysis of several experimental data (7). The energy range of the analysis was extended, taking advantage of a better knowledge of the shape of the resolution function for an accurate determination of the parameters in the high energy ranges (8). As a result of all these works, the new  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  resonance parameter files are of much better quality than those used in all the previous evaluated data files.

The comparison between several sets of resonance parameters proposed for the description of a fissile nucleus cross-sections is not straightforward. The properties and the quality of each set depend on the formalism used, the way of considering the

effect of the missed resonances, the number and the nature of the experimental data analysed, the energy range considered. The best set of resonance parameters should be obtained by a consistent analysis of all the available experimental data taking into account the most important experimental effects such as the accurate shape of the resolution function, the normalization and the background corrections which are mainly responsible for the discrepancies among the measured cross-sections. The best computer code for the analysis should use the generalized least square method using the BAYES equations, in such a way that a covariance matrix is obtained for the resonance parameters and the parameters describing the experimental effect corrections of the experimental data considered. The REICH-MOORE ORNL code SAMMY (7) has been developed for this kind of analysis and important results have been obtained for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  resonances parameters at ORNL.

### $^{239}\text{Pu}$

The last review of the status of Nuclear Data for  $^{239}\text{Pu}$  resonance region is found in the Proceedings of the Santa Fe international conference (19), with an extensive study of the resonance discrepancy problems. At the same time an important resonance analysis work was started at ORNL. The analysis (10) was performed on the recent ORELA measurements : fission cross-section measurements performed in 1984 by GWIN et al in the energy range 0.01 eV to 60 eV (4) and by WESTON et al. in the energy range 10 eV to 110 keV (3) ; total cross-section measurements by SPENCER et al. in 1986 over the thermal and first resonance energy range (11) ; total cross-section measurements by HARVEY et al. in 1984 (2) in the energy range 1 eV to several keV. HARVEY data were obtained from the transmission of 4 sample thicknesses cooled down at liquid nitrogen temperature. All these data were analyzed by the REICH-MOORE code SAMMY along with some older data, particularly the RPI-ORNL fission, capture and absorption data taken since 1970 by GWIN et al. in the thermal energy range (39), and WAGEMANS fission data (12) and BLONS fission data (13) in the resolved energy range. A set of resonance parameters has been obtained for the calculation of the cross-sections in the energy range thermal to 1 keV. The so-called

ENDF/B file 3 is not needed since the contribution of the external range was taken into account by 4 bound levels and 3 fictitious resonance in the energy range above 1 keV. The data have been proposed for JEF2 and ENDB/B-VI evaluated data files.

The new set of resonance parameters provides some answers to the problems which were reviewed in the Santa Fe paper :

1) The first recommendation was for a new measurement of the total cross-section in the thermal energy range. The measurement was done by SPENCER et al. (11) who obtained a 2200 m/s value with 0.6 % accuracy. SPENCER data were included in the ENDF/B-VI standard evaluation data base ; the thermal cross-sections obtained by the standard group are shown on Table I (14). After renormalization on the new standard, a SAMMY fit was performed on SPENCER transmissions and on all the fission, absorption and capture obtained by GWIN et al. since 1970, in the energy range 0.01 eV to 1 eV, showing the consistency of all the RPI-ORELA data. The values obtained from the fit at 2200 m/s are shown on Table I.

2) The spin of the 0.3 eV resonance is definitively  $1^+$  and the first negative resonance at - 0.15 eV has been assigned the spin  $0^+$ . That constitutes a new basis for the interpretation of  $E_k$  and  $v_p$  measurements in the thermal range.

3) The new parameters were compared to those obtained at SACLAY (15) in the energy range up to 660 eV from a multilevel BREIT-WIGNER fit of SACLAY transmissions and fission data. The agreement is very good for the narrow resonances (mainly  $1^+$  resonances) in which the interference effects due to the  $1^+$  fission channel are very small. The energy, the neutron width and the fission width of the broad resonances ( $0^+$  resonances) are generally different ; the new set of parameters is obviously more accurate for the reason that the multilevel BREIT-WIGNER formalism used in the old SACLAY analysis was not able to fit the strong interference effects due to the  $0^+$  open fission channel. The new value of the strength function in the energy range 0 eV to 600 eV is  $1.29 \times 10^{-4}$  compared to  $1.26 \times 10^{-4}$  for the old SACLAY data. If one considers only the narrow resonances ( $1^+$  resonances), the strength function is  $0.915 \times 10^{-4}$  from the new data and  $0.916 \times 10^{-4}$  from SACLAY. In view of this very good agreement, the 10 % discrepancy found between the average neutron width values from DUBNA and SACLAY (16) should be disregarded.

TABLE I - 2200 m/s VALUES OF CROSS-SECTIONS AT 300 K (barns)

	235U		239Pu		241Pu	
	Calculated value	Proposed standard	Calculated value	Proposed standard	Calculated value	Proposed standard
Total	696.92	698.67 + 1.71	1026.88	1027.30 + 5.00	1386.30	1386.14 + 8.64
Scattering	15.09	15.46 + 1.06	8.85	7.88 + 0.97	11.16	12.17 + 2.62
Absorption	681.83	683.22 + 1.34	1017.83	1019.42 + 4.00	1375.14	1373.97 + 8.23
Fission	582.93	584.25 + 1.11	747.34	747.99 + 1.87	1012.13	1012.68 + 6.58
Capture	98.90	98.96 + 0.74	270.49	271.43 + 2.14	363.01	361.29 + 4.95

4) There is no change for the average s wave level spacing value of 2.40 eV obtained in the energy range 0 eV to 450 eV where it is expected that only few resonances could be missed. However, following KEYWORTH et al. suggestion (17), the statistical tests applied to the neutron width distribution show that an important number of resonances is still missing ; the true value of the s wave level spacing could be between 1.90 eV and 2.00 eV, corresponding to at least 20 % of missed levels. Since the resolution (both SACLAY and ORNL) and the counting rate (SACLAY) of the transmission measurements were of very high quality, a doubt remains on the validity of the statistical tests applied to the  $^{239}\text{Pu}$  observed neutron width distribution.

5) The inconsistency between the measured total cross-section and the measured fission cross-section over the broad resonances, observed by SIMPSON et al. (18) when evaluating the data for ENDF/B-III and IV, has been explained. It was mainly due to a rather large remaining background in the BLONS fission data. The ORNL analysis of HARVEY transmissions and WESTON fission is consistent.

