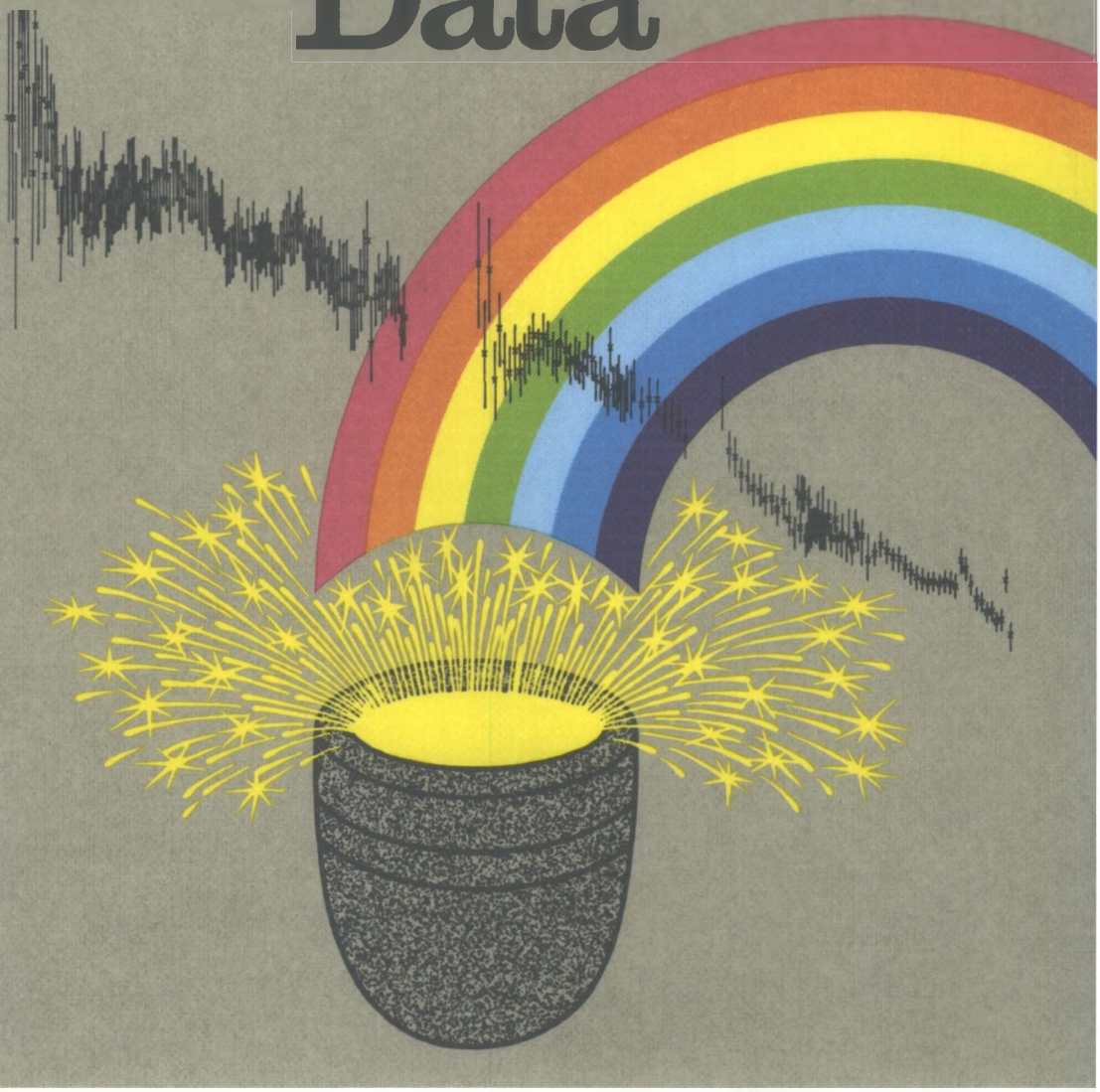
Neutron Standard Reference Data

Proceedings of a Panel, Vienna, 20-24 November 1972



INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1974

NEUTRON STANDARD REFERENCE DATA

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NEUTRON STANDARD REFERENCE DATA

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ON NEUTRON STANDARD REFERENCE DATA
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN VIENNA, 20-24 NOVEMBER 1972

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FOREWORD

Acting on the recommendation of the International Nuclear Data Committee (INDC) the Agency convened a Panel Meeting in Vienna, 20-24 November 1972, to review the progress in the field of neutron standard reference data. The meeting was a follow-up of a previous Agency panel entitled Nuclear Standards for Neutron Measurements, held 8-12 May 1967 in Brussels. The report of that meeting was issued as a technical document, IAEA-107.

