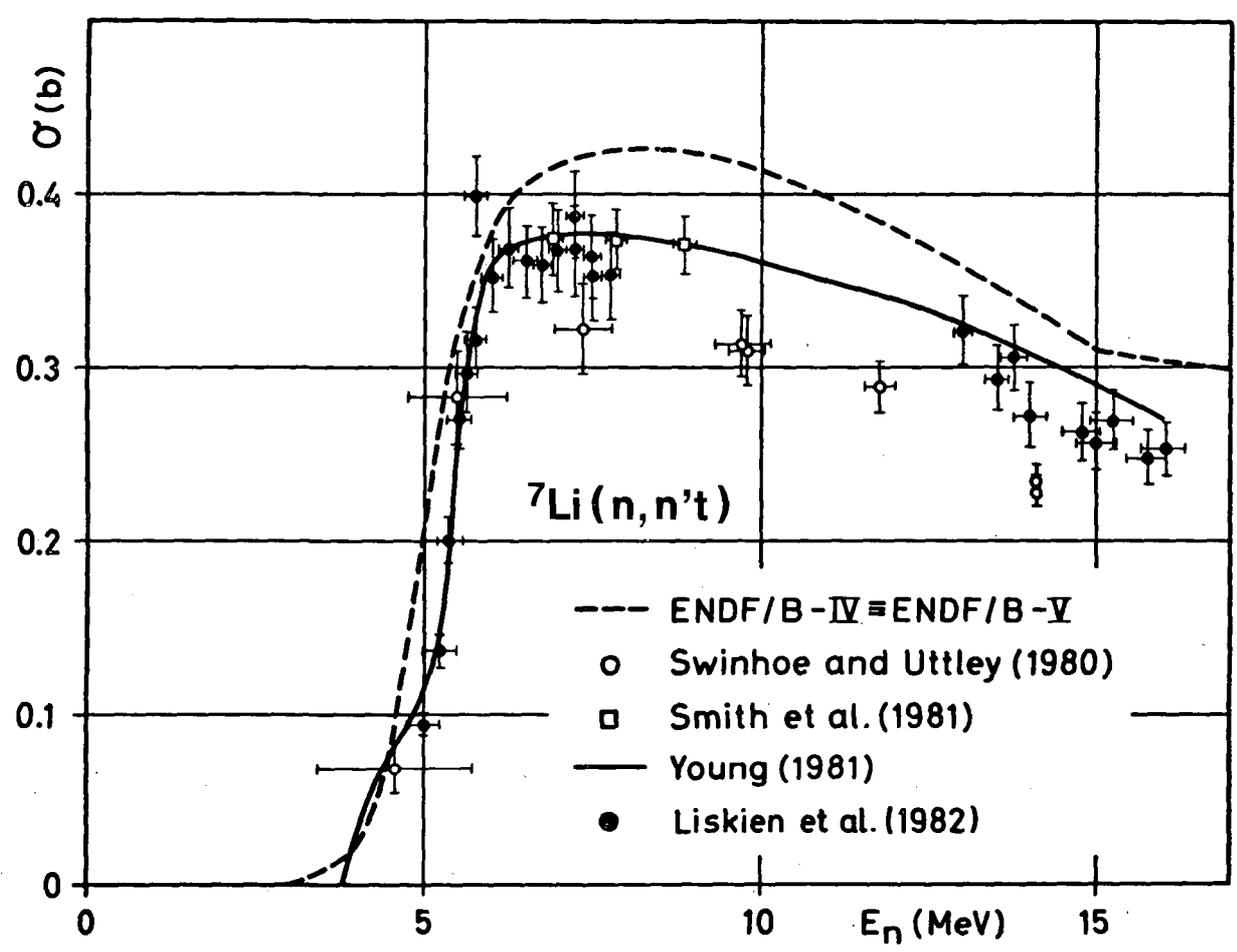


# NUCLEAR DATA DISCREPANCIES 1983



The NEANDC-INDC joint Discrepancy file





# **NUCLEAR DATA DISCREPANCIES**

The 1983 NEANDC-INDC Discrepancy file  
maintained jointly by  
OECD / NEA Nuclear Data Committee  
International Nuclear Data Committee (IAEA)

**OECD NUCLEAR ENERGY AGENCY**  
38 boulevard Suchet 75016 Paris

January 1984

- (20)  $^{239}\text{Pu}$  fission cross-section
- (21)  $^{109}\text{Ag}$  capture cross-section
- (22)  $^{243}\text{Am}$  capture resonance integral

Files are not available for 7 out of the 22 topics. For items (15) and (19) on the  $^{103}\text{Rh}(n,n')^{103m}\text{Rh}$  cross-section and the  $\Gamma_\gamma$  of the 2.85 keV resonance in  $^{23}\text{Na}$  there are no new data and so new File entries have not been made. (The most recent reviews of these two topics are given by Pitsaikin (INDC/P(83)-34) and Jackson (NEANDC-105/L, INDC 24/G p.73) respectively). The  $^{239}\text{Pu}$  decay power (item 12) was recently reviewed at the NEANDC Specialists' Meeting on "Yields and Decay Properties of Fission Products", Brookhaven 24-27th October 1983 (see also the Antwerp Conference proceedings (EUR 8355) pages 237, 245, 249). The  $^{238}\text{U}$  inelastic scattering data (item 10) were considered at the NEANDC Specialists' Meeting on "Fast Neutron Scattering on Actinide Nuclei", Paris 1981 (NEANDC-158U) and also at the Antwerp Conference (EUR 8355, p.9). The capture cross-section data on  $^{232}\text{Th}$  (item 5) have recently been reviewed by Poenitz at the NEANDC/NEACRP Specialists' Meeting on "Fast Neutron Capture Cross-sections" (NEANDC(US)-214/L, p.288) while the  $^{239}\text{Pu}/^{235}\text{U}$  fission ratio (relevant to item 20) has been discussed by Behrens (BNL-NCS-51123, Section B, p.XVII.1). Experimental work relevant to the discrepancy in Cr and Ni total and inelastic scattering cross-sections (item 3) has been published by ANL group (Guenther et al., Nucl. Sci. and Eng. 82 (1982) 402 on Cr and Budtz-Jorgensen et al., Zts. f. Physik 306 (1982) 265 on  $^{58}\text{Ni}$ ). It is also to be noted that the  $^{235}\text{U}$  fission cross-section data (item 8) have been reviewed in the 1982 INDC/NEANDC Nuclear Standards File (IAEA Tech. Report Series No. 227 (1983)).

The members of the NEANDC Sub-Committee on Discrepancies at the Chalk River Meeting in September 1982 were as follows:

|                |   |          |                 |   |         |
|----------------|---|----------|-----------------|---|---------|
| K. H. Böckhoff | - | CEC Geel | J. L. Rowlands  | - | UK      |
| R. E. Chrien   | - | USA      | S. M. Qaim      | - | Germany |
| S. Cierjacks   | - | Germany  | J. J. Schmidt   | - | IAEA    |
| C. Coceva      | - | Italy    | M. G. Sowerby   | - | UK      |
| H. Condé       | - | Sweden   | N. Tubbs        | - | NEA     |
| W. G. Cross    | - | Canada   | S. L. Whetstone | - | USA     |
| P. Garvey      | - | Canada   |                 |   |         |
| S. Igarasi     | - | Japan    |                 |   |         |
| A. Michaudon   | - | France   |                 |   |         |
| H. T. Motz     | - | USA      |                 |   |         |
| C. Nordborg    | - | NEA      |                 |   |         |
| F. G. Perey    | - | USA      |                 |   |         |

The  ${}^7\text{Li}(n,n't){}^4\text{He}$  cross-section

Of interest is the total tritium production cross-section for  ${}^7\text{Li}$  in the neutron energy range of 2.8 to 16 MeV. It is one of the key reactions for tritium breeding in fusion blankets and the desired maximum uncertainty is  $< 5\%$ .

The tritium production cross-section of  ${}^7\text{Li}$  below the  $(n,2n)$ -threshold of 8.3 MeV is essentially equal to the inelastic scattering cross-section. Only the first excited level of  ${}^7\text{Li}$  is particle-stable, but neutrons populating this level are typically not separated from ground state neutrons in TOF-experiments<sup>(1)</sup>. However, a determination as difference of the total and the elastic scattering cross-section leads to large uncertainties. A determination by observing the inelastic neutrons<sup>(1)</sup> or the emitted tritons<sup>(2)</sup> yields only results of modest accuracy. This is due to the fact that the observed energy spectra of the emitted particles have to be extrapolated to zero energy and an integration over the emission angle has to be performed. The most accurate method is certainly the activation method using quantitative tritium extraction from irradiated bulk samples and activity counting.

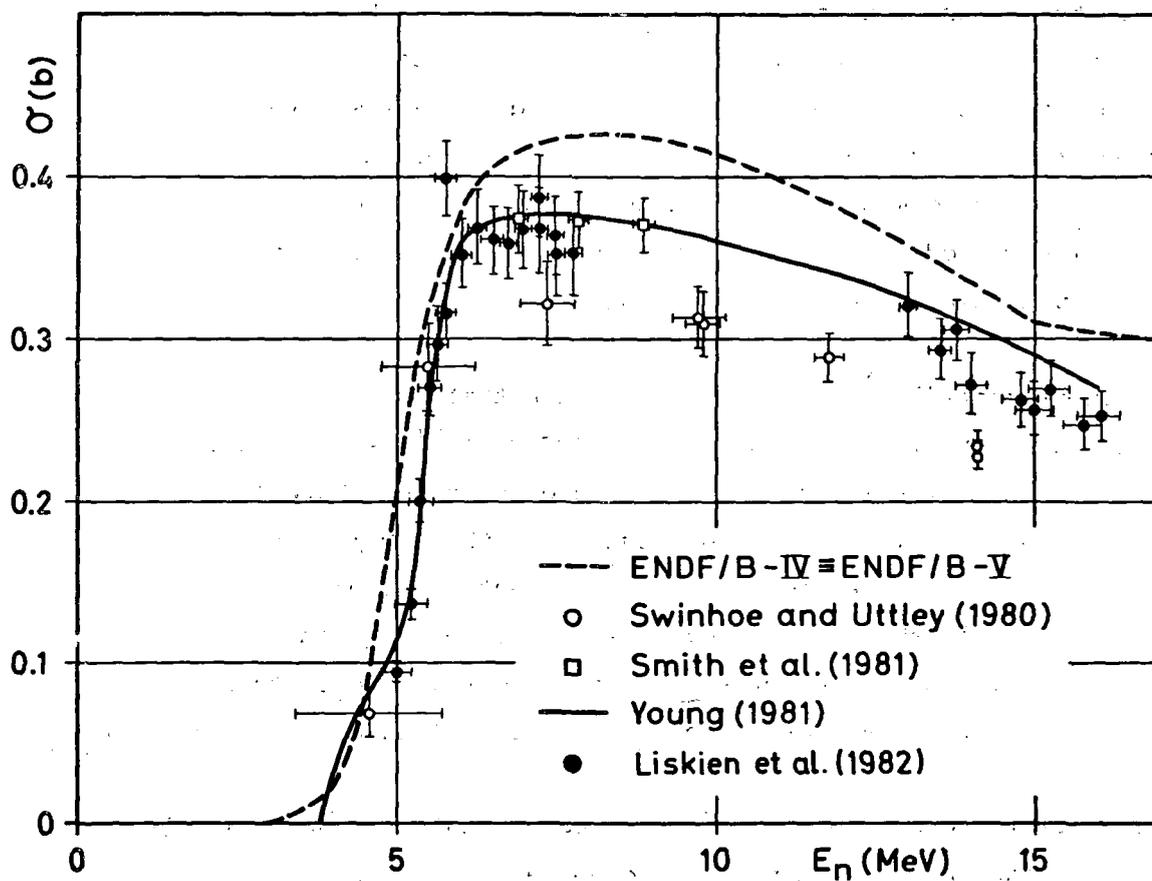
Integral experiments<sup>(3-5)</sup> led to the suspicion that the  ${}^7\text{Li}(n,n't){}^4\text{He}$  cross-section as predicted by ENDF/B-IV(=ENDF/B-V)<sup>(6)</sup> is too high. Indeed, a recent activation experiment conducted at AERE Harwell<sup>(7)</sup> resulted in cross-sections about 26% lower than the ENDF/B. This prompted two other activation experiments. One performed at Argonne National Laboratory<sup>(8)</sup> was restricted to measurements at 7, 8 and 9 MeV and yielded cross-sections about 17% lower than ENDF/B. The other work was carried out as a Geel-Jülich collaboration<sup>(9)</sup>.

The results of various recent activation measurements are shown in the accompanying diagram. Evidently the Argonne values (Smith et al) and the Geel-Jülich results (Liskien et al) are in good agreement. They are also consistent with a recent Los Alamos evaluation (Young<sup>(10)</sup>). The Harwell values (Swinhoe and Uttley) are too low.

In view of the good agreement between recent measurements and evaluation it is recommended that this reaction be deleted from the Discrepancies File. However, whether the 6 to 7% uncertainty in the cross-section is acceptable by the users is a different matter. It appears that with the presently available techniques the cross-section cannot be determined with uncertainties less than 5%.

S.M. Qaim, KFA Jülich

January 1983



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## Capture Cross-Sections for Cr, Fe and Ni.