6) The most important point is the 3 % discrepancy between WESTON 84 fission data and the values proposed by the ENDF/B-VI standard evaluation group in the energy range 100 eV to 1000 eV. Since the new set of resonance parameters reproduces WESTON data without normalization and background corrections, the same discrepancy exists between the calculated cross-sections and the proposed standard, as shown on Table II (19). The authors of ORNL analyses believe that most of the experimental fission data suffers of an under-evaluation of the experimental background, particularly BLONS data. Then, the corresponding systematic errors in the data base used by the standard evaluation group should not be randomly distributed with a mean value of 0. As a consequence, the proposed standard values could be wrong by as much as + 3 % in the energy range concerned. The ORNL SAMMY fits were performed on WESTON 84 and BLONS 73 fission cross-sections with the result that no background correction was needed in WESTON data and that a large background correction was needed in BLONS data. The next step could be to include more experimental fission data set in the SAMMY fits above 100 eV. But, data with a resolution good enough for a resonance analysis in the high energy range are scarce. At least, the new 1988 high resolution

TABLE 2

 $^{239}\text{Pu}$  AVERAGE FISSION CROSS-SECTIONS (barns)

Energy (KeV)	This work (17)	Weston and Todd (4)	Blons (20)	Proposed standard for ENDF/B-VI (21)
0.1 - 0.2	18.135 (2.9%)	18.095	18.93	18.66 + 0.13
0.2 - 0.3	17.311 (3.3%)	17.441	17.79	17.88 + 0.12
0.3 - 0.4	8.080 (4.3%)	8.130	8.91	8.43 + 0.06
0.4 - 0.5	9.389 (1.9%)	9.337	9.71	9.57 + 0.07
0.5 - 0.6	15.062 (3.3%)	15.170	15.51	15.56 + 0.11
0.6 - 0.7	4.129 (8.0%)	4.192	4.63	4.46 + 0.04
0.7 - 0.8	5.323 (5.8%)	5.385	5.94	5.63 + 0.04
0.8 - 0.9	4.729 (5.3%)	4.765	5.11	4.98 + 0.04
0.9 - 1.0	8.228 (1.0%)	8.165	8.57	8.30 + 0.07
0.1- 1.0	9.04 (3.4%)	9.07 (3.1%)	9.51	9.35

The figures between parenthesis represent the percentage deviation with the proposed standard values.

fission measurement by WESTON et al (20) should be included in the SAMMY analysis.

To conclude, on the  $^{239}\text{Pu}$  resonance parameters, the situation has greatly improved. A complete REICH-MOORE multichannel-multi-level set of parameters is available up to 1 keV and should meet the requests in the resolved resonance region. Nevertheless, some points could be reinvestigated in a possible future work, particularly the values of the fission cross-sections above 100 eV, in relation with the analysis of the new WESTON et al fission measurement. The problem of the covariance matrices issued from SAMMY has not been mentioned above. The full covariance matrix for the 393 resonances in the entire energy range analyzed was not obtained. Only the 100 eV range correlation matrices are available. But the method in introducing such large matrices in the evaluated data files has not been clearly defined.

It was recommended in the "INDC Discrepancy file 1979" (21) that new multilevel-multichannel analysis should be performed on the available total and fission experimental data with the new informations contained in the KEYWORTH et al. spin separated fission cross-sections (22). Preliminary results were presented by MOORE et al. at the Vienna 1981 meeting (23). In "Nuclear Data Discrepancies 1983" it was stressed that the definitive analysis should include total and capture data in view of obtaining an unique consistent set of neutron, fission and capture width for each spin state. In 1984-1987 new experimental data were obtained at ORELA : fission cross-sections with very low experimental background by GWIN et al. (4) and by WESTON et al. (3), total cross-sections by SPENCER et al. (11) in the thermal region and by HARVEY et al. (2) in the resolved energy range. HARVEY data were obtained from transmissions of samples cooled down at liquid nitrogen temperature on a 80 m flight path. With these new high quality experimental data it was possible to start in 1986 a consistent SAMMY analysis for the determination of the best multilevel-multichannel set of resonance parameters extended on a larger energy range. The work is in progress at ORNL by DE SAUSSURE et al. and should be completed in fall 1988 to be available for ENDF/B-VI and JEF2. Preliminary results are available (24). In the energy range thermal to 110 eV, 262 resonances are used for the calculation of the cross-sections ; in this energy range, where there is probably less than 20 % of missed levels, the resonance parameters are physically meaningful. Above 110 eV the resonances become more and more unresolved and the analysis must be considered as a parametrization which describes the available data fairly well but which should not be used to infer the average properties of the resonances ; this parametrization provides a more accurate representation of the cross-sections than does a statistical treatment such as that used in the unresolved region. These kind of "pseudo-parameters" will be given up to about 1250 eV. Parameters for the description of intermediate structure above 1250 eV could also be obtained.

The new ORNL set of resonance parameters reproduces quite well the 2200 m/s cross-sections proposed by the ENDF/B-VI standard evaluation group (14) as it is shown on Table I. But the

problem still exists for the fission standard averages above 100 eV. The cross-sections calculated from the ORNL resonance parameters will be about 2 % lower than the average values proposed by the standard group in the energy range 100 eV to 500 eV with an accuracy less than 0.5 %. The explanation given by the ORNL evaluators for this discrepancy is the same than the one proposed for  $^{239}\text{Pu}$  ; some of the experimental fission data have an obvious high fission background amounting to several % of the average, compared to the 1984 WESTON data where such a background does not appear to be required in the SAMMY fit.

Important new measurements in the subthermal region of  $n$  at Harwell and of fission cross-sections at Geel and at ILL (Grenoble), and new fission measurements at NBS are now considered for inclusion in ORNL analysis.

### $^{241}\text{Pu}$

A review of the problems concerning the  $^{241}\text{Pu}$  neutron data in the resonance region is found in the proceedings of the 1981 Vienna conference of U and Pu isotopes resonance parameters (25). This item was introduced later in the INDC discrepancy file (26) and concerned mainly the difficulty for the normalization of WESTON et al. experimental absorption data (27). WESTON and WRIGHT (28) reported that the measurement of WESTON indicated a capture cross-section 14 % higher over the 0.27 eV resonance than the ENDF/B-IV evaluation. ENDF/B-IV was consistent with SIMPSON et al (29) old total and fission measurements. WESTON data were normalized to an absorption cross-section inferred from the total cross-section of KOLAR et al. (40).

High resolution transmission measurements were performed in 1972 at ORELA by HARVEY et al. (30), but the resonance parameters were not obtained from these data. The analysis of the data was restarted in 1987 and a SAMMY fit was performed (31) in the energy range thermal to 300 eV including the fission cross-sections from MIGNECO et al. (32), WESTON et al. (27), BLONS et al. (33), WAGEMANS et al. (34) and the absorption cross-sections from WESTON et al. (27). The results of the analysis have shown that :

- 1) WESTON absorption should be renormalized on the absorption obtained from HARVEY total cross-sections ;
- 2) below 0.03 eV, WESTON fission and absorption data deviate from the expected  $\frac{1}{v}$



shape of the cross-sections and should not be used for the normalization in the thermal range. After consideration of these two points, the inconsistency observed by WESTON et al. over the 0.27 eV resonance disappears, and the  $\alpha$  values are on average 14 % lower than those given by WESTON et al. (27) in the energy range thermal to 300 eV.

The cross-section values calculated at 2200 m/s are shown on Table I, and compared to the recommended values.