Both meetings were convened as part of the Agency's general responsibilities to promote and coordinate the evaluation and exchange of scientific information in the field of nuclear data. Between 1967 and 1972 the initiative for reviewing improvements in neutron standard reference data was shared among many organizations. The 1970 symposium at the Argonne National Laboratory, which was sponsored by the European American Nuclear Data Committee, was devoted exclusively to neutron standards, and sessions covering various aspects of standards were included in several other meetings.

The present Proceedings contain all papers presented at the November 1972 meeting as well as the essential details of the technical discussions. In addition to the conclusions and recommendations the final section contains brief summaries of the status of currently available data for light-element standards and fission and capture standards. Some important conclusions of the meeting are the following.

The determination of the neutron flux no longer appears to be an outstanding source of uncertainty in standard cross-section measurements. Continued improvement in these data must be accomplished through small improvements in many aspects of the measurements.

For the light-element standard $^{6}Li(n,\alpha)T$, recent measurements of the (n,α) cross-section near the 250-keV resonance appear to agree within experimental error. These measurements tend to call into question either the total cross-section data or the assumptions of the theory used to interpret the total and (n,α) cross-sections.

Below about 500 keV it was concluded that the fission cross-section of ^{235}U was sufficiently well known for an evaluation of the available data to achieve 5% accuracy. However, it was emphasized that consequences of fluctuations in the cross-section must be carefully considered when ^{235}U is used as a standard below about 100 keV.

Measurements of the gold capture cross-section by counting prompt capture gamma rays appeared to be more consistent than measurements by the activation technique. It was concluded that resolution of discrepancies between the two methods would be a significant contribution toward making gold a more reliable standard.

Differences between values of $\bar{\nu}$ for spontaneous fission of ^{252}Cf determined by direct measurement and values derived indirectly from measurements of the parameter η are many times larger than the total errors

assigned to each method. The $\bar{\nu}$ experts attending the Panel, all of whom had made direct measurements of $\bar{\nu}$, challenged the indirect determinations or the uncertainties assigned to those measurements.

The Panel recommended that a standard fission neutron spectrum be included among the neutron standard reference data. The spectrum of neutrons from spontaneous fission of ^{252}Cf was proposed as the primary standard, and the neutron spectrum from fission of ^{235}U induced by neutrons of energy less than 150 keV was recommended as an associate standard.

When reporting experimental work on neutron standards, the participants insisted that it was essential to include detailed information about sources of uncertainty and possible systematic errors.

A third Agency-sponsored meeting devoted to nuclear standard reference data is tentatively planned for late 1976. The somewhat earlier date recommended by the participants and reported in these Proceedings has been postponed because of the possible partial overlap of subject matter with other meetings already scheduled during the intervening period.

The INDC has recommended that the scope of the next meeting be expanded to include all nuclear standard reference data instead of being restricted to the acknowledged neutron standards discussed at the present meeting. It was emphasized that the standard reference data to be discussed should have direct relevance to practical problems in nuclear science and its peaceful applications, which constitute the continuing responsibilities of the International Atomic Energy Agency.

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CURRENT ACTIVITIES IN THE NEUTRON STANDARD DATA FIELD IN THE UNITED STATES OF AMERICA

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At the time of the first IAEA Panel on Nuclear Standards for Neutron Measurements, held in 1967 in Brussels[1], very few endeavours in the standard field were being carried out in the United States of America. There were some measurements of the light nuclei reaction cross-sections and capture measurements on gold. However, a major effort was being made to determine $\bar{\nu}$ of ^{252}Cf.

Activities in the standard field have rapidly increased in recent years, reflecting the importance of this area for nuclear data in reactor evaluation and design. Starting with only one project on the absolute fission cross-section of ^{235}U in early 1968, we now have a wealth of experiments being carried out, presently under way or being planned for the near future. Some work on the light elements and other standard quantities is also being carried out; the work on $\bar{\nu}$ of ^{252}Cf was concluded.