### Description

The interest in structural material data for reactor applications has considerably increased since the last report on this subject; indicated by two activities :

- 1) The NEACRP and NEANDC have endorsed a High Priority List (NEACRP/A-500, NEANDC/A-156), where capture measurements for  $^{56}\text{Fe}$ , Fe, Ni and Cr are asked up to 1 MeV neutron energy (Cr up to 100 keV) with accuracy requirements of 10-15, 5-10, 10-20 and 20% respectively.
- 2) The discrepancies observed in iron capture cross-section measurements as identified in a review at the Argonne Specialists Meeting <sup>1)</sup> and in contributions to the Antwerp Conference has led the NEANDC to set up an international task force with the goal to understand and remove these discrepancies.

### Status

The status of structural material data has been reviewed at the Specialists Meetings at Geel and Argonne as well as in the contributions for the discrepancies file (NEANDC-105L (1976), NEANDC-124A (1980) ).

Since the last discrepancy file the following works concerned with structural material capture data have been published :

Capture data of  $^{56}\text{Fe}$  measured at Geel have been analysed at Harwell up to a neutron energy of 100 keV using REFIT <sup>2)</sup>.

Different types of Moxon-Rae detectors have been used to test the influence of the  $\gamma$ -ray detection linearity on the data obtained for broad s-wave resonances (27.7 keV resonance of  $^{56}\text{Fe}$  included) at the Karlsruhe pulsed Van de Graaff generator with time-of-flight discrimination of scattered neutrons <sup>3)</sup>

The status of structural material data in general <sup>1)</sup> and the status of the resonance parameters of the 27.7 keV resonance <sup>4)</sup> have been reviewed in contributions to the Argonne Meeting.

High resolution measurements of  $^{54}\text{Fe}$ ,  $^{56}\text{Fe}$  and  $^{57}\text{Fe}$  performed with  $\text{C}_6\text{D}_6$  detectors at the Linac of Geel and analysed up to a neutron energy of 200 keV ( $^{56}\text{Fe}$  up to 250 keV) have been published at the Antwerp Conference <sup>5),6),7)</sup>

Capture measurements of  $^{56}\text{Fe}$  and  $^{58}\text{Fe}$  have been performed at the Karlsruhe Van de Graaff using  $\text{C}_6\text{D}_6$ -detectors and resonance parameters have been obtained in the energy range 10-100 keV<sup>8)</sup>.

Moxon-Rae detector measurements of the broad 27.7 keV resonance, performed with pulsed Van de Graaff accelerators, which have been mentioned in the previous discrepancy file have the Refs. 9) and 10).

### Discrepancies

A review on the accuracy aspect of iron isotope resonance parameters has been given by G. Rohr at the Argonne Meeting where the following conclusions were made :

1. Comparing the capture area of five resonance data sets for  $^{56}\text{Fe}$  :

a) the largest deviation between data sets is of systematic origin and due to normalization.

Normalizing the data to one resonance, the agreement of data obtained with similar techniques agree much better.

b) the remaining deviation of data obtained with the Maier-Leibnitz method and with large liquid scintillators are correlated with the hardness of the  $\gamma$ -ray spectrum in the resonances.

The application of the normalization (a) and a suitable correction for data obtained with the Maier-Leibnitz method described in Ref. 1) reduces the discrepancy averaged over all considered resonances from 20% to 6%.

2. Comparing the capture data of the iron isotopes  $^{54,56,57}\text{Fe}$  :

There is an indication of a systematic 15% change in the flux below 30 keV by comparing data from ORNL and CBNM.

3. The 1.15 keV ( $^{56}\text{Fe}$ ) resonance is well suited for normalization purposes, but the published values for the neutron width obtained from capture and transmission measurements differ by up to 36%.

### Recommendation

1. Precise determination of the  $^{56}\text{Fe}$  1.15 keV resonance parameters by transmission measurements.
2. Capture measurements of  $^{56}\text{Fe}$  with different detectors, in particular with large liquid scintillators.

## References

- 1) G. Rohr, Proceedings of Specialists Meeting on Fast Neutron Capture, 20-23 April 1982, Argonne National Laboratory, Argonne, Ill., ANL-83-4, p.394.
- 2) M.C. Moxon, U.K. Nuclear Data Progress Report (Jan.-Dec. 1980) NEANDC (E) 222, Vol. 8, p. 22.
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- 8) F. Käppeler, K. Wisshak, L.D. Hong, KFK 3412, Nov. 1982
- 9) B.J. Allen, D.D. Cohen, F.Z. Company, J. Phys. G-6 (1980) 1173
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G. Rohr

CBNM Geel

January 1983

ON THE NEED FOR FURTHER EXPERIMENTAL WORK TO IMPROVE THE Ni NEUTRON  
CROSS-SECTION DATA

At the NEACRP/NEANDC specialist meeting held in May 1973 at Karlsruhe, M.C. MOXON (1) in his evaluation of the neutron cross-sections of Ni came to the conclusion that "the available experimental data were scant and often of poor quality". He gave several recommendations for accurate measurements of total and capture cross-sections to be analysed in terms of resonance parameters in the energy range 1 keV to hundreds of keV, with special need of obtaining the spin assignment of the  $l > 0$  resonances and accurate values of the average radiation widths. The next specialist meeting on neutron data for structural materials was held at Geel in December 1977 and reports were given by GAYTHER et al. (2) on capture cross-section measurements, by JAMES et al. (3) and by SYME et al. (4) on transmission measurements, and by PEREY et al. (5) on transmission and capture measurements. These works, performed at HARWELL and OAK-RIDGE, gave some answers to the 1973 recommendations of MOXON. The following improvements of the data were obtained :

- 1/ Capture area for 45  $l > 0$  resonances of  $^{58}\text{Ni}$  in the energy range 7 keV to 120 keV (5) ;
- 2/  $g\Gamma_n$  values for 117  $l > 0$  resonances of  $^{58}\text{Ni}$  in the energy range 7 keV to 650 keV (4), (5) ;
- 3/  $g\Gamma_n$  values for 29  $l > 0$  resonances of  $^{60}\text{Ni}$  in the energy range 30 keV to 300 keV (4) ;
- 4/ more accurate values of  $\Gamma_n$  and  $\Gamma_\gamma$  of the s wave resonances for  $^{58}\text{Ni}$  and  $^{60}\text{Ni}$  (4), (5) ;

However, the results from PEREY et al. were only preliminary and no spin assignment was given for  $l > 0$  resonances.

In the proceeding of the HARWELL conference (september 1978), one finds one paper by SYME and BOWEN (6) on resonance analysis of  $^{58}\text{Ni}$  transmission data, which was the continuation of the work presented at Geel in reference (4), (5). They gave the  $\Gamma_n$  values of 41 s wave resonances up to 640 kev neutron energy and further work was foreseen (70 s wave resonances up to about 1.2 Mev as it is shown on fig.4 and 5 of SYME and BOWEN paper, and neutron widths for 230  $l > 0$  resonances below 1 Mev). Nothing was given on the status of the OAK-RIDGE work.

At the recent Antwerp conference (september 1982) nothing was presented concerning the work at HARWELL and at OAK-RIDGE or by other laboratories, in the resonance region. The newest data found in the EXFOR international file are still from GEEL 77 and HARWELL 78 (accession number 10691, 20858, 20861, 21212).

Before concluding on the needs of further experimental works up to 1 Mev, one should know from HARWELL and OAK-RIDGE experimenters if one could expect in the near future more informations from their experimental transmission, capture and scattering data. Particularly, is it possible to obtain from OAK-RIDGE a definitive set of  $^{58}\text{Ni}$  resonance parameters with the spin assignment for  $l > 0$  resonances, as it was done by the same team for  $^{56}\text{Fe}$  resonance parameters ?

One find in the "INDC discrepancy file 1979" (7), two recommendations for Ni isotopes : 1/ measurement of the capture in the broadest resonances at Van de Graaf accelerators with a new method for discrimination against quasi prompt capture of resonance-scattered neutrons ; that should give more confidence in the capture width of s wave resonances ; 2/ high resolution scattering measurements in order to establish p and d wave level spins for the calculation of temperature dependent self - shielding factors - The same recommendations are still present in the conclusions of the eleventh INDC meeting (june 1970)(8). In october 1981, at the twelfth INDC meeting, it was concluded in the status of the discrepancy list that : "it may be hoped that the new data, once completely analysed and the evaluations in progress at ORNL, BOLOGNA, JAPAN and CADARACHE, help to define the capture cross sections of Ni with the requested accuracies up to 300 kev or higher", the requested accuracy being 8 % on capture. The new data are those above mentioned (HARWELL and OAK-RIDGE). A preliminary evaluation has been achieved at CADARACHE for  $^{58}\text{Ni}$  and  $^{60}\text{Ni}$  (10), but the requested accuracy is not obtained.

Therefore, I will conclude in the following way :

1/ it is not possible, at the present time, to calculate the Ni capture cross sections from the available resonance parameters with the requested accuracies,

2/ recommendations on further high resolution transmission experiments cannot be made before the complete analysis of the HARWELL and OAK-RIDGE experimental data,

3/ concerning capture and scattering measurements the recommendations of the 1979 INDC discrepancy file should be maintained.

4/ the recommendation by MOXON (1) on attempts "to measure the total width of some of the narrow resonances observed in the capture to try to confirm the radiation width required to fit the average capture cross section" at high energy, is still valid.

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- 4/ D.B. SYME et al., idem, page 703
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- 6/ D.B. SYME et al. Proceedings of HARWELL meeting, september 1978, page 319
- 7/ INDC Discrepancy file 1979, INDC-32/L
- 8/ Minutes of the eleventh INDC meeting, INDC-35/L
- 9/ Minutes of the twelfth INDC meeting, 37/L
- 10/ H. DERRIEN, not published

H. Derrien

C.E.N. Cadarache

December 1982

## The $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$ reaction.

### A.) Significance

Because of its low threshold (30 keV) and long half-life (~15 years) the  $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$  reaction is potentially an excellent long-term fast neutron fluence monitor especially for radiation damage studies.

### B.) Status

The present status of our knowledge of the excitation function of the  $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$  reaction is still unsatisfactory. Activation cross-section measurements for monoenergetic neutrons exist to date only for 14 MeV /1/. Some information on the  $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$  cross-section in the neutron energy range 0.8 - 2.7 MeV can be derived from  $(nn')$  measurements by summing up the gamma production cross-sections for all transitions populating the isomer. However the existing data /2 - 5/ strongly disagree within each other and also with the existing data on the  $^{93}\text{Nb}$  nonelastic cross-sections derived from sphere-transmission /6/ and  $(n,n')$  measurements /7 - 9/. Thus at best cross-section values accurate to about 30% are derivable from this information.

Thus at present it seems, that the most accurate method of prediction of this cross-section are nuclear reaction calculations based on the statistical model of nuclear reactions including preequilibrium particle emission.

Calculations of this kind have been performed 1980 at the Institut für Radiumforschung und Kernphysik. An evaluation of the  $^{93}\text{Nb}(n,n')^{93\text{m}}\text{Nb}$  cross-section up to 20 MeV based largely on the above calculations has been published in Physics Data by Fachinformationzentrum Karlsruhe, Germany and included in the International Neutron Dosimetry File /10/. The results of this evaluation has been confirmed in the meantime by 3 different measurements.

- 1) The non-elastic cross-section assumed for the model calculations has been confirmed by ANL measurements /11/.
- 2) The activation measurement at 14 MeV already mentioned, which was done after the completion of ref. /10/, is in excellent agreement with the predictions of the evaluation.
- 3) The results of the evaluation /10/ have been tested in integral spectra and excellent agreement between calculated and measured reaction rates (ratio .9 - 1.) were found /12/.

Thus the results of ref. /10/ are certainly accurate within their rather large stated uncertainties, for energies above 10 MeV the actual uncertainties are probably less than the tabulated values of ref. 10 by at least a factor of 2.

#### C.) Current work

Measurements of the  $^{93m}\text{Nb}$  activation cross-section, with mono-energetic neutrons have been started by Dr. Uttley, Harwell for  $E_n = 4 - 6$  MeV and will be extended to the 1 - 2 MeV range; results are expected before the end of 1983. At the IRK Vienna a measurement of the activation cross-section is planned for  $E_n = 2.6$  MeV. Subsequently the IRK evaluation will be updated.

#### D.) Conclusion and Recommendation

The activation measurements described should be supplemented by new accurate (nn' $\gamma$ ) measurements in the energy range .8 - 3.0 MeV.

Inclusion of such (nn' $\gamma$ ) data and the new activation measurements mentioned in the evaluation of ref. 10 should then allow the prediction of the whole excitation curve with an accuracy of 10-15%.

#### Literature

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- /4/ V.C. Rogers, L.E. Beghian and F.M. Clikeman, Nucl. Sci. Eng. 45 (1971) 297
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- /7/ A.B. Smith et al., Z. Physik 264 (1973) 379
- /8/ D. Reitmann et al., Nucl. Phys. 48 (1963) 593
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- /11/ A.B. Smith, P.T. Guenther and J.F. Whalen, ANL/TDM-70 (1982)
- /12/ K. Sakurai, NEANDC(J)-83/U, p. 37

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January 1983

$^{232}\text{Th}$  Fast Neutron Fission

Fission cross-section results are now available from the following work:

- (1) ORNL 0.1 - 1.6 MeV (a)
- (2) ORNL/LANL 0.9 - 10 MeV (b)
- (3) LLNL 0.7 - 30 MeV (c)

Olsen presented a summary of cross-sections for the thorium cycle at the Meeting in Japan in October, 1982 (d). He states that the ORELA data from experiments (1) and (2) compare well in the overlap region of 0.9 to 1.6 MeV.

- (a) Perez, et al., Phys. Rev. C 28 (1983) 1635.
- (b) Auchampaugh et al., Phys. Rev. C 24 (1981) 503.
- (c) Behrens, et al., Nucl. Sci. Eng. 81 (1982) 512.
- (d) Olsen D. K., "ORELA Contribution to Thorium Cycle Nuclear Data", US-Japan Seminar on the Thorium Fuel Cycle, Nara, Japan, October 1982, CONF-821082-2.