## CONCLUSION

This note is mainly a description of the very important improvements obtained since 1985 for the resonance analysis of  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  neutron cross-sections at ORNL. The analysis utilizes a physically meaningful formalism to represent the available data without introducing an artificial file 3 contribution. A spin is assigned to each level. The resolved resonance region is extended up to 500 eV, 1000 eV and 300 eV for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  respectively. The cross-sections in the thermal region is consistent with the values currently proposed for ENDF/B-VI by the standard evaluation committee. Some covariance matrices are available. One important point of discrepancy remains. The average fission cross-sections recommended by the standard evaluation committee above 100 eV are larger than those calculated from the resonance parameters. These discrepancies should be due to the systematic errors in the background corrections in some of the experimental fission cross-sections ; the importance of these systematic errors, which can be revealed only by a resonance analysis of the data, should be reconsidered by the standard evaluation group.

Important works were also performed in the eastern country on  $^{235}\text{U}$  and  $^{239}\text{Pu}$  by KONSHIN et al. (35), VANKOV et al. (36), BAKALOV et al (37), KOLESOV et al. (38). Most of these works are not well documented and concern a rather limited energy range ; a modified ADLER-ADLER formalism is generally used for multilevel analysis. G.B. MOROGOVSII (41) reported on self-consistent evaluation of the  $^{235}\text{U}$  total, fission and capture cross-sections in the energy range 1 eV to 100 eV using Breit-Wigner and Adler-

Adler formalism. Parameters of 201 resonances were adjusted to provide the best description of the available experimental data.

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## PROMPT NEUTRONS FROM FISSION

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The last compilation of  $\bar{\nu}_p$ , the average number of prompt neutrons emitted per fission, was established by KONSHIN and MANERO in 1972 [1] and is always largely used. However, a number of important results have since been published and therefore a brief status of the data will be presented. The aim of this status is not to provide recommended values, but rather to compare the most accurate sets of data obtained on broad energy ranges, in order to have a good basis to discuss the present state of the techniques.

$\bar{\nu}_p$  values are generally measured relative to  $\bar{\nu}_p$  for the spontaneous fission of  $^{252}\text{Cf}$  used as a standard. The review of absolute  $\bar{\nu}_p$  measurements for  $^{252}\text{Cf}$  is beyond the scope of the present investigation. An overview of the present situation can be found in one of the last papers published on that subject [20].

### 1. $\bar{\nu}_p$ for $^{235}\text{U}$

Two sets of data have been published for  $^{235}\text{U}$ : a measurement by GWIN et al. on ORELA [2] from thermal up to 10 MeV incident neutron energy, and our measurement between 1 and 15 MeV [3].

The most accurate data published to date for  $E_n \leq 1$  MeV are compared in Fig. 1. In this energy range, the BRC data and those of BOLDEMAN et al. have been corrected [4] and are in good agreement. They agree also with the data of GWIN et al. although these latter are about 0.5% higher.

The data of ORELA [2] and BRC [3] for  $E_n \geq 1$  MeV are compared in Fig. 2. They agree fairly well. The present BRC data confirm earlier measurements made in the same laboratory [5].

### 2. $\bar{\nu}_p$ for $^{238}\text{U}$

Two series of measurements have been made in USSR for this nucleus [6,7]. These data are compared with BRC data [5] in Fig. 3. The agreement is fairly good, except around 3 MeV incident neutron energy, where the change in slope

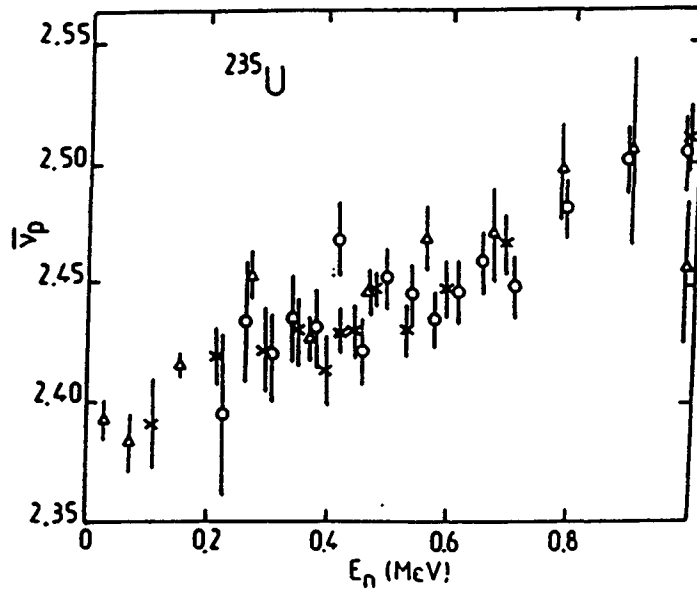


FIGURE III-1

$\bar{\nu}_p$  for  $^{235}\text{U}$  for  $E_n < 1$  MeV.

$\Delta$  : ref. [2]      X : ref. [4] (Boldeman)      O : Ref. [4] (BRC)

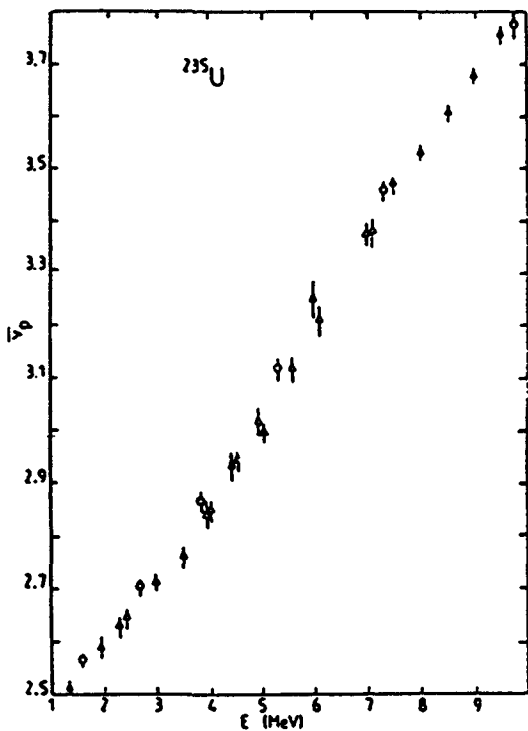


FIGURE III-2

$\bar{\nu}_p$  for  $^{235}\text{U}$  for  $E_n > 1$  MeV.

$\Delta$  : Ref. [3]      O : Ref. [2]

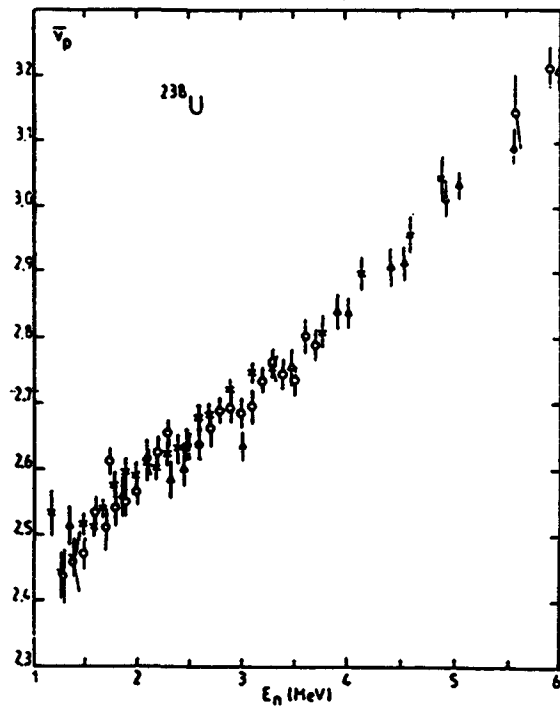


FIGURE III-3

$\bar{\nu}_p$  for  $^{238}\text{U}$ .