LOW-ENERGY RANGE

The standard cross-section programme at Gulf Radiation Technology, San Diego, Calif., includes measurements of capture cross-sections and reactions in the light-nuclei region. Absolute measurements of the capture cross-section of gold (and ^{238}U) and ^{3}He(n, p) were concluded and reported at the 1970 Helsinki IAEA Conference, the 1970 Argonne Symposium and the 1971 Knoxville Conference. Newer measurements were carried out for ^{10}B(n, α) in the 4- to 150-keV energy range relative to the hydrogen scattering cross-section with an uncertainty of 2-3%. The ^{10}B(n, $\alpha'\gamma$) cross-section was measured in the 4- to 1000-keV energy range with 1-2% uncertainty. Measurements of the ^{6}Li(n, α) cross-section are planned for the future.

Measurements are in progress at the Lawrence Livermore Laboratory which will yield ratios of the capture cross-section of gold to ^{6}Li(n, α) and ^{235}U(n, f). These measurements extend from 100 eV to 1 MeV. Measurements of the fission cross-section of ^{235}U relative to the hydrogen scattering cross-section are now under way in the 50-keV to 16-MeV energy range. Results with an accuracy of better than 2% are expected in 1974.

At Oak Ridge National Laboratory, measurements of the ^{235}U cross-section are being carried out up to 80 keV relative to the ^{10}B(n, α) cross-section. Three different groups are working on this problem and are

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, unpriced document IAEA-107 (1968).

attempting to resolve the present international discrepancies. Work on $^{238}U(n, \gamma)$ is continuing and being extended to the 0.5-MeV region. Total cross-section measurements of ^{6}Li are presently under way in the 20- to 500-keV energy range, and the possibility of measurements of $(^{10}B(n, \alpha_0))/(^{10}B(n, \alpha_1))$ is being considered.

INTERMEDIATE AND HIGH-ENERGY RANGE

The standard cross-section programme at Argonne National Laboratory (ANL) includes measurements on the light nuclei, the fission cross-section of ^{235}U, capture measurements on gold and ^{238}U, and several standard-related quantities. Measurements of the total cross-section of ^{6}Li in the 100- to 1500-keV range were completed and published. Recent measurements of the $^{6}Li(n, \alpha)$ cross-section between 90 and 600 keV are presented in paper IAEA-PL-246-2/16 in these Proceedings. The absolute cross-section of ^{235}U was measured between 30 keV and 3.5 MeV by applying three absolute and two relative measuring techniques. Some of these data are presented in paper IAEA-PL-246-2/25 in these Proceedings. Measurements of the fission cross-section in the higher MeV energy range are planned, as well as $^{10}B(n, \alpha, \gamma)$ measurements in the keV energy range.

Recent measurements of the ^{235}U fission cross-section at Los Alamos Scientific Laboratory were carried out relative to the hydrogen scattering cross-section in the 1- to 6-MeV energy range. An uncertainty of about 3% is anticipated. Preliminary values will be presented subject to final mass assay of the fissile samples.

At the University of Michigan, a programme is under way in which absolute ^{235}U and ^{239}Pu cross-section values are measured using radioactive (γ, n) neutron sources, absolutely calibrated with a manganese bath. At present, a value of $1.216 \pm 2\%$ for ^{235}U using a NaBe source (966 keV) was obtained. The next measurement will be on ^{235}U using a NaD source.

TOTAL ENERGY RANGE

At the National Bureau of Standards (NBS), measurements are planned utilizing a white-neutron source (linac) and a monoenergetic neutron source (Van de Graaff). The measurements are planned to start within about one year and will cover the energy range from 1 keV to 16 MeV. An accuracy of 1% is anticipated. Presently, measurements of the total, scattered and (n, γ) cross-sections of ^{6}Li are being carried out. The objective is to establish these cross-sections in the 1-keV to 1-MeV range with better than 2% accuracy.

OTHER STANDARD DATA

Additional measurements of $\bar{\nu}$ of ^{252}Cf were proposed recently. Such measurements are also being considered at the University of Michigan.

Measurements of the fission cross-section averaged over the ^{252}Cf spectra were carried out at NBS. Age measurements are planned for the near future.

Measurements of the branching ratios of the associated activities ^{7}Be and ^{65}Zn were completed at ANL. The value for ^{7}Be is 0.1042 ± 0.0018.

UTILIZATION OF DATA

Two different committees are concerned with the nuclear standard data field. The US Nuclear Data Committee (USNDC) is reviewing and fostering experimental work on standard data. The Cross-Section Evaluation Working Group (CSEWG) is concerned with the evaluation of data to be included in the ENDF/B data file. Both committees have subcommittees on standards with overlapping membership.