H. T. Motz

LANL

June 1983

## Fission Cross-section and Fission Cross-section Ratios of $^{233}\text{U}$ ( $E \geq 100$ keV)

Since the last entry in the NEANDC Discrepancy File (NEANDC 124A) a few new experimental data appeared which were mainly presented at the 1980 Kiev Conference.

Zhagrov et al.<sup>(1)</sup> (energy range of interest:  $4.6 \cdot 10^4$  eV -  $1.3 \cdot 10^5$  eV). There is one experimental result at  $120 \pm 9$  keV relative to  $^{235}\text{U}(n,f)$  cross-sections. Neutron flux has been measured by the  $\text{MnSO}_4$  bath method. The measured value for fission cross-section of  $^{235}\text{U}$  is  $1.51 \pm 0.06$  b (to be compared with 1.52 b in ENDF-BV) while the corresponding value for  $^{233}\text{U}$  fission cross-section is  $2.15 \pm 0.09$  b. This information is significant since it confirms the low values of Carlson and Behrens<sup>(8)</sup> in an energy range where the experimental data spread is over 10%.

Shpak et al.<sup>(2)</sup> (energy range: 0.06 MeV - 3.28 MeV). The fission cross-section ratio has been determined versus energy according to a classical technique in which neutron fluxes are monitored by glass detectors. Neutrons were produced from  $\text{T}(p,n)$  and  $^7\text{Li}(p,n)$  reactions by electrostatic accelerator. The values confirm quite perfectly those presented by Fursov et al.<sup>(3)</sup> in 1977 at the Fourth Kiev Conference.

Mostovoya et al.<sup>(4)</sup> (energy range: 0.1 keV - 2 MeV). The fission cross-section of  $^{233}\text{U}$  and  $^{235}\text{U}$  have been separately measured from 0.1 keV up to 100 keV. The measurements up to 2 MeV concern the ratio.

The experiment has been performed on a 60 MeV linac using the time-of-flight technique.  $^{233}\text{U}$  and  $^{235}\text{U}$  foils were placed back-to-back in a fission chamber which was protected against backscattered neutrons by Boron and against gamma flux by 60 cm of lead. Neutron background flux (small: 1.5% for  $E > 10$  keV) has been determined by the technique of the saturated resonance (Ag, Co, Mn).  $^{235}\text{U}$  fission cross-section has been normalised on the value of 13.704 b in the interval 0.2 - 1 keV while for  $^{233}\text{U}$  the normalisation has been performed on the value of 32.25 b given by Gwin<sup>(5)</sup> for the resonance integral in the interval 166.9 eV - 1233.3 eV. The ratio values have been normalized in the interval 1 - 2 MeV to an average value of 1.523. The fission cross-section data are consistent with those of Gwin (within 2%) except in the interval (10 - 20 keV) where the discrepancy reaches 5%. At 100 keV for  $^{235}\text{U}$   $\sigma_{n,f} = 1.51 \pm 0.01$  b and for  $^{233}\text{U}$   $\sigma_{n,f} = 2.28 \pm 0.05$  b. Above, the ratio

values are consistent with those of Behrens<sup>(6)</sup>, James<sup>(7)</sup> and Carlson<sup>(8)</sup> within 2% with a few exceptions of disagreements by 4%.

In addition the following work is in progress:

- 1) At the Technical University of Dresden by Arlt et al.<sup>(9)</sup> at  $E_n = 14.7$  MeV using the associated charged particle method.
- 2) At Geel by Wagemans et al.<sup>(10)</sup> (with GELINA) who measured  $^{233}\text{U}(n,f)$  cross-section relatively to  $^{10}\text{B}(n,\alpha)$  with time-of-flight method in the energy range: thermal energy - 100 keV.

Unfortunately there are no data given for these experiments.

### Conclusions and recommendations

Some improvements in ratio data appear to have been obtained with an accumulation of lower values in the energy range 0.4 to 2 MeV.

Below 400 keV, an energy range characterised by strong fluctuations in both ratio and cross-section data, the data presented here show a tendency towards lower values (cf. data at 100 keV), but there are still unacceptable discrepancies (over 5%). Since  $^{10}\text{B}(n,\alpha)$  cross-section does not exhibit fluctuation it should be considered as a suitable standard for measurements in this energy range.

Now, there is an urgent need for a careful evaluation:

- to establish the state of the art
- to decide about the priorities for further measurements - which kind? (probably absolute measurements) and in which energy range? (probably preferentially below 400 keV)

The author is indebted to J. Gourdon for translation of the 1980 Kiev Conference proceedings at a moment's notice.

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- (2) Shpak et al., Kiev Conference proceedings (1980) 3, 35.
- (3) Fursov et al., Kiev Conference proceedings (1980) 3, 144.
- (4) Mostovoya et al., Kiev Conference proceedings (1980) 3, 30.
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CONCLUSIONS AND RECOMMENDATIONS OF WORKING GROUP ON THE U-235 FAST  
FISSION CROSS-SECTION

(IAEA Consultants Meeting on the Fast Fission Cross-section of U-235,  
Bratislava, March 1983)

1. INTRODUCTION

The meeting re-affirmed the importance of the U-235 fission cross-section as a primary standard in the energy range 100 keV to 20 MeV. The accuracy required was briefly discussed and the current uncertainty was established as a function of neutron energy. There was considerable discussion on the main contributions to the uncertainties in the measurements of the cross-section and on what steps might be taken to reduce those uncertainties. Lastly, the work which must be carried out to allow the cross-section to be determined to within the required accuracy was identified.

2. ACCURACY REQUIREMENTS

It was agreed that the use of the U-235 fast fission cross-section as a primary standard is the main reason for requiring high accuracy and it was felt that the objective should be an uncertainty of  $\pm 1\%$  (1 standard deviation) over the energy range 100 keV to 20 MeV. Such a value will ensure that the contribution from this standard to the overall error in any measurement should be very small. In addition, the U-235 fission cross-section is used as a 'test-bed' against which new techniques are tried and tested. If measurements are unable to produce accurate values for the favourable case of the U-235 fission cross-section, what hope is there of obtaining high quality data for unfavourable nuclides with, for example, high alpha-activity or low cross-sections?

Besides its importance as a standard, the U-235 fast fission cross-section has application in the fast reactor field. For that purpose, requests in WRENDAs are typically seeking an accuracy of between 1 and 2% up to  $\sim 5$  MeV, and above that energy an uncertainty of  $\gtrsim 2\%$  is probably acceptable. If an accuracy of 1% for a standard can be achieved over the energy range 100 keV to 20 MeV, all of the requests for U-235 fast fission cross-sections in WRENDAs would be satisfied.

The nuclear data needs for fusion are not yet sufficiently detailed to require the U-235 high energy fission cross-section to a high accuracy, although the situation may change in the course of time.

### 3. CURRENT UNCERTAINTIES IN THE U-235 FAST FISSION CROSS-SECTION

An accurate estimate of the uncertainties in any cross-section can only be obtained by detailed evaluation of the appropriate measurements and therefore it is quite clear that the present meeting could do no more than make an intelligent estimate of the accuracy to which the U-235 fast fission cross-section is currently known. Rather than quoting accuracies in energy intervals, which leads to apparent discontinuities in the uncertainties, it was decided to specify values at specific energies from which the magnitudes at other energies can be inferred by a smooth interpolation. The meeting felt that the following uncertainties apply at the present time.

| $E_n$<br>(MeV) | $\frac{\Delta\sigma_{nf}}{\sigma_{nf}}$<br>(%) |
|----------------|--|
| 0.1            | 2-3  |
| 1.0            | 2-3  |
| 3.0            | 2-3  |
| 5.0            | 3-4  |
| 8.0            | 3-4  |
| 13             | 4  |
| 14             | 1.0  |
| 15             | 2  |
| 20             | 6  |

However, one participant felt that the cross-section is known to 1-2% over much of the energy range below 13 MeV.

The region giving most cause for concern at the present time is that from ~3 to 6 MeV. Here, the data tend to divide into two groups, one having a convex shape and high cross-section values, and the other with a concave shape and low values. (See, for example, Fig. 3 of the contribution by Bhat). The two groups differ by up to ~10% although the contributing measurements claim accuracies in some cases of ~2%. During the meeting, some reasons

were put forward for doubting the accuracy of certain measurements (see the paper by Gayther and Patrick) and those reasons might lead to a downweighting of the data from those measurements.

There was some slight concern about the uncertainty in the region of 600 keV, where it was felt that the spread in the data might warrant a larger uncertainty than quoted in the above table.

It is encouraging to note that there appears to be very good agreement among the 'modern' (i.e. post 1975) 14 MeV measurements. However, this apparent strength must be viewed with caution as all of the measurements have employed the same technique, namely the time correlated associated particle (TCAP) method, and there may be unknown systematic errors present.

It was noted that the U-235 cross-section averaged over the Cf-252 fission neutron spectrum is very insensitive to the shape of that spectrum and therefore such measurements provide a useful normalisation value for shape measurements and evaluations. At the present time, the most accurate measurements are in agreement within the experimental uncertainties of 1.5-2%.

#### 4. MAIN CONTRIBUTIONS TO UNCERTAINTIES IN MEASUREMENTS

It was clear from the discussions that, as experimental techniques are examined more critically, and, as a result, refined, more and more of the problems of making accurate measurements are being understood. As a consequence, a comparison of older to newer measurements might appear to indicate that little progress has been made, but this may only be because the older measurements may have been too optimistic in their error estimates. There are clear signs that the main problem areas are now recognised and being tackled with vigour. These areas must be divided into two main components, covering problems arising from fission counting and those from neutron flux determination.

On the fission counting side of any measurement, the following are the problem areas.

(a) Extrapolation of the fission fragment pulse height spectrum to zero energy

In a well-designed experiment, the extrapolation to zero pulse height introduces a correction of  $\sim 1-1.5\%$ , and in most cases, it is assumed that

this extrapolation is linear and has a constant magnitude equal to the measured pulse height spectrum in the region just above the bias. But is this assumption justified or could the neglect of the ionisation defect cause undetected systematic errors? More work needs to be done to understand these effects and also more attention paid to the electronic methods used in processing the signals from fission chambers.

(b) Loss of fission fragments

Corrections for loss of fragments require a knowledge of the angular distribution as a function of the neutron energy and of the range of fragments in the fissionable material used. In the case of U-235, the uncertainty in the range may have a significant effect and may be more of a problem than any inaccuracies in the angular distributions. More work needs to be done to understand the effect of surface conditions (of both the backing and the deposit) on the loss of fission fragments. Direct measurements of fission chamber efficiency might be the best way of solving problems (a) and (b).

(c) Sample assay

Considerable attention is currently being focussed on the problems of sample assay and the international intercomparison of the mass determinations of fission foils should produce very valuable results. Of the main methods employed, comparisons of fission counting in a thermal field relative to very accurately assayed foils, seem to lead to more accurate results than alpha-particle assay. Further details of the results of the international intercomparison are to be found in the Appendix.

For the TCAP method, however, the areal density, rather than the total mass, is the important quantity. In this case, the alpha-counting technique using at least two apertures seems to be favourable.

Turning to the neutron counting aspect of a typical experiment, it appears that the most accurate method currently in use is the TCAP method. Problem areas which could lead to systematic errors are:

- (i) inscattering through large angles of the associated particle. Estimations for the worst case of the 2.6 MeV TCAP measurements on a neutron generator suggest that this effect could lead to an error  $\sim 0.5\%$ .

- (ii) the shape of the neutron cone corresponding to the associated particle acceptance. Scattering effects need to be carefully calculated and verified by direct measurement.
- (iii) pile-up and count loss corrections. As high counting rates are employed, very careful attention needs to be paid to these effects if undetermined systematic errors are to be avoided.
- (iv) high sample uniformity. Sample non-uniformity has to be carefully measured and taken into account.

The meeting also discussed the errors arising out of attempting to produce accurate cross-sections up to high energy by successive normalisations of essentially independent measurements, beginning at thermal energy. It was concluded that this is very unlikely to lead to accurate values due to the fact that the statistical uncertainty at each normalisation region becomes a systematic error in the succeeding values and these errors add up to produce large uncertainties at high energy.

It must also be remembered that problems may arise in the application of a standard. For example, the measurement of the response of a detector using the TCAP method is largely unaffected by scattered neutrons. However, if this detector is then used in a different experimental set-up, corrections for scattering may be required.

#### 5. WHAT CAN BE DONE TO REDUCE THE UNCERTAINTIES IN MEASUREMENTS?

There is no doubt that the problems identified in the previous section must be solved if we are to reach the 1% goal which seems desirable. Currently, new measurements are in progress or planned in a number of laboratories, in particular at the National Bureau of Standards, Argonne National Laboratory, Khlopin Radium Institute and at the Technical University of Dresden. The following suggestions for methods, either currently in use or to be used, were put forward.

The inefficiency of fission chambers may be investigated by one or more of several possible techniques. For example, neutron fragment coincidence/anticoincidence measurements may be useful in determining missing events and hence shed light on the problem of extrapolating the fission fragment pulse height spectrum to zero pulse height, as well as on the absorption

problem. Another method, the angular distribution approach described in a contribution from Budtz-Jørgensen, may prove to be a very powerful tool for these purposes. Yet another approach may be the use of U-235/Cf-252 mixed source counting techniques.