$\Delta$  : Ref. [5]      X : Ref. [6]  
O : Ref. [7]

observed for the BRC data is not reproduced by Russian data. However, the agreement should be better if the Russian data were lowered by about 1%.

### 3. $\bar{\nu}_p$ for $^{239}\text{Pu}$

Four sets of data, the most accurate published to date, have been compared: the measurements of BRC [5,8], of BOLDEMEN et al. [9], of NURPEISOV et al. [6] and of GWIN et al. [2]. The data below 1.5 MeV incident neutron energy are compared in Fig. 4. An overall good agreement is observed, with however the data of GWIN et al. higher by about 0.5% than others, such as in the case of  $^{235}\text{U}$ .

The data for  $E_n \geq 1.5$  MeV are compared in Fig. 5. The data of NURPEISOV et al. [6] are about 1% higher than the BRC data [5]. In this energy range the data of GWIN et al. are not accurate enough to permit a fine comparison with other data.

### 4. $\bar{\nu}_p$ for $^{237}\text{Np}$

The three published measurements of BRC [3], VEESER [10] and VOROBEOVA et al. [11] are compared in Fig. 6. The BRC data are about 3% lower than those of VEESER and about 2% lower than those of VOROBEOVA et al. It should be noted however that  $^{235}\text{U}$  and  $^{237}\text{Np}$  were measured simultaneously in a single experiment at BRC. Since the BRC  $^{235}\text{U}$  data, as discussed previously, are consistent with the other published data for this nucleus, we suspect the two other sets to be in error. This point will be discussed later on.

### 5. $\bar{\nu}_p$ for $^{232}\text{Th}$

Three measurements have been made for  $^{232}\text{Th}$ , by BRC [3,17], by VOROBEOVA et al. [12] and by HOWE et al. [13]. The accuracy of this latter measurement is quite poor, but the data agree on the average with the BRC measurements.

Fig. 7 compares the data of VOROBEOVA et al. and those of BRC. Russian data are higher by about 4% than the BRC data below 2 MeV incident neutron energy. At higher energies the difference is about 2%.

### 6. Other isotopes

Besides  $^{238}\text{U}$  and  $^{239}\text{Pu}$ , NURPEISOV et al. [6] have also investigated  $^{233}\text{U}$  in the energy range from thermal to 5 MeV. A measurement on  $^{236}\text{U}$  between 0.8 and 6 MeV has also been performed by MALINOVSKY et al. [18].

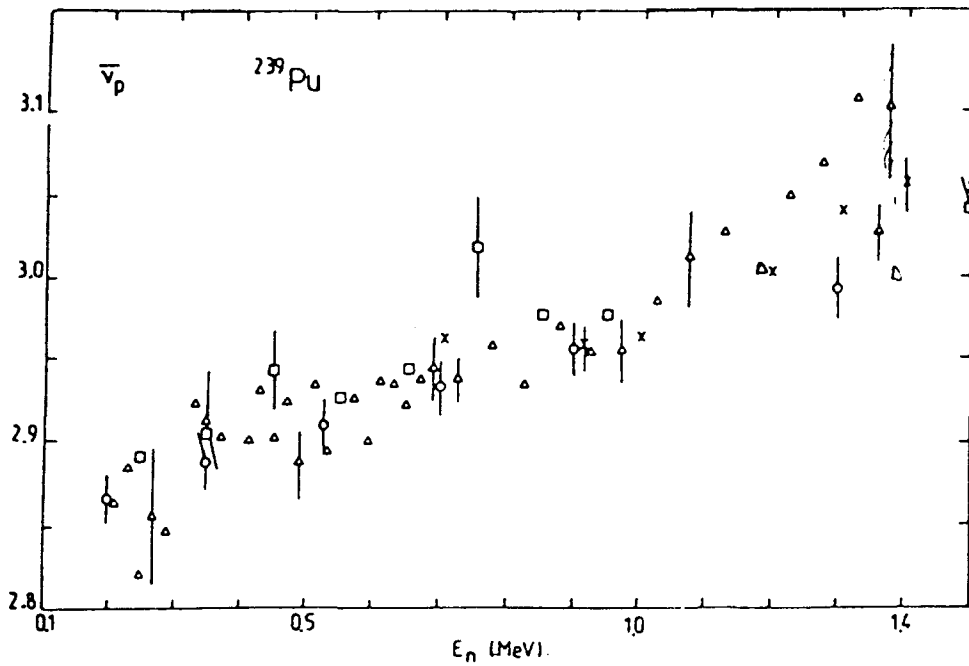


FIGURE III-4

$\bar{\nu}_p$  for  $^{239}\text{Pu}$  for  $E_n < 1.5$  MeV

$\Delta$  : Ref. [8]     $\square$  : Ref. [2]     $\circ$  : Ref. [9]     $\times$  : Ref. [6]

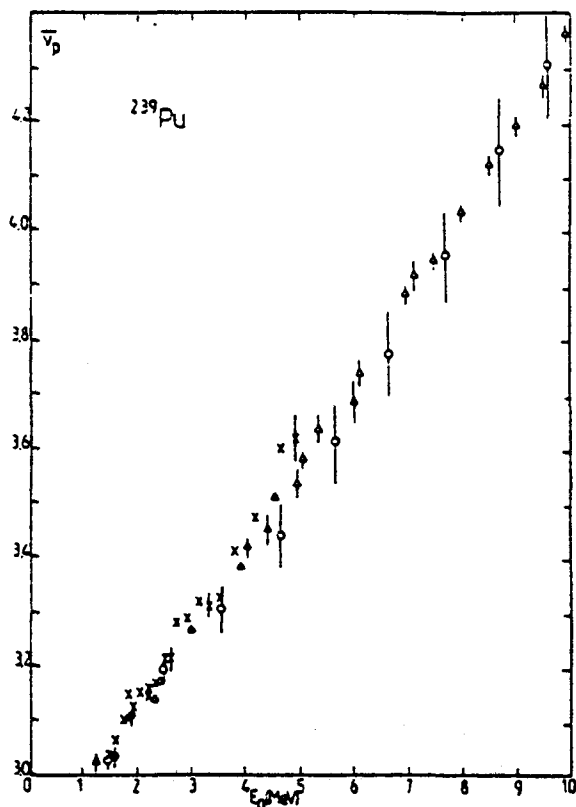


FIGURE III-5

$\bar{\nu}_p$  for  $^{239}\text{Pu}$  for  $E_n > 1.5$  MeV.