CSEWG has recently reviewed the standard cross-sections of H(n, n), $^{3}He(n, p)$, $^{6}Li(n, \alpha)$, $^{10}B(n, \alpha)$, C(n, n), and $^{235}U(n, f)$. The data were included in ENDF/B-III and are thus utilized by reactor evaluators. This data set was made available by the USAEC to the IAEA.

DISCUSSION

C.D. BOWMAN: I have a comment on the age measurement of californium. Those measurements at the Bureau of Standards are complete. The analysis is not yet finished, but I would like to present some results from that work which I think have significance for the fission spectrum of californium.

I also have a comment on the measurement of the fission cross-section which is being made at the University of Michigan. Recently, this group attended the American Nuclear Society Meeting in Washington, D.C., and the result presented was 1.223 b at 960 keV, which is higher than the value quoted by Mr. Poenitz.

W.P. POENITZ: Mr. Knoll of the University of Michigan called me especially about this point. A correction was omitted from the value of 1.223 b. The correct value is 1.216 b ± 2% at 966 keV, which is what I quoted.

PRESENT AND PLANNED WORK ON NEUTRON STANDARDS IN AUSTRALIA

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This is a short summary of work on standards in Australia over the last few years. We have made several $\bar{\nu}$ measurements for thermal fission of ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu, all relative to ^{252}Cf. We have also measured $\bar{\nu}$ for spontaneous fission of ^{240}Pu and ^{242}Pu. We have measured the variation with neutron energy of $\bar{\nu}$ for ^{233}U, ^{235}U and ^{239}Pu relative to ^{252}Cf. As a complementary work, we have measured the variation of the average total kinetic energy of the fission fragments for ^{233}U and ^{235}U. Recently, we have measured the absolute number of neutrons emitted in the spontaneous fission of ^{252}Cf. This measurement is almost complete although there are a few little things that must still be examined. A full report on that work is given in paper IAEA-PL-246-2/33 *in these Proceedings*.

For the future, there are plans to consider fission neutron spectra (^{252}Cf and ^{235}U), primarily because these are of interest in our $\bar{\nu}$ measurements and could result in quite significant corrections; they might possibly cause difficulties. There are also plans to measure the capture cross-section of ^{238}U using a white source. We are not yet quite sure who will supply us with a white source but we plan to use one and to check some spot points with a Van de Graaff. Depending upon the results of this Panel, we may in fact examine some fission cross-sections of ^{235}U. We hope that we will have more people working in this area for the next years.

DISCUSSION

A.J. DERUYTTER: Do you mean that you have absolute measurements on ^{235}U at 2200 m/s or only ratios to californium?

J.W. BOLDEMAN: We have the original ratios to californium; we have not repeated the measurements recently. However, we have used the data which we obtained in the recent californium measurements to correct the early ^{233}U and ^{235}U data. In other words, these measurements can now be quoted independently of californium.

WORK ON NEUTRON STANDARDS AT THE KERNFORSCHUNGSZENTRUM KARLSRUHE

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At the Kernforschungszentrum, the fission cross-sections of ^{235}U in the energy region above 500 keV are being measured. One measurement between 500 keV and 1.2 MeV is reported in paper IAEA-PL-246-2/27 in these Proceedings. Other work is being done at higher energies, up to about 20 MeV, using the cyclotron. Furthermore, the total cross-section of ^{10}B was measured between 10 and 300 keV. For future activities, there are plans to measure the capture cross-section of ^{238}U up to about 500 keV and also the capture cross-section of gold up to that energy.

DISCUSSION

T.A. BYER: When do you begin the measurements on the capture cross-sections of ^{238}U and gold?

F. KÄPPELER: We hope to start these experiments at the end of this year or the beginning of next year.

PRESENT AND PLANNED NEUTRON STANDARD REFERENCE DATA AT THE CENTRAL BUREAU FOR NUCLEAR MEASUREMENTS

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One of the main objectives of the Central Bureau for Nuclear Measurements (CBNM) was and remains to provide experimenters within Euratom (and also outside on request) with very well defined targets that may be used for neutron standard reference data evaluations. In particular, the CBNM provides alloys and layers of fissile materials and absorption standards such as boron and lithium. More information on these services can be obtained upon request from our laboratory.

For neutron standard reference data evaluations, certain quantities are sometimes needed in cross-section calculations, such as alpha half-life values. The half-life for alpha decay of ^{234}U was recently published and is relevant to the standard 2200-m/s fission cross-section of ^{235}U which, in its turn, is used for the normalization of this cross-section in the resonance region and even higher up in energy. The CBNM is at present measuring the alpha half-life values for ^{233}U and ^{239}Pu which are also needed for a similar purpose. In both cases, the recent calorimetric measurements of Oetting are lower than the best values obtained from precise alpha-counting combined with destructive methods of target definition.