The loss of fission fragments may also be determined by the application of thermal beams to low geometry fission counting with varying foil thicknesses, coupled with alpha-particle assay. Another method is to compare  $2\pi$  fission counting with low geometry counting, again using thermal fields.

There is a general need to investigate these problems and all possible sources of systematic error. It was suggested that this should be done through more international intercomparisons. Already such an intercomparison of fissile foil mass assay is in progress and a second round of intercomparisons of flux determinations has been started. This can only lead to improved methods and one suggestion for a possible new intercomparison would involve an exchange of fission chambers.

In the case of shape measurements done with white neutron spectra, the main points which require further investigation are the determination of the neutron energy scale at high energies and the backgrounds associated with the fission counting and flux measurements. The energy scale of experiments in which the flux is measured by detecting recoil protons in a silicon detector can be affected by a considerable time walk in the discriminator from which the timing is derived. The time walk may not be the same as that measured using an alpha-particle source (or a pulser) and simulating protons by altering the pulse height using an amplifier.

Background determination at high neutron energies (i.e. energies above the region where notch filters can be used) in time-of-flight measurements on white neutron sources has long been a problem. The methods used are sometimes rather indirect and of a "hand-waving" variety and more attention needs to be paid to this aspect.

Experiments on linear accelerators have to cope with the intense gamma-flash which precedes the arrival of the neutron burst and as a result they have to recover from the effect of the gamma-ray burst in sufficient time to record neutron reactions with full efficiency. In a typical case of a 50m flight path, this means recovering in less than  $1\mu\text{s}$ . It is essential for measurers to show that their equipment has properly recovered during the full duration of their counting period.

During the course of the meeting, it became apparent that there were significant differences in the fission fragment ranges adopted by different groups in making calculations of corrections. There appears to be no single compilation of this type of data and such a publication should be encouraged. Following that, an evaluation should be performed so that consistent values can be used by all groups and to identify where any further work is required. However, it has to be realised that ranges obtained elsewhere may not solve the problems for a particular measurer, as the chemical composition of the foil may not be well-known and surface effects as well as impurities may cause errors.

## 6. RECOMMENDATIONS

The need for full documentation of measurements was stressed; this is vital if evaluators are to be able to make proper assessments of data. If sufficient detail cannot be given in a journal, because of space restrictions, then it is recommended that more detailed laboratory reports should be issued.

It was agreed that there is essentially no value in producing more measurements of the fast fission cross-section of U-235 with accuracies in the range 2-3% or worse using established techniques. As we already have a number of such measurements, additional ones will contribute little towards the reduction of the uncertainties. However, this should not be interpreted as an attempt to stifle new and innovative techniques. On the contrary, new methods, which may provide independent determinations, are to be encouraged even if they produce accuracies in the 2-3% range.

It is clear that if higher accuracies are to be achieved, one must pay particular attention to understanding the properties of the fission and flux detectors and to the corrections which are applied (e.g. loss of fragments in fission detectors).

The only foreseeable way of improving the accuracy of the cross-section is to perform

- (a) accurate mono-energetic measurements (using the TCAP method at as many energies as possible) with the focus on discrepant regions, but also paying attention to lower neutron energies where the applied needs are greatest.

- (b) accurate shape measurements which can be used to determine the cross-section between the spot point data.

These recommendations are certainly not novel but that can hardly be surprising. However, that does not make them any less valid. There can be no substitute for very careful, detailed and thorough investigations of all possible sources of systematic errors, using a variety of techniques. Science is founded on questioning and this approach must be applied vigorously and nothing must be taken for granted. Given a concerted effort, the problems can be solved and the U-235 fission cross-section can be determined to an accuracy of  $\pm 1\%$  using existing experimental techniques.

## APPENDIX

### Results of Sample Comparison (W. P. Poenitz, Argonne National Laboratory, USA)

Absolute alpha-decay rates and relative fission ratios were determined for 15 samples from ANL, LANL, NBS, KRI, BRC, Harwell, and CBNM in measurements at ANL. Comparisons for the alpha-decay rates can be made for those samples for which such values have been stated or can be inferred from stated masses based on alpha counting:

| Sample  | Quoted, aps      | ANL, aps         | $\Delta$ , % |
|---------|------------------|------------------|--------------|
| NBS     | 50.89 $\pm$ 0.25 | 50.97 $\pm$ 0.13 | $\sim$ 0.1   |
| KRI VI  | 62.6 $\pm$ 2.0   | 62.94 $\pm$ 0.2  | $\sim$ 0.5   |
| KRI XV  | 74.4 $\pm$ 2.2   | 73.97 $\pm$ 0.2  | $\sim$ 0.6   |
| HAR A   | 911.7 $\pm$ 4.6* | 914.1 $\pm$ 3.2  | $\sim$ 0.3   |
| HAR B   | 915.8 $\pm$ 4.6* | 914.9 $\pm$ 3.2  | $\sim$ 0.1   |
| CBNM 33 | 477.6 $\pm$ 4.1  | 476.7 $\pm$ 1.2  | $\sim$ 0.2   |
| CBNM 36 | 979.4 $\pm$ 8.3  | 977.3 $\pm$ 2.5  | $\sim$ 0.2   |

\*subject to revision

The comparison of sample masses, which includes questions of fission fragment absorption, is shown here only for some selected samples. The quoted values are shown as well as those derived from the ratio determination and final masses where available:

| Sample  | Quoted, $\mu$ g                 | Av. Ratios x Quoted, $\mu$ g | $\Delta$ , % |
|---------|---------------------------------|------------------------------|--------------|
| ANL 5-1 | 834.6 $\pm$ 2.7                 | 834.2 $\pm$ 3.3              | 0.05         |
| LANL S1 | 298.7 $\pm$ 0.3                 | 298.6 $\pm$ 1.2              | 0.03         |
| NBS     | 228.5 $\pm$ 1.2                 | 228.6 $\pm$ 0.9              | 0.04         |
| KRI     | 758 $\pm$ 25<br>757.9 $\pm$ 7.6 | 760.2 $\pm$ 3.0              | 0.29<br>0.30 |
| CBNM 36 | 250.0 $\pm$ 0.4                 | 251.3 $\pm$ 1.0              | 0.52         |
| HAR A   | 343.4 $\pm$ 2.7*                | 346.6 $\pm$ 1.4*             | 0.93         |

\*subject to clarification of the units of the isotopic composition

## $^{238}\text{U}(\text{n}, \gamma)$ CROSS-SECTION BELOW 100 keV AND $^{238}\text{U}$ RESONANCE PARAMETERS

### DESCRIPTION

The  $^{238}\text{U}$  capture cross-section and resonance parameters are of major importance for the calculation of performance parameters of thermal and fast reactors, such as the effective multiplication constants, the breeding ratio as well as the Doppler coefficient of reactivity.

In most recent evaluations the  $^{238}\text{U}$  cross-sections are represented by resolved resonance parameters up to about 4 keV and by unresolved (statistical) parameters above 4 keV, up to 45 keV in ENDF/B-IV and up to 149 keV in ENDF/B-V.

The resolved resonance parameters are obtained by a consistent analysis of transmission, self-indication, capture, and scattering high-resolution measurements in conjunction with theoretical models of statistical properties and whatever other information may be available on the properties of specific resonances.

The unresolved parameters in ENDF/B versions IV and V were generated by using "conventional values" for the average s-wave parameters and adjusting the average p-wave neutron width to "fit" evaluated average capture and inelastic-scattering cross-sections. In this procedure the average p-wave neutron widths are redefined every few hundred eV.

## STATUS

### I. Thermal Range (below 1 eV)

Some 10 measurements have been reported of the  $^{238}\text{U}$  capture cross-section at 0.0253 eV. All the measurements quote errors larger than 1% except the following two:

Bigham, Durham and Ungrin<sup>1</sup> (1969)  $2.721 \pm 0.016$  b

Poenitz, Fawcett and Smith<sup>2</sup> (1981)  $2.680 \pm 0.019$  b

The measurement of Poenitz et al., was relative to the  $^{197}\text{Au}$  thermal capture cross-section for which the current value  $98.65 \pm 0.09$  b evaluated by Holden<sup>3</sup> was used. The measurement of Bigham et al. was relative to  $^{235}\text{U}$  for which an absorption cross-section averaged over a 296K Maxwellian,  $g_a \sigma_a = .9771 * 679.9 = 664.3$  b was used. If the current value of the  $^{235}\text{U}$  absorption cross-section is used (Stehn et al.<sup>4</sup>:  $g_a \sigma_a = 680.4 \pm 1.4$  b) then the result of Bigham et al. becomes 2.792 b. A resolution of the discrepancy is desirable (both measurements are clearly described) since the thermal capture cross-section of  $^{238}\text{U}$  impacts strongly the calculated initial conversion rate in thermal reactors, and the epithermal to thermal ratio  $\rho^{28}$  is often used as a benchmark.<sup>5</sup>

There is no published accurate measurement of the shape of the  $^{238}\text{U}$  capture cross-section below 0.1 eV. This shape is sometimes assumed to be  $v^{-1}$ , but this assumption appears to lead to an incorrect calculation of the moderator temperature coefficient of reactivity in thermal lattices. This issue has recently been reviewed by Bouchard et al.,<sup>6</sup> who recommend that high accuracy measurements, particularly below 0.01 eV be attempted.

## II. Parameters of the First Few Large Resonances

Neutron capture in the first few large levels dominates the  $^{238}\text{U}$  resonance capture in thermal reactors.<sup>7</sup> Designers have requested accuracies of 1 meV in the capture widths and 2, 3 and 5%, respectively, in the neutron widths of the first three large resonances.<sup>8</sup> The spread among values obtained in recent measurements suggests that these accuracies have been met.<sup>9,10</sup> Calculations of resonance absorption in thermal lattices are now in reasonable agreement with the result of direct measurements.<sup>5,7,11</sup>

Though the spread among results from recent measurements is small compared to the accuracies needed for reactor design, there remain discrepancies among these results which are large compared to the small estimated uncertainties of the measurements. In particular, the neutron widths of the 20.9 eV and 36.8 eV resonances obtained from the RPI self-indication data<sup>12</sup> are 4 to 5 standard deviations smaller than the widths obtained from the other, mostly transmission data. These discrepancies imply unrecognized systematic errors which should be further investigated.

A covariance matrix describing the estimated uncertainties in the evaluated parameters of the large levels below 100 eV and their correlations has been requested by designers.<sup>8</sup>

## III. Resolved Resonances

An accurate knowledge of the parameters of the important resonances up to several tens keV is desired for the calculation of the Doppler coefficient of reactivity and other safety parameters in fast reactors.<sup>13,14,15</sup>

In the past ten years or so several laboratories have undertaken major programs of measurement of the  $^{238}\text{U}$  resonance parameters in the keV energy range.<sup>16-20</sup> Neutron widths have been published up to 6 keV. Up to 1.5 keV the values of the neutron widths obtained from the different experiments are consistent to within a few percent. Above 1.5 keV the results show systematic differences which increase with energy and are of the order of 10 to 20% above 3 keV. This is illustrated in Fig. 1 where values of the local s-wave strength function obtained from five sets of data are compared. A great deal of effort was expended in trying to understand the sources of systematic discrepancies between the various data sets.<sup>10,22,23</sup> The present consensus appears to be that a resolution of the discrepancies will require a general review of the methods of resonance analysis in the keV region, with particular attention to a realistic estimate of the systematic errors. Coates et al. have provided a particularly pertinent discussion of the problems.<sup>24</sup>

#### IV. Unresolved Resonance Parameters

Several authors have discussed the inadequacy of the present treatment of the  $^{238}\text{U}$  unresolved range in several major evaluations and have recommended an extension of the resolved range treatment to at least 10 keV.<sup>23,25,26</sup>

#### V. The C/E Discrepancy of $^{28}\text{c}/^{49}\text{f}$ in Fast Assembly

The persistent overprediction by a few percents of the  $^{238}\text{U}$  capture rate,  $^{28}\text{c}$ , relative to the  $^{239}\text{Pu}$  fission rate,  $^{49}\text{f}$ , in typical fast reactor spectra has been reduced but not eliminated by the latest U.S. evaluation, ENDF/B-V.<sup>27,28</sup> The discrepancy has often been

"blamed" on the  $^{238}\text{U}$  differential capture data because these data exhibit a spread which suggests relatively large uncertainties, yet the recent evaluations of the  $^{238}\text{U}$  capture cross-sections are already lower than most basic data and further lowering to resolve the discrepancy is not technically justified.<sup>29</sup> Recent work on this discrepancy suggests that it may result from several problems at the 2 to 3% level, with possibly also the  $^{239}\text{Pu}$  data and the direct measurements and their representation.<sup>30,31</sup>

Measurements of the  $^{238}\text{U}$  dilute capture cross-section in the range 1 to 100 keV and with accuracy of 3% or better have been requested.<sup>8</sup> This accuracy may not be achievable, particularly below 20 keV, until the parameters of the main resonances in the range 4 to 20 keV become better known, to permit a reliable calculation of resonance self-shielding and multiple scattering corrections to the measurements.<sup>32</sup>

#### SUMMARY AND RECOMMENDATIONS

1. The discrepancy between the  $^{238}\text{U}$  thermal capture cross-section measurements of Bigham et al.<sup>1</sup> and Poenitz et al.<sup>2</sup> needs to be resolved. This might be accomplished by an update of the corrections in the measurement of Bigham et al., which is well documented. If not, an additional accurate measurement may be desirable.
2. Existing data on the  $^{238}\text{U}$  capture cross-section shape below 0.1 eV should be documented (including a table of typical values of  $\sigma_{n\gamma}(E)$  with uncertainties.) Accurate measurements have been

requested:<sup>6</sup> accuracy of 10% or or better at 0.01 eV or below.