$\Delta$  : Ref. [5]     $\times$  : Ref. [6]

$\circ$  : Ref. [2]     $\bullet$  : Ref. [9]



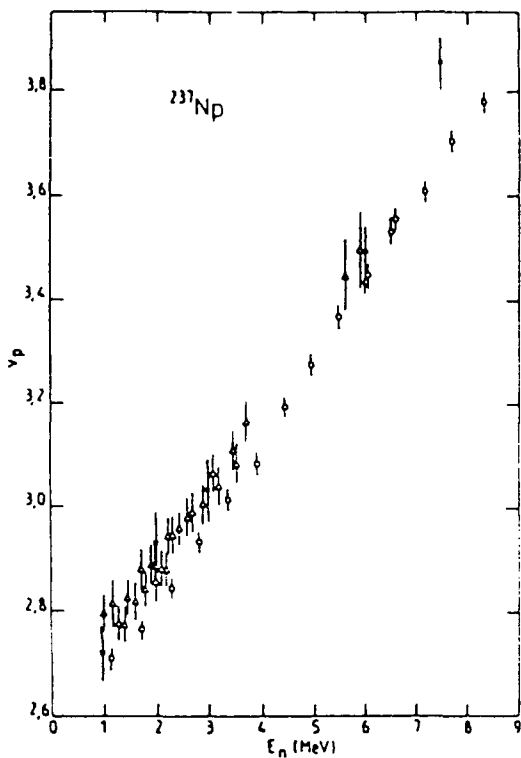


FIGURE III-6

$\bar{\nu}_p$  for  $^{237}\text{Np}$

O : Ref. [3] X : Ref. [10]

$\Delta$  : Ref. [11]

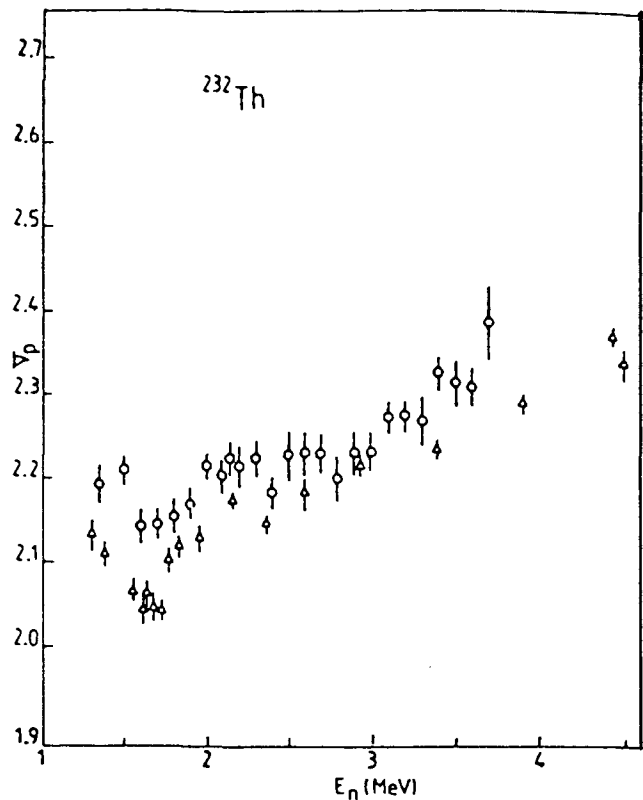


FIGURE III-7

$\bar{\nu}_p$  for  $^{232}\text{Th}$

$\Delta$  : Ref. [3] and [17] O : Ref. [12].

For the first time the isotopes  $^{242\text{m}}\text{Am}$  and  $^{245}\text{Cm}$  have been investigated at LIVERMORE in the energy range below 14 MeV [19]. Although the statistical accuracy is quite poor, the data for  $^{245}\text{Cm}$  show clearly that the slope  $d\bar{\nu}_p/dE_n$  is significantly smaller than estimated from systematic trends.

$^{243}\text{Am}$  has been investigated very recently in our laboratory between 6 and 15 MeV with a statistical accuracy of  $\sim 1\%$ . The preliminary data, unpublished, are well represented by a straight line of equation:

$$\bar{\nu}_p = 0.139 E_n + 3.28$$

## 7. Discussion and comments

The data which have been compared in the previous sections have been obtained using essentially two experimental techniques:

- $^3\text{He}$  counters embedded in a polyethylen block used as a neutron moderator. This technique was used by the two Russian groups.
- Large Gd-loaded liquid scintillator; this technique was used by the other groups.

With the exception of  $^{237}\text{Np}$ , the results obtained with the liquid scintillator technique are particularly coherent since the observed systematic differences are of the order of 0.5%. This kind of detector has a large neutron efficiency (~80%), is quite isotropic, its main characteristics have been carefully studied; accurate Monte Carlo simulations of its operation have been made for the absolute  $\bar{\nu}_p$  measurements on  $^{252}\text{Cf}$ . Therefore the corrections to be applied are quite small and can be determined accurately.

On the contrary, the  $^3\text{He}$  counter detector technique has not been investigated as carefully. Furthermore, its neutron detection efficiency is of the order of 30%, and this efficiency is more sensitive to the detected neutron energy spectrum than in the case of the liquid scintillator. Also, this detector is less isotropic, and therefore more sensitive to the angular distribution of the neutrons to be detected; the variation of the detector efficiency as a function of the source position along the detector axis is quite large. The correlation between these two latter effects has not been considered in corrections, but should not be negligible when long fission chambers are used [6,11].

NURPEISOV et al. [6] have improved significantly the angular response of their detector. The systematic difference of about 1% existing between their data and the results obtained using the scintillator technique can be considered as reasonable taking into account the present state of the techniques.

An incorrect estimation of the correction for the fragment emission anisotropy could explain the large difference observed in the case of  $^{232}\text{Th}$  around the fission threshold between the data of BRC and those of VOROBEVA et al. (Fig. 7). In this case, the fission fragments are emitted preferentially at large angles relative to the direction of incident neutrons inducing fission. Since the angular distribution of the fission neutrons is correlated to the fragment direction, these neutrons are detected with a better efficiency than in the case of an isotropic emission. The correction is quite

negligible in the case of the liquid scintillator, but not for the  $^3\text{He}$  counter detector.

An opposite effect can be predicted in the threshold region for  $^{238}\text{U}$ . In fact, Russian data appear to be quite low in that region (Fig. 3).

The identification of the fission events is quite different for the two types of detectors.

For the liquid scintillator technique, the identification results from a coincidence between the detection signal of a fission fragment in a fission chamber placed at the center of the detector and the scintillator signal corresponding to the detection of the prompt fission  $\gamma$ -rays. This procedure virtually eliminates the alpha piling-up problem for active materials such as  $^{239}\text{Pu}$  or  $^{237}\text{Np}$  and allows using relatively low thresholds on the fission chambers. Typically, 80% of the fission fragments are detected in our chambers.

The  $^3\text{He}$  counters are quite insensitive to  $\gamma$ -rays, and thus alpha piling-up rejection is obtained by using relatively high thresholds on the fission chambers: only 55% of the fragments were detected above threshold for  $^{237}\text{Np}$  in the measurement of VOROBEVA et al. [11].