Concerning the low neutron energy standard activity, two papers were recently published in the Journal of Nuclear Energy[1] on the normalization of the fission cross-sections of ^{235}U and ^{239}Pu to the 2200-m/s values for these isotopes, and values of suitable resonance integrals are suggested for further normalization of these cross-sections. Two detailed papers were prepared on the CBNM measurements to deduce the 2200-m/s fission cross-sections of ^{235}U and ^{239}Pu. They were discussed in detail in the Concultants' Meeting on the Third Evaluation of the Thermal Fission Constants, held in Vienna from 15 to 17 November 1972.

Our low-energy programme includes measurements of the 2200-m/s σ_f-values of ^{233}U and ^{241}Pu. Especially ^{241}Pu is important as there is practically no precise direct measurement available.

Recently, transmission experiments were performed on solutions of Li_2SO_4 in heavy water for natural and highly enriched lithium in the neighbourhood of 2200 m/s. However, for the highly enriched solutions, the results obtained for the 2200-m/s absorption cross-section of ^{6}Li were quite different from the generally accepted value. For this reason, we are now planning transmission experiments on ^{6}LiF layers to try to resolve this discrepancy.

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[1] DERUYTTER, A.J., WAGEMANS, C., J. Nucl. Energy 25 (1971) 263.
DERUYTTER, A.J., et al., J. Nucl. Energy 26 (1972) 293.

Further, we plan to study the scattering of a collimated beam of fission fragments from different scatterers (A and Z values) and under different angles to try to improve the precision of 2π, 4π and low-geometry fission counting experiments.

Measurements on the fission resonance integrals of ^{233}U were made with a linac, and further measurements are planned on ^{241}Pu in the low-resonance region with the intention of normalizing them to the reference 2200-m/s values and of suggesting absolute resonance integrals for further normalization.

It is also planned to measure the $^{6}Li(n,\alpha)$ to $^{10}B(n,\alpha)$ ratio with a linac and to begin this direct comparison at 2200 m/s with well defined layers and geometry. This implies an evaluation of the number of boron and lithium atoms and the availability of sufficiently thin layers to resolve the reaction product spectrum.

Further, we began to measure the fission cross-section of ^{235}U relative to ^{10}B, including a low-energy part for normalization, and intend to go up to 100 keV.

Measurements of $\sigma_{tot}(^{6}Li)$ through the first resonance are being performed by Böckhoff et al.

Our programme of comparing the results of different flux determination methods (associated-particle method, proton recoil telescope, methane- or hydrogen-filled proportional counter) at different neutron energies is described in the proceedings of the 1970 Argonne symposium. In the meantime, we have added to this series a comparison at 250 keV (between the associated-particle method and a proportional counter with γ-discrimination). A corresponding paper has been accepted by Nucl. Instrum. Methods. At present, these flux methods are applied to the determination of $^{197}Au(n,\gamma)$ cross-sections in the energy range 0.1-1 MeV by the activation method, but no results exist so far.

Some effort has been put into compiling and evaluating the information on differential cross-sections for the source reactions T(p,n), D(d,n) and T(d,n). Preliminary results have been published as EANDC(E)-"L" documents. New or additional information has been received in the meantime, and at present a re-evaluation is performed. The final results will be published in Nuclear Data. Using the reciprocity theorem, the results of the $T(p,n)^{3}He$ and $D(d,n)^{3}He$ evaluation may also be interpreted as evaluations for differential cross-sections of the reactions $^{3}He(n,p)$ and $^{3}He(n,d)$. A report of this investigation is presented in paper IAEA-PL-246-2/21 in these Proceedings.

Finally, there are experiments under way to determine the fission neutron spectra for ^{235}U and ^{252}Cf.

We also helped in a comparison with a high-intensity neutron beam of a Szabo and White fission chamber with the aim of resolving some systematic differences between the two chambers for ^{235}U.

As concerns the relationship of the CBNM activities to the national nuclear energy requirements, it can be said that, on one side, the activities in the field of standards have always been guided by the European-American Nuclear Data Committee (EANDC) and especially its subcommittee on standards. On the other side, the recommendations of the Joint European Nuclear Data and Reactor Physics Committee (JENDRPC) and the Euratom Working Group on Reactor Dosimetry have always been carefully considered.