3. Efforts to resolve the systematic discrepancies in the values of the neutron widths above 1.5 keV, obtained from several recent experiments, should be pursued. The suggested approach is a systematic investigation of the sensitivity of the data to experimental corrections (background, resolution, etc.) and analysis assumptions (multilevel effects, resonance overlap, truncations, etc.)<sup>24</sup>
4. The extension of the resolved resonance range, at least for the major resonances, from 4 to 10 keV has been recommended.<sup>8,25</sup>
5. Dilute capture cross-section measurements to an accuracy of at least 3%, over the range 1 to 100 keV have been requested.<sup>8</sup> The interpretation of such measurements, at least below 20 keV, will probably require a better knowledge of the resonance structure.

Note: The present discussion is confined to the resonance regions.

The issues in the energy-averaged region (above 45 keV) have been discussed in a recent review of the <sup>238</sup>U data.<sup>9</sup>

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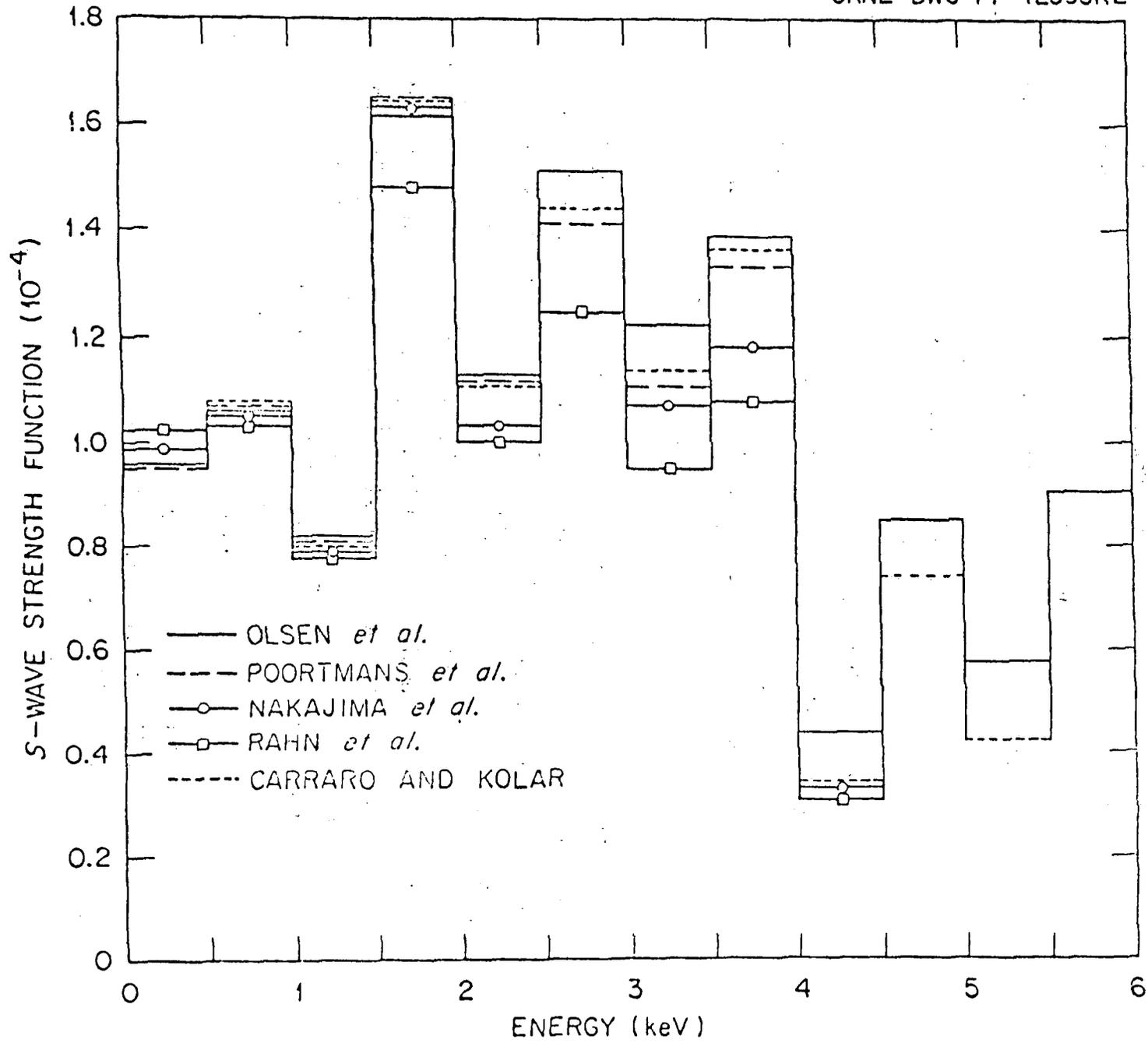
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## Present Status of $^{237}\text{Np}(n,2n)$ Cross-section for $^{236}\text{Pu}$ Formation

At the Antwerp Conference Wiese et al.<sup>(1)</sup> and Fort et al.<sup>(2)</sup> presented papers on the  $^{237}\text{Np}(n,2n)$  cross-sections in which the KEDAK and French evaluations were compared with integral measurements. The two papers essentially considered the same 3 steps in the evaluation of the  $^{236}\text{Pu}$  production following the (n,2n) reaction:

- Calculation of the cross-section for the (n,2n) process (Step 1).
- Evaluation of the fraction of the (n,2n) process giving rise to  $^{236}\text{Pu}$  formation. Let us call R this fraction. (Step 2).
- Analysis of integral data using the evaluated cross-section of  $^{236}\text{Pu}$  formation. (Step 3).

However, they differ at each step and this leads to a factor 2 discrepancy in the  $^{236}\text{Pu}$  formation cross-section.

When the two Antwerp Conference papers were written it was believed that the published data of Nishi et al.<sup>(3)</sup> had to be reduced by ~30% but it is now clear that the original data are correct<sup>(4)</sup>. This fortunately has little effect, except perhaps in Step 2 for Fort et al., and does not significantly alter the discrepancy.

### Step 1

There are no direct measurements of the (n,2n) cross-section. All the available experimental data are derived from the data for production of  $^{236}\text{Pu}$  obtained by the activation technique. With the exception of one datum at 9.6 MeV by Nishi<sup>(3)</sup>, they are located around 14 MeV and are in reasonably good agreement. The ratio ( $\frac{1}{R}$ ) to convert the  $^{236}\text{Pu}$  formation cross-section into the total (n,2n) cross-section has been experimentally found by Landrum et al.<sup>(5)</sup> to be equal to 2.76 from data obtained in thermonuclear neutron fluxes. The experimental data from Nishi, Lindeke<sup>(6)</sup>, Perkin<sup>(7)</sup> and Landrum<sup>(5)</sup> shown in Fig. 1 have been converted assuming that the value of R of Landrum et al. applies at all neutron energies.

Both evaluations agree very well above 12.5 MeV. At lower energies, KEDAK evaluation (performed by Caner et al. in 1977<sup>(8)</sup>) is higher by 15% to 40% (Fig. 1).

There are at least two reasons, taken into account in the French evaluations which are in favour of lower values:

- pre-equilibrium component
- constraints to reproduce fission probability data for fissioning  $^{237}\text{Np}$  and  $^{236}\text{Np}$  nuclei, which have been proved to play an important role.

### Step 2

In the German work R was assumed to be energy independent and in the calculations its experimental value at 14.5 MeV was considered. Patrick in a previous entry in the INDC Discrepancy File suggested R might depend on incident energy. If r stands for

$$\frac{\sigma[^{237}\text{Np}(n,2n) \longrightarrow ^{236}\text{Np}^m]}{\sigma[^{237}\text{Np}(n,2n) \longrightarrow ^{236}\text{Np}^g]}, \text{ R can be expressed as } R = \frac{0.48}{1+r} \text{ or}$$

$R = \frac{0.48r}{1+r}$  according to whether the short lived state ( $1^-$ ) is the ground state or the excited state. (In the following, these two situations will be labelled case 1 or case 2 respectively).

The direct or derived information concerning R (Fig. 2) is very scarce:

- Around 14 MeV, Landrum et al. have obtained  $R = \frac{1}{2.76} = 0.36$
- In 1972 Lecoq and Veyssiere measured the photoneutron emission cross-section of  $^{237}\text{Np}$  from threshold to 10 MeV with the Saclay Linac. (The data have been confidentially reported). The interpretation of experimental data has been performed in the context of case 1 and the authors concluded that the isomeric ratio was a smooth function of energy. At 9.6 MeV the derived value for R is  $R^{\text{exp}}(9.6) = 0.34$ .
- At 9.6 MeV also, an estimate of R deduced from the value of Nishi<sup>(3)</sup> and the French evaluation on one side is  $R_F^{\text{Cal}} = 0.43$ , and the KEDAK evaluation on the other side is  $R_G^{\text{Cal}} = 0.30$ .
- At threshold energy (6.8 MeV) the calculated value is  $R^{\text{C}}(6.8) = 0.48$  (case 1) or  $R^{\text{C}}(6.8) = 0$  (case 2). From these statements it is concluded that R, r and consequently isomeric ratio of  $^{236}\text{Np}$  are energy dependent.

### Step 3

The basic change with respect to the previous entry in the NEANDC Discrepancy File (May 1981) is the integral information<sup>(1)</sup> obtained from post-irradiation analysis of PWR TRIN02 pellets. The KEDAK evaluation and associated calculations result in a slight over-estimation of  $^{236}\text{Pu}$  content measured in TRIN02 pellets and give a value averaged over a fission spectrum for the  $^{236}\text{Pu}$  formation cross-section equal to 1.21 mb. The value obtained from the integral measurement of Paulson and Hennelly<sup>(9)</sup> is  $0.53 \pm 0.05$  mb while the evaluation of Fort et al. gives 0.51 mb. The fission spectrum averages of the total (n,2n) cross-section given by the two evaluations are 3.35 mb (KEDAK) and 1.97 mb (Fort et al.).

### Concluding remarks and recommendations

The two competing evaluations are discrepant by a factor 2, each being supported by integral information.

The origins of the discrepancy of these evaluated data are very clear:

- . Difference in the total (n,2n) cross-section below 12.5 MeV.
- . Difference in the adopted energy dependence of  $R(E)$ .

To solve these discrepancies it is recommended that (1) microscopic measurements be made, particularly below 12 MeV, of the partial cross-section for the formation of  $^{236}\text{Pu}$  by the activation method and of the total (n,2n) cross-section by the large liquid scintillator technique, and (2) improved calculations of  $R(E)$  should be made using the most sophisticated methods. For this it is essential that the level spectroscopy of  $^{236}\text{Np}$  is extended above the first two states whose parameters have recently been confirmed. The behaviour of the isomeric ratio with energy depends on an exact description of the excited level density. In particular for the energy range 1 to 1.5 MeV above threshold, which is crucial for practical applications, a complete and accurate description of the low lying levels (<1 MeV) is essential. In the absence of this information a preliminary calculation of  $R(E)$  can be made (the curves  $R_1(E)$  and  $R_2(E)$  in Fig. 2 are purely speculative) by assuming a statistical description of the level density of the

residual nucleus using parameters deduced from systematics.

Concerning the integral data the situation is by far more obscure.

It has to be kept in mind that the analysis of burned fuel is extremely difficult and requires highly qualified specialists. Often, the isotopes of interest are by-products of the irradiation, resulting from several competing channels whose relative importance has to be known. Shortly the difficulties are located at two levels:

- Accurate determination of the produced quantities of isotopes marking the various formation chains.
- Determination of the neutron flux along the period of irradiation (history of irradiation, evolution) and its description as a function of energy.

In reactor calculations the description of flux versus energy depends obviously on the data base but also on the calculational methods (collapsing of microscopic data into group constants, group structure scheme, upper energy limit...). The correctness of such description has to be carefully checked, especially the part above (n,2n) reaction threshold. A possible source of error is the evolution of the neutron spectrum during the irradiation period and resulting from actinide production which is treated by empirical methods (for example, parametrization of the one group cross-sections by the burn-up rate).

In light water reactors  $^{236}\text{Pu}$  formation is the result of the chains illustrated in Fig. 3.

In the measurement by Paulson and Hennely the  $^{237}\text{Np}(n,2n)$  cross-section was adjusted until the calculated value for the ratio of contents  $^{236}\text{Pu}/^{238}\text{Pu}$  agreed with the measurements. The other measured contents were well reproduced by calculations made with the burn-up code CASPER.

In the post-irradiation analysis of pellets from TRINO, the KEDAK  $^{237}\text{Np}(n,2n)$  cross-section was used to calculate and compare with the measured content in  $^{236}\text{Pu}$ . The correctness of calculations and data base were checked by comparing experimental and calculated values (by Korigen) for concentration of U, Pu, Am, Cm isotopes.

It appears clearly that integral experiment of separated isotope

sample irradiations are probably more appropriate for validating the microscopic information on the  $^{237}\text{Np}(n,2n)^{236}\text{Np} \xrightarrow{\beta^-} ^{236}\text{Pu}$  reaction.

The results, expected for 1983 of the PROFIL II experiment performed in PHENIX core are much awaited, in order to solve the discrepancy in the integral information. Another experiment is planned at KNK to give some additional information on the  $^{236}\text{Pu}$  formation cross-section.

The author is indebted to Dr. Antonio Giacometti for helpful comments on the techniques used in the burned fuel analysis.