The correlation between the fission threshold and the neutron detection efficiency has been investigated by BOLDEMAN et al. [14] and by GWIN et al [2] in the case of the scintillator technique. The efficiency remained almost constant, even when 80% of the fission events were lost. This is because they used fast fission chambers specially designed to obtain a good discrimination between fragment and alpha pulses, for which the height of the delivered pulses was not correlated to the fission fragment energy. Since we use very similar fission chambers at BRC, we have no correction to apply for this effect. We only correct for the loss of fragments in the deposit thickness using the data from our recent investigation [15].

On the contrary, Russian groups do observe a correlation between the neutron detection efficiency and the fission threshold: the height of the pulses delivered by their chambers remains certainly more or less correlated to the fragment energy. We have plotted in Fig. 8 the  $\bar{\nu}_p$  corrective factors given in ref. [6] and [11] as a function of the fragment detection

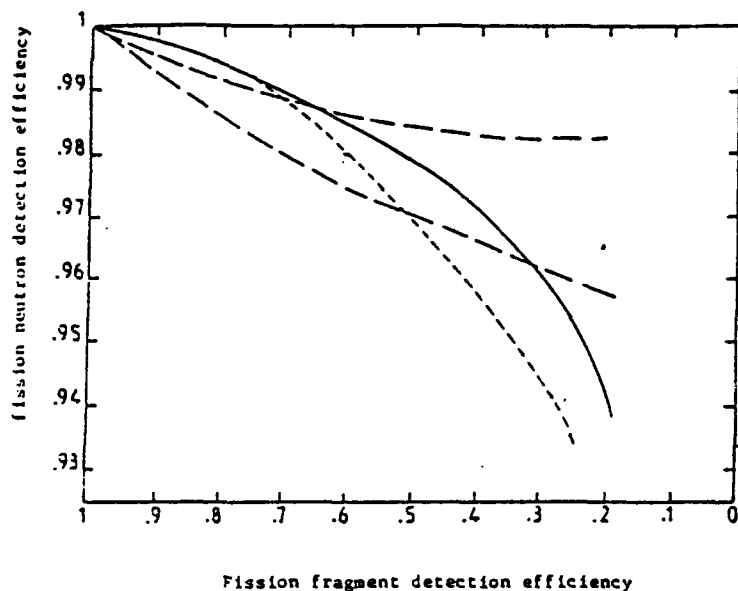


FIGURE III-8

Fission neutron detection efficiency  
(relative scale) as a function of  
fission fragment detection efficiency

- BRC measurement
- - - - - NURPEISOV et al. [6]
- . - . - VOROBEVA et al. [11]
- upper curve : deposit on grounded electrode
- lower curve : deposit on biased electrode.

efficiency. For comparison we have also plotted data obtained in our laboratory with the liquid scintillator using a  $^{252}\text{Cf}$  source placed in front of a solid state detector. The curve from NURPEISOV et al. is similar to the one we measured, which tends to confirm a linear response for their fission chamber.

VOROBEVA et al. [11] find a more important effect. But surprisingly and difficult to understand, the observed effect is different according to whether the fissile deposit is on the grounded or the biased electrode. Moreover, these authors do not use their curves, but only the point they measured for 55% efficiency with their  $N_p$  fission chamber, to deduce a correction of 3.7% for their data, which is larger by about 0.8% than the correction deduced from the curve for biased electrodes. And still, although this information is not given explicitly in the paper, it seems that half of the material was deposited on grounded electrodes. On the other hand their curves were

obtained from relative data measured between 55% and 80% fission detection efficiency. The shape of the curves and thus the absolute scale are rather arbitrary. Under these conditions the correction could be of the order of 2% for  $^{237}\text{Np}$ , in agreement with the curve of NURPEISOV (Fig. 8), and thus their data would be in agreement with the BRC measurement. It should be mentioned also that this fission threshold effect on neutron efficiency is correlated to the deposit thickness and to the detector anisotropy and thus the 3 corresponding corrections are not independent. This correlation could be at the origin of the difference between the curves of NURPEISOV et al. and BRC on Fig. 8.

For the measurements on  $^{232}\text{Th}$ , the systematic difference of ~2% observed above 2 MeV between the data of VOROBÉVA et al. [12] and the data of BRC [3] is larger than the 1.1% correction applied for the fragment detection efficiency by VOROBÉVA et al. (to be compared with a correction of 0.2% for  $^{238}\text{U}$ ) [7]. However their publication is not documented enough to draw any valuable conclusion.

We brought attention in 1972 [16] on background problems connected with induced fission measurements. Briefly, the neutron detector background rate and the induced fission rate are proportional to the incident neutron flux. Thus, for the background to be measured correctly, the corresponding counting gates must be opened either systematically after each fission event, or randomly by using as a trigger pulses from a neutron detector placed in the incident beam. If the counting gates were opened using an external generator as a trigger - i.e. without reference to the incident neutron flux - any fluctuation in the incident beam intensity would necessarily result in an underestimation of the real background rate, and subsequently to an overestimation of  $\bar{\nu}_p$ . Such an effect has been also clearly seen by GWIN et al. [2] for measurements on OREIA, where deviations as large as 3% were observed on  $\bar{\nu}_p$ . Such an effect is certainly at the origin of the high values of  $\bar{\nu}_p$  obtained by VEESER [10] for  $^{237}\text{Np}$ .

In fact, all  $\bar{\nu}_p$  measurements where an external generator was used to trigger the background measurement are likely to be overestimated and should not be used in an evaluation.

Incidentally, we questioned in the above mentioned 1972 publication [16] our data published in 1969 [5] and provisional corrected values were given ( $\bar{v}_p$  was increased by 1 to 3% for incident neutron energies below 8 MeV) for  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$ . Such a correction was not confirmed by subsequent control measurements, and thus the data of ref. [5] are always valid. However use was made of our provisional corrected values in the compilation of KONSHIN and MANEKO [1] which is often used as a reference. Therefore this compilation overestimates  $\bar{v}_p$  below 8 MeV for  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$ .

The foil thickness effect presented in details in ref. [21] might have been underestimated up to now. Fragments emitted at large angles relative to the normal of the deposits are not detected. Since the direction of the fission neutrons is correlated to the direction of the fragments, the measured  $\bar{v}_p$  value is correlated to the deposit thickness when the neutron detector has not a perfect  $4\pi$  geometry. This effect has been investigated at BRC [15] for the Gd-loaded liquid scintillator. Corrections of the order of 0.25% have been found for deposits of oxides  $1 \text{ mg/cm}^2$  thick. However the amplitude of the correction should depend dramatically on the deposit homogeneity. The presence of grains at the surface, for example, should increase considerably the effect. Thus the quality of the deposit seems to be essential for accurate measurements. The investigation of that effect was originally motivated to explain a discrepancy observed in thermal  $\bar{v}_p$  measurements by GWIN [2] and BOLDEMAN [14]. While their  $^{239}\text{Pu}$  data are in agreement, their  $^{235}\text{U}$  data differ by 0.8%. The deposit thickness was of  $0.1 \text{ mg/cm}^2$  for both the  $^{239}\text{Pu}$  measurements and for the  $^{235}\text{U}$  measurement of GWIN, but of  $0.8 \text{ mg/cm}^2$  for the  $^{235}\text{U}$  measurement of BOLDEMAN. Only one third of the difference was attributed to the deposit thickness effect [15], but the influence of the possible inhomogeneities remains an open question.