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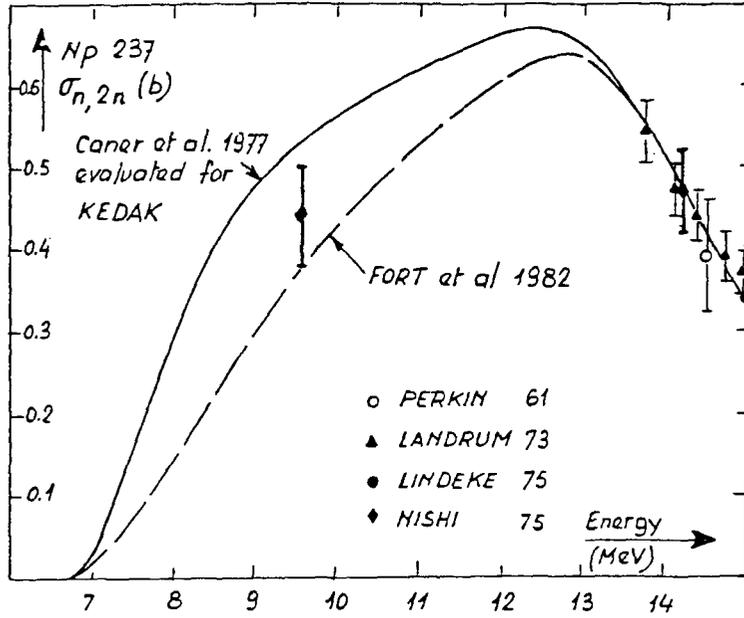
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November 1983

Fig. 1. Comparison of local s-wave strength function for 0.5 keV intervals for five sets of data.



EVALUATED AND MEASURED (n,2n) CROSS-SECTIONS FOR Np237

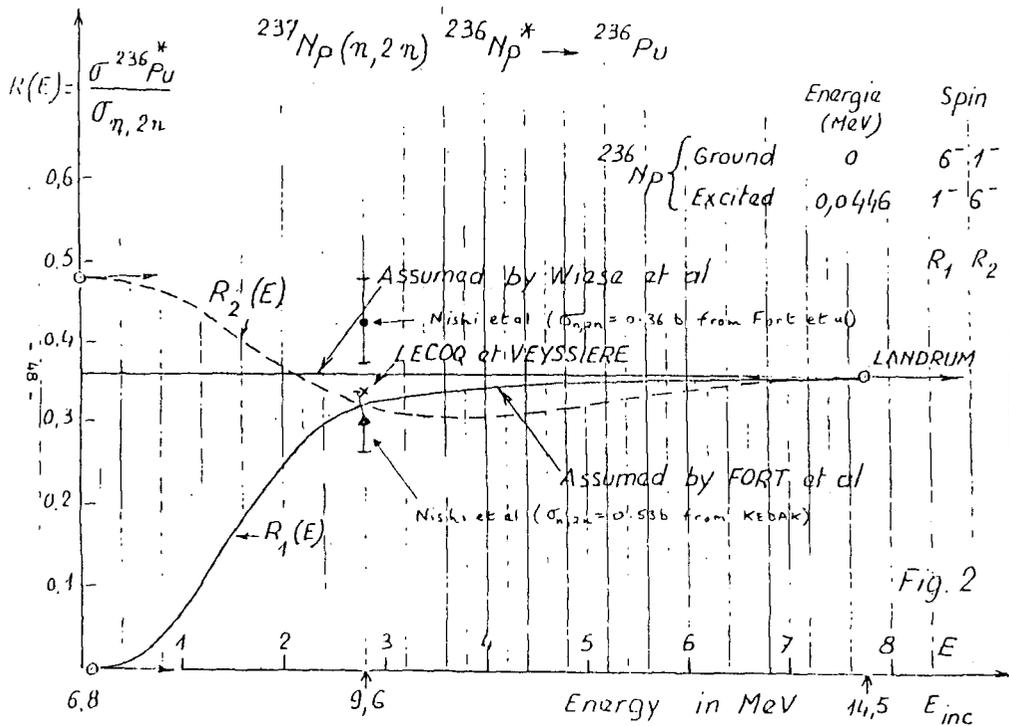


Fig. 2

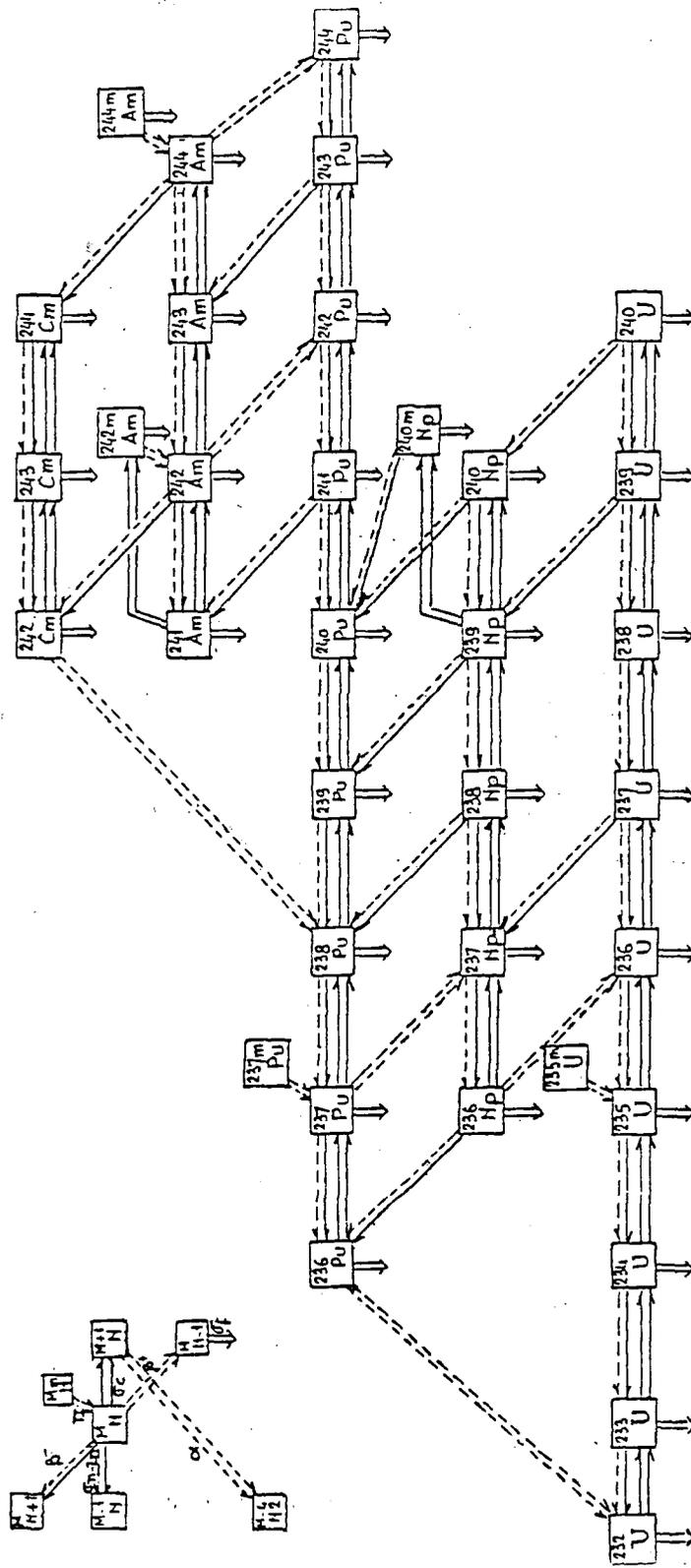


FIG 3

## $^{241}\text{Am}$ Fission Resonance Integral

A detailed account of the discrepancy between the measured and calculated values of the  $^{241}\text{Am}$  fission resonance integral is to be found in the "Review of Important Nuclear Data Discrepancies: An NEANDC Contribution to the INDC/NEANDC Discrepancy File", NEANDC 124A (May, 1980).

A measurement of the  $^{241}\text{Am}$  fission cross-section at ORELA, extending from 0.02 eV to 20 MeV, has recently been published by Dabbs et al. (Nucl. Sci. Eng. 83 (1983) 22). The fission resonance integral derived from this measurement is  $14.1 \pm 0.9$  barns for a cut-off energy of 0.5 eV. This value is significantly lower than results obtained from measurements in reactor (typically  $I_{nf} \sim 22 \pm$  barns) but still higher than values estimated from resonance parameter data (typically 8-10 barns). It appears therefore that there is still a discrepancy. It would be useful if Dabbs et al. could analyse their measurement to produce fission widths for a few resonances above 0.5 eV (assuming  $g\Gamma_n$  values obtained from published transmission measurements) so that they can be compared with existing values of  $\Gamma_f$ .

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A.E.R.E., Harwell  
May 1983

$^{235}\text{U}$

New experimental data

Since the 1979 publication of the NEANDC/INDC discrepancy file<sup>(1)</sup>, very little experimental works have been performed in the  $^{235}\text{U}$  resonance parameter field. One finds only one publication by Y.V. ADAMCHUK et al.<sup>(2)</sup> giving the alpha values for 19 resonances in the energy range 2-32 ev from the analysis of absolute alpha measurements by G.V. MURADYAN et al.<sup>(3)</sup>. The data were compared to those obtained by R. GWIN et al.<sup>(4)</sup> and a discrepancy of about 20 % was observed, MURADYAN et al. values being smaller than those from GWIN et al. Alpha measurements were also performed by F. CORVI et al.<sup>(5)</sup> in the energy range 6-140 ev and were used to normalize the measurements in higher energy ranges. F. CORVI et al. found some difficulties in this normalization using  $\Gamma_f$  and  $\Gamma_\gamma$  of some supposed well-isolated resonances, such normalization procedure being subject to uncertainties because different evaluations give quite discrepant values for resonance parameters, particularly for the radiation widths. As a matter of fact, it is quite difficult to find well-isolated resonances for  $^{235}\text{U}$ , as it is shown in reference<sup>(1)</sup>, and more difficult to find accurate values of  $\Gamma_\gamma$ . Nevertheless, the normalization coefficient obtained by F. CORVI from ENDF/B-V resonance parameters agrees with those obtained from other methods with 3.3 % accuracy. Finally, the  $\alpha$  values obtained by F. CORVI et al. are on average 10 % lower than those from R. GWIN et al. The accuracy claimed by Y.V. ADAMCHUK et al. is 2 % in the resonance region. According to F. CORVI, this accuracy is too optimistic with regard to the discrepancy existing between the three above mentioned experimental results.

The recent evaluations

The status of the most recent evaluations have been discussed at the consultant meeting on Uranium and Plutonium Isotope Resonance Parameters (Vienna, 1981). ENDF/B-V still contains the SMITH-YOUNG<sup>(6)</sup> parameter set and uses the single-level formalism

associated to a fluctuating background for the description of the cross-sections. The same procedure is used in JENDL-2<sup>(7)</sup> with a parameter set obtained by averaging all the available data and modified in the energy regions where the discrepancies between the calculated and the experimental cross-sections appear to be too large. SOKRATOR file contains the KONSHIN-ANTSIPOV evaluation<sup>(8)</sup>. The cross-sections are calculated using either the one-level Breit-Wigner formalism or a modified Adler-Adler formalism. Resonance spins are given in accordance with the assignments made by MOORE et al.<sup>(9)</sup>. However not all the resonances observed by MOORE et al. are included in SOKRATOR. There is not much information concerning KEDAK-IV evaluation<sup>(10)</sup>; the cross-sections are given point by point in the resonance region and 30 percent more resonances are used than in the three preceding evaluations, which would be in agreement with the results of MOORE et al.

A numerical comparison of these four evaluations has been made by D.E. CULLEN et al.<sup>(11)</sup> for group cross-sections integrated over a  $1/E$  spectrum. There are large differences between the calculated average cross-sections. One notes, in particular, abnormally high values for SOKRATOR in the 1.0 eV to 2.15 eV group. In the other groups the discrepancies may be as high as 25 percent. Such large differences should be avoided at least for the fission cross-sections. As a matter of fact, if one examines the experimental data published since 1972, one finds discrepancies not larger than 5 percent on the average cross-sections (see for example the table published by C. WAGEMANS et al. in reference 12). Then, it is obvious that some of the available evaluated data files do not calculate the fission cross-sections with an accuracy comparable to that achieved with the experimental results for the resonance region.

It is worthwhile to mention that the discrepancy observed in the calculated resonance self-shielded fission when compared to BRAMBLET and CZIRR measurements<sup>(13)</sup>, has been largely removed by the results of a new measurement performed recently by CZIRR<sup>(14)</sup>; the earlier measurements were not properly corrected for background.

### Works in progress

No new experimental resonance parameter data set is expected for the immediate future from new experiments. But new evaluations or new analysis of existing experimental data are planned especially for ENDF/B-VI. M. MOORE<sup>(15)</sup> reported, at the Consultant Meeting on Uranium and Plutonium isotope resonance parameters, on a preliminary result of a multilevel analysis of KEYWORTH et al.<sup>(16)</sup> spin-separated fission cross-sections. This multilevel analysis used two fission channels in each spin state and the uniqueness of the fit was ensured by some constraints from PATTENDEN-POSTMA<sup>(17)</sup> angular distribution data. The definitive analysis will include total and capture data in view of obtaining a coherent set of neutron widths, fission widths and capture widths. Work of this kind is also undertaken at Mass Transfer Institute (Minsk, USSR) by V.F. ZHARKOV et al.<sup>(18)</sup>.

<sup>239</sup>Pu

### New experimental data

As for <sup>235</sup>U one finds very little new informations on <sup>239</sup>Pu resonance parameters since 1979. Fission measurements have been performed by I. BERCEANU et al.<sup>(19)</sup> in the energy rang 0.01 ev to 0.72 ev. These data have been analysed with the single level Breit-Wigner formalism and the parameters for the five low energy resonances, including a negative level, were obtained. These data could be used for the cross-section evaluation in the thermal region. A new set of single level parameters was also obtained by V.A. KONSHIN et al.<sup>(20)</sup> from a least square shape analysis of thick sample transmission data with 100 ns/m resolution. It is unlikely that these results could be better than those obtained at Saclay several years ago<sup>(21)</sup> from transmission measurements with 0.5-16 ns/m resolution at liquid nitrogen temperature.

The recent fission measurements by C. WAGEMANS et al. <sup>(22)</sup> were not analysed in term of resonance parameters, the purpose of this experiment being to obtain accurate values of the absolute fission cross-sections.

### The recent evaluations

The status of the evaluations was also discussed at the Consultant Meeting of Uranium and Plutonium Isotope Resonance Parameters (Vienna, 1981). ENDF/B-V contains the evaluated parameters of J.R. SMITH et al. <sup>(23)</sup> ; the evaluation report has not been published. According to M. MOORE <sup>(15)</sup> this data set results from an incompleted analysis and new evaluation is foreseen for ENDF/B-VI. JENDL-2 is based on RIBON et al. evaluation <sup>(24)</sup> which is almost identical to the Saclay experimental single level set up to 660 ev. <sup>(21)</sup> In SOKRATOR file the resolved resonance parameters are also given up to 660 ev. Up to 500 ev a least square analysis fit using the one level Breit-Wigner formalism was carried out <sup>(25)</sup> for all the cross-sections for which experimental data are available. The results are similar (if not identical) to RIBON evaluation. Above 500 ev, the parameters are those from RIBON. None of these evaluations uses a multilevel formalism ; at best a very complex background is specified to account for interference effects.

Numeral comparisons have also been performed by D.E. CULLEN <sup>(11)</sup> in group cross-sections for ENDF/B, JENDL, SOKRATOR and KEDAK. The discrepancies between the calculated capture cross-sections fall in the range 10 % to 50 %. For fission cross-sections, the differences are 7 % on average in the energy interval 1 ev to 100 ev. As for <sup>235</sup>U, better agreement is found in the most recent fission experiments of J. BLONS et al. <sup>(26)</sup> , R. GWIN et al. <sup>(4)</sup> and C. WAGEMANS et al. <sup>(22)</sup> . At least, the average values of these three fission experiments should be reproduced in the fission evaluation ; an accuracy of 3 % could then be obtained.

In a preliminary evaluation carried out by SIMPSON and SIMPSON<sup>(27)</sup> an internal inconsistency was found in the description of the measured total and partial cross-sections. It seemed that this inconsistency was removed by taking into account the resonance-spin information<sup>(28)</sup>. According to M. MOORE<sup>(15)</sup>, the inconsistency is not completely removed in the not yet published SMITH et al. evaluation on which ENDF/B-V is based. SMITH et al. argued that, since the total cross-sections were obtained from transmissions using several sample thicknesses, problems could arise due to uncertainties in the knowledge of the number of atoms in the samples. This possibility has been discussed in reference 28 and it appears unlikely that the problems associated with the accuracy of the sample thicknesses could result in important errors in the Saclay total cross-sections. More should be learnt about this discrepancy, when the SMITH et al. evaluation is published. If the inconsistency is a real one, due to the total cross-sections, it is then more urgent to undertake a new precise transmission measurement to replace the old Saclay data.

#### Works in progress

A new evaluation is in progress at Cadarache. Below 200 eV, this evaluation is based on the Reich-Moore multilevel set of resonance parameters obtained in reference 29. In the energy range 200-660 eV, it will be essentially RIBON et al. evaluation<sup>(24)</sup>. The cross-sections will be calculated by using an optimized energy ladder at 0° temperature, allowing accurate calculation at any other temperature from an appropriate convolution procedure. Works are also under way for ENDF/B - VI<sup>(15)</sup>.

H. Derrien  
C.E.N. Cadarache  
May 1983

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## Comments on the Scandium Discrepancy

A number of very different values have been reported on the value of the total cross-section of scandium at a minimum near 2 keV. The value is needed for application of beam filters, and the value plays a role in the determination of the optimum filter length. The reason for the wide variation in reported values clearly lies in the problem of obtaining pure scandium. In the U.S., the scandium normally supplied is produced by sublimation on a Ta surface. Accordingly, there is a chance for a sizable Ta impurity to exist. Furthermore scandium is slightly reactive in the atmosphere, especially in humid conditions, when a hydroxide can be formed. Cu and Mn impurities in Sc have also been noted. The presence of small impurities can seriously affect the measured value, and when the impurities are sizable, make an accurate correction impossible. Most experimenters recognize the problem, but it is not easy to assess the presence of H and O atoms, and the possibility of sample deterioration with time is present.

The values that have been reported are tabulated below, as a function of chronological time.

The most recent values of Fujita and Yamamuro are in good agreement with recent USSR results. I understand that both the Japanese and Russians used scandium obtained from the same (Russian) source. The Japanese measurements, obtained by TOF at the KURRI Institute at Kyoto, agree well with Liou et al., except near the 2 keV minimum. The ORNL results of Harvey et al. were performed on samples of Sc + Ti (obtained from Los Alamos) and on  $\text{Sc}_2\text{O}_3$ .

It seems clear that the impurity problem plagues these measurements and that a check on the Russian-Japanese results would be useful. In any practical application, however, the impurity effects will dominate the transmission near 2 keV.

R. E. Chrien

Brookhaven National Laboratory  
October 1982

| $\sigma_{\min}$ (2 keV) (mb)              | Author  |
|---|---|
| $\approx 450$                             | Pattenden, Proc. Phys. Soc. A68 (1955)  |
| +100                                      |   |
| 50  | Wilson (unpublished thesis) (1966)  |
| -50                                       |   |
| 85 mb                                     | Magurno and Mughabghab (evaluation), Nuclear Cross Sections and Technology Conf., Vol. I, p. 357 (1975) |
| $710 \pm 30$ mb                           | Liou et al., Nucl. Sci. Eng. <u>67</u> , 326 (1978)   |
| $270 \pm 70$                              | Razbudey et al., Knoxville Conf. (1979) p.890   |
| $250 \pm 50$                              | Fujita and Yamamuro (1981), quoted in JAERI-M 9981  |
| $230 \pm 20$                              |   |
| $266 \pm 77$                              | Kirilyuk et al. (1981) PC from Vertebnyi  |
| $278 \pm 90$                              |   |
| $360 \pm 30$<br>(Corrected for Ti and Mn) | J. A. Harvey et al., Antwerp Conf. (1982) p.856   |
| $470 \pm 40$<br>(Corrected for O)         |   |
| $210 \pm 30$                              | Fujita Y., J. Nucl. Sci. and Tech. <u>20</u> (1983) 191   |

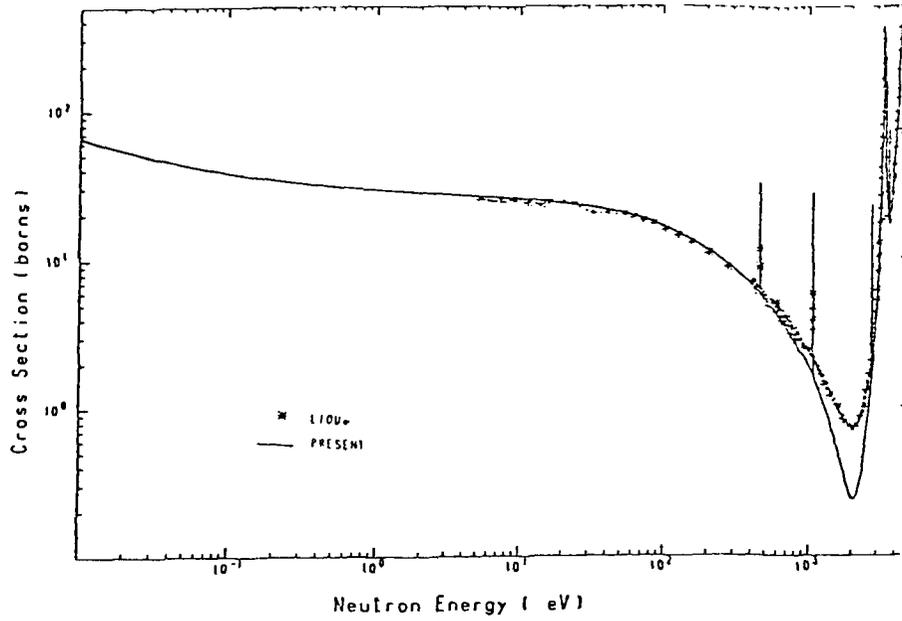


Fig. 1 The total cross-section of  $^{45}\text{Sc}$  calculated from the present resonance parameters with the measured data by Liou et al. in the energy range below 5 keV.

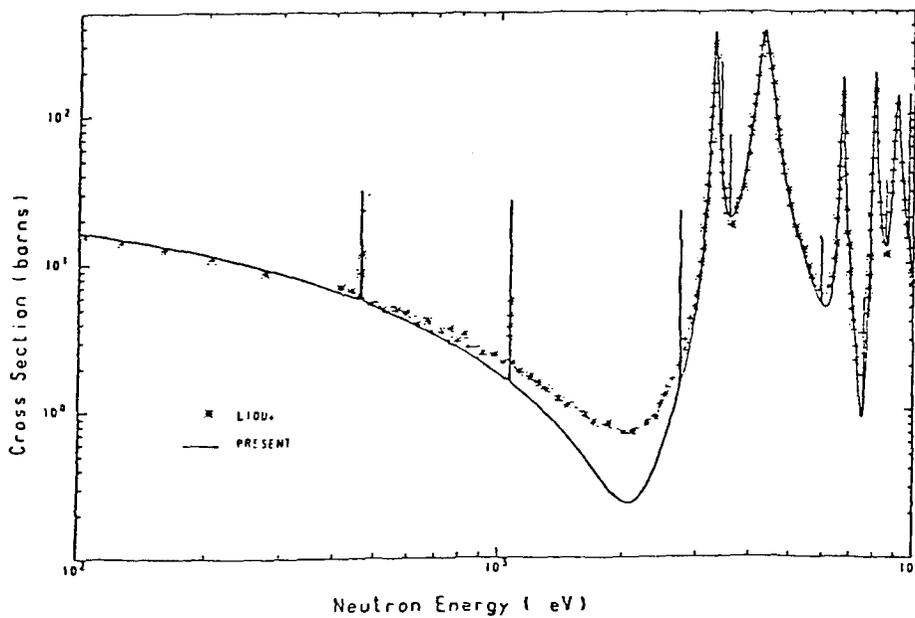


Fig. 2 The total cross-section of  $^{45}\text{Sc}$  calculated from the present resonance parameters with the measured data by Liou et al. in the energy range between 100 eV and 10 keV.

## DELAYED NEUTRONS

The following conclusions are largely extracted from the results of the IAEA Consultants' Meeting in Vienna, 26 - 30 March 1979<sup>1)</sup> and completed with results which have appeared in the literature 1979-1982.

Integral properties, i.e. those resulting from the bulk of fission products in nuclear fuel, and properties of individual products are treated separately.

### 1. Integral properties of delayed neutrons in nuclear fuel

#### 1.1 Delayed neutron yields

The most stringent requirements are for the interpretation of critical experiments which demand an accuracy of about  $\pm 2\%$  of the yields. We are close to this goal for  $^{235}\text{U}$  and  $^{238}\text{U}$ , but more work is needed especially for  $^{232}\text{Th}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$ . Moreover, the dependence of the yields on neutron energy should be better investigated.

Since the properties of individual delayed-neutron precursors, for instance the branching ratios, are often quite well known, the yield can be obtained not only by integral measurements but also by summing the contributions from the precursors. This might well be the best way to

obtain yields for the heavy plutonium isotopes and for isotopes of americium and still heavier elements. It must be borne in mind, however, that this procedure throws the difficulties over to the field of fission yields. The accuracy of the evaluation will strongly depend on how well the fission yield pattern is known.

## 1.2 Delayed-neutron energy spectra

Data on the time dependence of group spectra is sparse and should be improved by new measurements. As an alternative approach, especially for fissionable materials which are difficult to measure, group spectra can be constructed from individual precursor data. This method has been used in a recent study<sup>1)</sup> which provides data for the evaluation of the energy spectra of six half-life groups, and their time dependence, of a number of fissile materials.

## 2. Properties of individual delayed-neutron precursors

### 2.1 Branching ratios

There are a number of cases for which several determinations exist but where the values and errors given clearly indicate systematic deviations between different

laboratories and/or methods. The reason for such deviations should be tracked down and the measurements, if possible, corrected. The following precursors belong to this group:

$^{85}\text{As}$ ,  $^{88}\text{Se}$ ,  $^{91}\text{Br}$ ,  $^{97-99}\text{Sr}$ ,  $^{97-99}\text{Y}$ ,  $^{128-131}\text{In}$ ,  $^{138}\text{I}$ ,  $^{140}\text{I}$ ,  $^{141,142}\text{Cs}$  and  $^{147}\text{Ba}$ .

Occasionally one measurement for which a high accuracy is claimed deviates seriously from the bulk of data. This measurement will then have an appreciable influence on the mean value. In the first place one should try to explain the reason for the discrepancy. If this is not possible one may consider discarding the discrepant measurement when calculating the mean value of the branching ratio. Examples are:

$^{135}\text{Sb}$ ,  $^{137}\text{I}$ ,  $^{138}\text{I}$ ,  $^{144}\text{Cs}$ ,  $^{145}\text{Cs}$ ,  $^{147}\text{La}$  and  $^{148}\text{Ba}$ .