In all  $\bar{v}_p$  measurements a correction must be applied to account for the difference between the fission neutron energy spectrum of the investigated isotope and that of  $^{252}\text{Cf}$ . The energy dependence of the neutron detector efficiency is generally well established, but the fission neutron energy spectrum is rather uncertain. Most of the measurements rely on the TERRELL law [22] which correlates the average maxwellian spectrum energy with  $\bar{v}_p$ .

This relation is certainly not valid above the second chance fission threshold. A better approach would be now to use the formalism developed by MADLAND and NIX [23].

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$\bar{\nu}_p$  for  $^{237}\text{Np}$

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The situation concerning  $\bar{\nu}_p$  for neutron induced fission of  $^{237}\text{Np}$  remains unchanged, since there is no work in progress in that domain.  $\bar{\nu}_p$  values from BRC are ~3% lower than the Los Alamos data of Lynn Vesser, and ~2% lower than the data of Vorobeva et al.

The BRC measurement has been made using a single fission chamber containing  $^{235}\text{U}$  and  $^{237}\text{Np}$  deposits; the data obtained for  $^{235}\text{U}$  are in agreement with other published data as well as with previous BRC measurements by Soleilhac et al. in 1969.

An Analysis of experimental techniques used for  $\bar{\nu}_p$  measurements [1] brings to light some problems which are probably at the origin of the observed discrepancies for  $^{237}\text{Np}$  as well as for  $^{232}\text{Th}$  and, to a lower extent, for  $^{238}\text{U}$ . A more thorough experimental study of these problems would be worthwhile.

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IAEA-TECDOC-336 (1985)



## FICHE SUR LES DESACCORDS CONCERNANT $^{237}\text{Np}$

J. FREHAUT

$^{237}\text{Np}(n,2n)^{236}\text{Np}(22,5h)$

L'intérêt de bien connaître cette section efficace est que le  $^{236}\text{Np}(22,5h)$  donne par désintégration  $\beta$   $^{236}\text{Pu}(5,8\text{ a})$  dont les produits de décroissance sont émetteurs de rayonnements  $\gamma$  pénétrants. Elle joue un rôle important dans les calculs de blindages pour le retraitement du combustible.

Jusqu'en 1984 on disposait de quelques résultats expérimentaux (1 valeur à 9,6 MeV et quelques valeurs autour de 14 MeV) [1] qui ne permettaient pas d'être très confiant dans les évaluations faites (pour ENDF/B.V la section efficace paraissait nettement sur-évaluée entre le seuil et 10-11 MeV).

Les mesures de N.V. KORNILOV [2] entre 7,1 et 9,9 MeV ont complété très utilement en 1984 les mesures antérieures. Les résultats des évaluations qui ont suivi [3], en bon accord avec les données expérimentales, ont finalement permis d'obtenir une fonction d'excitation couvrant la gamme d'énergie demandée [4] et en laquelle on avait confiance.

Cette situation a conduit à considérer qu'il n'existait plus de désaccords importants [5]. Ceci a été confirmé par les résultats de l'expérience PROFIL 2 (irradiation de  $^{237}\text{Np}$  très pur dans le réacteur Phénix) qui, pour la valeur moyenne de la section efficace et le rapport isomérique, sont en accord avec les résultats des calculs faits à partir des dernières données évaluées [6].

$\bar{\nu}_p$  de  $^{237}\text{Np}$

Faute de nouvelles études, la situation n'a pas évolué en ce qui concerne le  $\bar{\nu}_p$  du  $^{237}\text{Np}$  : Ces valeurs mesurées à BRC sont  $\sim 3\%$  plus basses que la mesure de Lynn Veaser [7] à Los Alamos, et  $\sim 2\%$  plus basse que la mesure de Vorobeva et al. [8].

La mesure de BRC a été effectuée à l'aide d'une chambre à fission contenant du  $^{235}\text{U}$  et du  $^{237}\text{Np}$  ; les résultats obtenus pour  $^{235}\text{U}$  sont en accord avec les autres résultats publiés ainsi qu'avec des mesures antérieures de Soleilhac et al. [9].

Une étude des techniques expérimentales utilisées pour les mesures de  $\bar{\nu}_p$  [3] met en lumière un certain nombre de problèmes qui sont

vraisemblablement à l'origine des désaccords constatés, non seulement sur  $^{237}\text{Np}$ , mais aussi sur  $^{232}\text{Th}$  et, à un moindre degré, sur  $^{238}\text{U}$ . Ces problèmes mériteraient une analyse plus détaillée, en particulier sur le plan expérimental, mais rien ne semble avoir été entrepris dans ce domaine.

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## $^{239}\text{Pu}$ DECAY POWER DISCREPANCY

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### A. BACKGROUND

An extensive effort was undertaken in the time period 1973-1978 to improve the basic decay-power standard used in U.S. reactor calculations. The following activities were included in this effort and were completed in 1978:

1. The U.S. Department of Energy (DOE--then ERDA) formed a task force to add fission-product decay, cross section, and yield data for 825 nuclides to its evaluated data file (ENDF/B-IV) and supported efforts to combine analyses using these data with experiments to produce an improved standard.
2. The U.S. Nuclear Regulatory Commission (NRC) supported new benchmark experiments at the Los Alamos National Laboratory (LANL) and Oak Ridge National Laboratory (ORNL) to accurately measure decay heat following fission of  $^{233}\text{U}^*$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and  $^{241}\text{Pu}$ .\*\*
3. The Electric Power Research Institute (EPRI) supported new decay-heat experiments at the University of California (Berkeley) and at Intelcom Rad Tech (IRT).

The participants and results of these activities were brought together in an American Nuclear Society (ANS) standards committee, and a new ANS Decay Power Standard<sup>1</sup> was approved in 1979. The final standard was determined from a generalized least-squares analysis that combines summation calculations using the ENDF/B-IV data base with these and other decay-heat measurements. In obtaining the standard, a concerted effort was made to reconcile the various decay-heat measurements with the yield and decay data in the ENDF/B-IV data file. The development of the standard, the experimental data that were used, and the  $^{239}\text{Pu}$  discrepancy noted below are described in Refs. 2 and 3.

### B. $^{239}\text{Pu}$ DISCREPANCY

The decay-heat experiments carried out at LANL<sup>4</sup> and ORNL<sup>5,6</sup> are the most precise of the various measurements and therefore exert the most influence on the new decay-heat standard. The two experiments employ entirely different techniques. In the LANL experiment the decay heat of irradiated fissile samples was measured directly in a precision calorimeter, whereas energy spectra of decay gamma rays and beta particles were measured at various times following relatively short irradiations in the ORNL experiment. The uncertainties in the two experiments range from 2-1/2 to 4 %; however, the results differ by approximately 10% for the  $^{239}\text{Pu}$  measurements. (The  $^{235}\text{U}$  results also differ but by a smaller amount.) The discrepancy between the LANL and ORNL measurements is shown in Fig. 1, where the plots have been derived for a common irradiation time.<sup>2</sup> The measurement in France by Lott and Fiche tends to support the LANL result but has a relatively large uncertainty.

Summation calculations using the ENDF/B-IV data base (and calculations using an independent U.K. data base by Tobias) do not clarify the discrepancy. There is excellent

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\* LANL only.

\*\* ORNL only

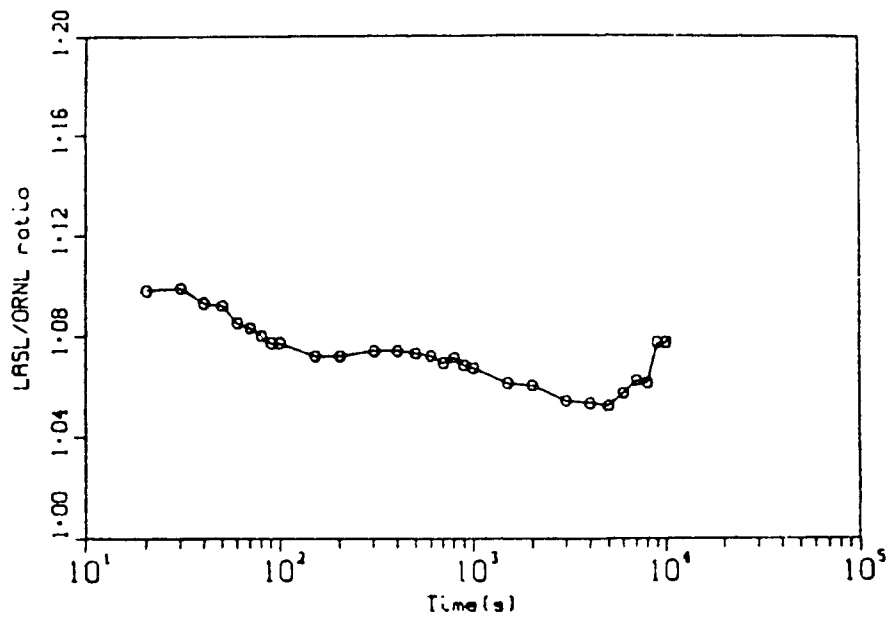


Fig. 1. <sup>239</sup>Pu decay-heat ratio of LANL to ORNL following a 20,000-s irradiation.

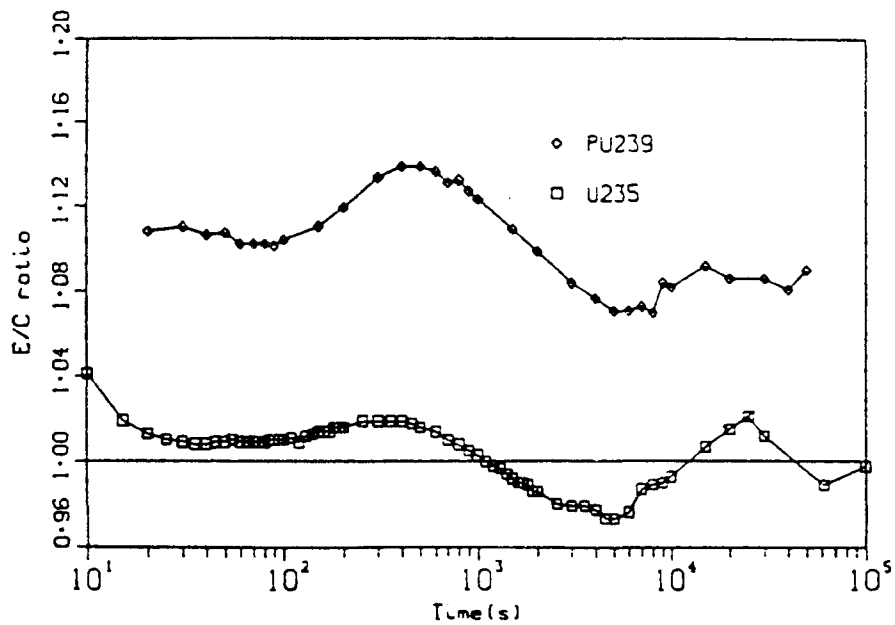


Fig. 2. Ratio of LANL decay-heat measurements to ENDF/B-IV calculations (20,000-s irradiation).

agreement between the calculations and the LANL measurement of  $^{235}\text{U}$  decay heating, but for  $^{239}\text{Pu}$  the calculations agree better with the ORNL results. Figure 2 shows the ratio of calculated to measured decay heat for the LANL  $^{235}\text{U}$  and  $^{239}\text{Pu}$  results following a 20,000-s irradiation.

References 2 and 3 discuss the unsuccessful efforts to resolve this discrepancy in deriving the new decay-heat standard. The new standard for  $^{239}\text{Pu}$  is intermediate between the LANL and ORNL measurements, whereas the  $^{235}\text{U}$  standard is consistent with the measurements from both labs. However, the  $^{239}\text{Pu}$  discrepancy could also reflect problems for the  $^{235}\text{U}$  standard and should be resolved as soon as possible.

### C. NEANDC SPECIALISTS' MEETING AT STUDSVIK

The status of decay heat measurements was reviewed at an NEANDC Specialists' Meeting held in Sweden.<sup>7</sup> Although two new measurements of  $^{239}\text{Pu}$  decay heat are available<sup>8</sup> or are in progress,<sup>9</sup> the possibility of resolving the differences between the earlier Los Alamos<sup>4</sup> and ORNL<sup>5,6</sup> measurements appears remote. In particular, the University of Tokyo experiment<sup>8</sup> utilized similar techniques to the earlier ORNL measurement (separate detection of beta and gamma rays) in a fast-neutron fission experiment, and the  $^{239}\text{Pu}$  decay heat results agree with the ORNL data. The Tokyo results for  $^{235}\text{U}$ , however, show a discrepancy with the ORNL data for gamma decay heat. The second experiment<sup>9</sup> was carried out with thermal neutrons at the University of Uppsala, again using methods similar to those employed for the ORNL measurements. As of this writing, only relative values are available for the  $^{239}\text{Pu}$  gamma decay heat, but the Swedish results for beta decay heat are absolute and agree well with the ORNL measurements. The Swedish data for  $^{235}\text{U}$  gamma heating, however, are about 12% higher than the equivalent ORNL results, and differences in the shape as a function of time are also seen.

Finally, it should be pointed out that a new measurement of total decay heat for  $^{235}\text{U}$  has been made at Karlsruhe<sup>10</sup> using thermal neutrons. This experiment was performed using a calorimetric technique, and the results are very similar to those from Los Alamos.<sup>4</sup>

The conclusion reached at the Specialists' Meeting is that the discrepancy in  $^{239}\text{Pu}$  decay heat is part of a broader problem between integral measurements for  $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , and other fissioning systems on the one hand, and between these measurements and summation calculations using evaluated data files on the other. It was recommended that a task force be set up to consider not only total decay heat, but also the separate beta and gamma components and their spectra. It was also recommended that sensitivity studies should be considered by the task force.

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