In certain cases only one experimental determination is available, which calls for at least one independent determination. To this group belong:

$^{79-83}\text{Ga}$ ,  $^{84}\text{As}$ ,  $^{87}\text{As}$ ,  $^{89}\text{Se}$ ,  $^{94}\text{Kr}$ ,  $^{121-124}\text{Ag}$ ,  $^{129\text{m}}\text{In}$ ,  $^{132}\text{In}$ ,  $^{134}\text{Sn}$ ,  $^{137}\text{Te}$ ,  $^{138}\text{Te}$  and  $^{147}\text{Cs}$ .

The  $P_n$ -determinations of the nuclides:  $^{85-87}\text{As}$ ,  $^{92}\text{Br}$ , and  $^{136}\text{Sb}$  are derived from neutron per fission determinations and estimated fission yields. This method should be regarded with caution because of the fine structure which may be present in the yield pattern. There is a need for

experimental determinations, either direct measurements of the branching ratios or experimental determinations of the fission yields.

In particular, we would like to stress the importance of removing discrepancies in branching ratio determinations for certain precursors with large fission yields, such as  $^{98}\text{Y}$ .

For a few precursors,  $^{83}\text{Ga}$ ,  $^{83}\text{Ge}$ ,  $^{84}\text{Ge}$ ,  $^{133}\text{Sn}$ , and  $^{143}\text{Xe}$ , the branching ratios still remain to be measured.

## 2.2 Energy spectra

Energy spectra of delayed-neutrons have been measured using different techniques. There is a definite disagreement between results obtained, however, especially between the  $^3\text{He}$ -spectrometer results and results obtained with a proton-recoil spectrometer. This problem has to be resolved. Also alternative techniques should be tried.

Most of the measured spectra extend from about 60-100 keV up to a few hundred keV below the upper limit. Both the low energy part and the high energy part are missing which means that complementary measurements should be carried out. Among the more important precursors to be further studied are  $^{88-91}\text{Br}$ ,  $^{139}\text{I}$ , and  $^{140}\text{I}$ .

No information about the neutron spectrum is available for the following group of precursors:  $^{92}\text{Br}$ ,  $^{93}\text{Kr}$ ,  $^{94}\text{Kr}$ ,  $^{98-100}\text{Sr}$ ,  $^{97-101}\text{Y}$ ,  $^{121-124}\text{Ag}$ ,  $^{137-39}\text{Te}$ ,  $^{141}\text{I}$ ,  $^{147,148}\text{Ba}$ ,  $^{147,148}\text{La}$ , and  $^{131,132}\text{In}$ .

It is important that the experimentalists evaluate the uncertainty in the measured spectra.

### 2.3 Mean neutron energies

If the spectrum is known the mean neutron energy can be evaluated. The mean energy can also be measured directly. This has been done for a few cases<sup>2)</sup>, and the work should be extended. Directly measured mean values are valuable as consistency checks on the spectra.

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## $^{91}\text{Zr}$ and $^{96}\text{Zr}$ Resonance Parameters

### Description

Because of its low thermal absorption cross-section and good mechanical and corrosion properties, Zirconium is used as cladding material in most of the existing pressurized water, boiling water and heavy water reactors.

A precise knowledge of resonance parameters is essential to reach a good approximation in calculating the performance of thermal reactors and the parameters of their operation as, for example, multiplication constant and Doppler coefficient. Most of neutron absorption occurs in the odd isotope  $^{91}\text{Zr}$ , having a natural abundance of 11.2%.

In the "High Priority Measurement Request List" prepared by NEANDC, total cross-section and resonance parameters of  $^{91}\text{Zr}$  and  $^{96}\text{Zr}$  are required with a typical accuracy of 10%. The resonance integral of  $^{91}\text{Zr}$  is required with an accuracy of 5%.

### Status

Extensive capture and transmission measurements both on  $^{91}\text{Zr}$  and  $^{96}\text{Zr}$  have been performed at the Oak Ridge linac in an Australian-U.S. co-operative work<sup>(1,2)</sup>. More recently, capture and transmission measurements on  $^{91}\text{Zr}$  and transmission on  $^{96}\text{Zr}$  were carried out at the Geel linac by a joint Italian-Euratom group<sup>(3,4)</sup>. The latter work included also the spin assignment of several  $^{91}\text{Zr}$  resonances by means of capture gamma-ray measurements. Capture measurements on  $^{96}\text{Zr}$  have successively been carried out at Geel, but the results have not yet been published.

### Discrepancies

#### $^{91}\text{Zr}$

The dominant contribution to the resonance integral is given by the 292 eV resonance. With respect to previous data<sup>(1,5)</sup>, the Geel measurements have definitely changed the spin assignment of this resonance. Moreover Geel gives higher  $2g\Gamma_n$  and  $\Gamma_\gamma$  values, such that, all in all,  $g\Gamma_n\Gamma_\gamma/\Gamma$  is almost doubled.

The  $\Gamma_n$  values of ref. 3 are systematically higher than in ref. 1; the average ratio is 1.33.

#### $^{96}\text{Zr}$

Since the natural abundance of  $^{96}\text{Zr}$  is only 2.8%, discrepancies in this isotope have smaller practical consequences. However, the very high systematic discrepancies between refs. 2 and 4 are an indication of possible errors also in other important data. The ratio of the  $\Gamma_n$  values given in ref. 4 over those of ref. 2 is, on average, 1.65.

The analysis of the capture measurements at Geel is not likely to modify substantially the data of ref. 4. In particular, preliminary results indicate that the resonance integral, which is dominated by the 301 eV resonance, should be left practically unchanged.

#### Comments

The authors of ref. 4 suggest that the systematic discrepancy might be produced by the different data analysis procedures employed at Oak Ridge and Bologna and work on  $^{91}\text{Zr}$  is going on at Bologna to explore this hypothesis. The resolution function of ORELA has now been calculated<sup>(6)</sup> by the method adopted in refs. 3 and 4 and using this the transmission data of Musgrove et al.<sup>(1)</sup> are being re-analyzed at Bologna employing the same code used for the analysis of the data obtained at Geel. The first analyzed resonances gave excellent fits and  $2g\Gamma_n$  values very close to those previously obtained by the Geel-Bologna collaboration.

The Bologna group have now modified their code to fit simultaneously transmission data having different resolution functions and different isotopic compositions.

The following table reports, for the first few resonances, the discrepant data and the preliminary results of the simultaneous fit of ORELA and Geel measurements.

| $E_0$  | $J^\pi ; 2g\Gamma_n$   |                       |               |
|--------|------------------------|-----------------------|---------------|
|        | Brusegan et al. (3)    | Musgrove et al. (1)   | Present work  |
| 181.73 | $4^- ; 10.04 \pm 0.10$ | $3^+ ; 8.51 \pm 0.12$ | $4^- ; 10.40$ |
| 240.10 | $2^- ; 3.6 \pm 0.1$    | $2^- ; 3.3 \pm 0.1$   | $2^- ; 3.63$  |
| 292.41 | $3^+ ; 714 \pm 7$      | $2^+ ; 639 \pm 17$    | $3^+ ; 735$   |
| 681.43 | $3^+ ; 962 \pm 20$     | $3^+ ; 926 \pm 23$    | $3^+ ; 984$   |

Work is going on at Bologna to obtain more resonance parameters from such simultaneous fits; it is expected that significantly improved results will be obtained in consideration of the fact that ORELA data correspond to a much thicker sample than those used at Geel.

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## $^{109}\text{Ag}$ Capture Cross-section

At the 23rd NEANDC Meeting in September 1983 it was agreed that the  $^{109}\text{Ag}$  capture cross-section should be added to the NEANDC Discrepancy List because of the discrepancies between the results of Weston et al.<sup>(1)</sup>, Kononov and Stavisskii<sup>(2)</sup> and Mizumoto et al.<sup>(3)</sup>. However, shortly after the meeting, the data of Macklin<sup>(4)</sup> became available which agree well with the recent measurement of Mizumoto et al. It is now considered that there is no discrepancy and the  $^{109}\text{Ag}$  capture cross-section should be deleted from the Discrepancy List.

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## $^{243}\text{Am}$ Capture Resonance Integral

### Introduction

Measurements of the  $^{243}\text{Am}$  capture resonance integral give a value (2250 b) which is about 20% higher than the same value calculated from differential measurements (1830 b). This discrepancy cannot be attributed to the differences in the cadmium cut off energy or admixtures of  $^{242}\text{Am}$ . More measurements are needed to solve this discrepancy.

Short descriptions are given of existing integral and differential measurements.

### Integral Measurements

Measurements of the capture resonance integral of  $^{243}\text{Am}$  are listed in Table 1.

Eberle<sup>(1)</sup> irradiated Pu-Al alloy in a reactor and measured the production of different actinides in the sample. Nuclear data were taken from the literature and adjusted to reproduce the isotopic composition from  $^{238}\text{Pu}$  to  $^{246}\text{Cm}$  obtained experimentally. Also the ratio of thermal to epithermal flux was varied to obtain best agreement between experiment and calculation. The experiment is not suited to extract nuclear cross-sections but to check the adequacy of the data used for a particular purpose.

Schuman<sup>(2)</sup> irradiated pure cadmium-covered  $^{241}\text{Am}$  samples in a high flux core position of ETR to determine heavy element build-up and measure resonance integrals.  $^{197}\text{Au}$  and  $^{59}\text{Co}$  monitors were used. The isomeric ratio obtained for the two states of  $^{242}\text{Am}$  was 0.8 which is much lower than that obtained in other experiments. No cadmium cut-off is given.

Butler et al.<sup>(3)</sup> irradiated pure  $^{241}\text{Am}$  samples in the NRX reactor. The isotopic composition was experimentally determined before and after the irradiation. The cadmium cut-off energy was 0.5 eV.

Folger et al.<sup>(4)</sup> made a reactor activation measurement both with bare and cadmium-covered samples. A fairly high cadmium cut-off energy (0.83 eV) was claimed.

Bak et al.<sup>(5)</sup> made reactor irradiation build-up studies using both bare and cadmium-covered  $^{241,243}\text{Am}$  samples. Information about the cadmium cut-off energy is uncertain.

Gavrilov et al.<sup>(6)</sup> measured both  $I_\gamma$  and  $I_f$  for  $^{241,243}\text{Am}$  using almost pure isotopes. In calculating the thermal cross-section cadmium correction, a cut-off energy of 0.68 eV was applied. Some doubt can be expressed if this also was the cadmium cut-off energy in the experiment.

### Differential Measurements

The capture resonance integral of  $^{243}\text{Am}$  calculated from resonance parameters and adopted in recent evaluations for ENDF/B, JENDL, KEDAK and UKNDL gives a value of the resonance integral of about 1850 b (Table 2). The resonance parameters were obtained from differential measurements of the total cross-section of  $^{243}\text{Am}$ <sup>(7,8,9)</sup>.

Coté et al.<sup>(7)</sup> made a transmission measurement at the Argonne fast chopper using samples enriched to 99.5%  $^{243}\text{Am}$ . Eleven resonances were analysed for  $E_0$ ,  $\Gamma_n$  and  $\Gamma_\gamma$  from 0.9 to 15.3 eV.

Simpson et al.<sup>(8)</sup> collected transmission data from 0.5 to 1000 eV using ORELA and two high-purity samples of  $^{243}\text{Am}$ . Two hundred and thirty-eight resonances were analysed to get  $E_0$ ,  $\Gamma_n$  and  $\Gamma_\gamma$ . The resonance absorption integral for neutrons with energies above 0.625 eV was determined to be  $1810 \pm 70$  b.

Belanova et al.<sup>(9)</sup> measured the  $^{243}\text{Am}$  transmission for neutrons at the SM-2 reactor between 0.4 and 30 eV. The  $\text{AmO}_2$  sample contained 79.8% americium with an isotopic purity of 96.9%  $^{243}\text{Am}$ . Forty-eight levels were analysed. The value of the total resonance integral was calculated to be 1740 b.

### Summary

The measured resonance capture integral of  $^{243}\text{Am}$  is about 20% larger than that calculated from differential resonance data (Tables 1 and 2). The deviation cannot be explained by differences in the cadmium cut-off energy or sample impurities. More measurements are needed to solve this discrepancy.

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Table 1

Measured capture resonance integrals for  $^{243}\text{Am}$

| Author   | Year | Ref. | $I_{\gamma}$ (b) | $E_{\text{Cd}}$ (eV) |
|----------|------|------|------------------|----------------------|
| Eberle   | 1972 | 1    | 2250             | 0.5                  |
| Schuman  | 1969 | 2    | 2160±120         | -                    |
| Butler   | 1957 | 3    | 2290±50          | 0.5                  |
| Folger   | 1968 | 4    | 2250             | 0.83                 |
| Bak      | 1967 | 5    | 2300±200         | (0.3)                |
| Gavrilov | 1977 | 6    | 2300±200         | 0.68                 |

Table 2

Calculated capture resonance integrals for  $^{243}\text{Am}$  from differential measurements

| Author   | Year | Ref. | $I_{\gamma}$ (b) | $E_{\text{Cd}}$ (eV) |
|----------|------|------|------------------|----------------------|
| ENDF/B V | 1972 |      | 1818.4           | 0.5                  |
| JENDL 2  | 1982 |      | 1816             | 0.5                  |
| UKNDL 3  | 1982 |      | 1841             | 0.5                  |
| KEDAK 4  | 1982 |      | 1847             | 0.5                  |
| Benjamin | 1975 | 10   | 1810             | 0.632                |
| BNL 325  | 1973 |      | 1820±70          | 0.5                  |