DISCUSSION

E. MIGNECO: Do you plan to push the measurements on the ratio of the (n,α) cross-sections of ^{6}Li and ^{10}B to 100 keV?

A.J. DERUYTTER: Measurements up to 100 keV are intended for the ^{235}U fission cross-section relative to $^{10}B(n,\alpha)$. The work on the ratio of $^{10}B(n,\alpha)$ to $^{6}Li(n,\alpha)$ is still being planned. We intend to begin with a comparison at thermal energies. Then using the same technique, we shall try to go to higher energies. The ratio will be determined directly by counting the alpha particles.

E. MIGNECO: Some years ago, precision measurements at high energies were difficult because of the gamma flash. Has this problem been solved?

A.J. DERUYTTER: That is the reason why our previous measurements did not extend to higher energies; we could not measure closely enough to zero-time. We now have a much improved arrangement for gamma-ray shielding.

NEUTRON STANDARD REFERENCE DATA ACTIVITIES IN JAPAN

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The present activities in Japan concerning neutron standard reference data deal with the evaluation of $^{6}Li(n,\alpha)$ and $^{235}U(n,f)$ cross-sections. The evaluation work is one of the activities of the Japanese Nuclear Data Committee. It is expected that some of the neutron standard reference data, such as the $^{197}Au(n,\gamma)$ cross-section, can be measured using the JAERI electron linear accelerator.

1. Evaluation of the $^{6}Li(n,\alpha)$ cross-sections in the range of 10-500 keV [1]

The cross-section of this reaction is one of the important nuclear standards. However, the number of cross-section measurements in the range of 10-100 keV is rather meagre, and the experimental values are scattered considerably. Furthermore, a resonance around 250 keV has not yet been fixed unambiguously. In the present evaluation work, the cross-sections in the region where the 1/v law applies (< 100 keV) and the 250-keV resonance are analysed with the following formula

$$\sigma_{n\alpha}(E) = \frac{(\text{const. } 1)\sqrt{E}}{(E_0 - E)^2 + \frac{1}{4}\Gamma^2} + \frac{(\text{const. } 2)}{\sqrt{E}} - \Delta\sigma \qquad (1)$$

Here, the 250-keV resonance is considered to be p-wave, and the energy dependence of Γ and Γ_α is neglected. The so-called Shapiro term $\Delta\sigma$ is taken into account. The five parameters, i.e. E_0, Γ, const. 1, const. 2, and $\Delta\sigma$, are derived from the observed cross-sections by using the least-squares method. The experimental data collected in the range of 10-500 keV are grouped into two parts, i.e. data obtained before 1964 and those obtained after 1964. The difference between the two groups is much larger than the experimental errors shown by the authors. For the present work, the data after 1964 were chosen, i.e. those published by Schwarz (1965), Fort (1970) and Sowerby (1970). (Condé (1965) and Barry (1966) are excluded because of scarcity of the data points.) These data are treated with equal weight.

The result of the evaluation is given in the following equation:

$$\sigma_{n\alpha}(E) = \frac{0.0144\sqrt{E}}{(0.245 - E)^2 + (0.107)^2/4} + \frac{0.1491}{\sqrt{E}} - 0.033 \qquad (2)$$

where the energy is expressed in MeV.

The χ^2-value in the range of 0.001-500 keV is 1.45 per point. The obtained value of Γ, i.e. 107 keV, is narrower than previously reported values, while the resonance energies agree well with each other. The thermal value, σ_{th} = 937.5 b, which is obtained by extrapolating Eq. (2), agrees well with directly measured values, i.e. 938 ± 6 b by Meadows et al. (1970), 936 ± 4 b by Meadows (1970), and 944 b ± a few per cent by Becker (1970), as well as a recommended value in Rep. BNL-325, i.e. 945 b. We are expecting the results of new measurements of the ^{6}Li(n, α) cross-sections by the Harwell group.

2. Evaluation of the ^{235}U(n, f) cross-sections in the range of 1 keV - 20 MeV [2]

The review and evaluation work by members of the Japan Nuclear Data Committee (JNDC) is progressing. Most of the experimental data on fission cross-sections of ^{235}U reported before 1965 are compiled in Rep. BNL-325, 2nd Edition and Supplement No. 2. The JNDC has collected the cross-section data after 1965 as far as possible. 32 data have been collected. In the same period, three important evaluations have been published, i.e. Davey's (1966, 1968), ENDF/B-III (1972), and Konshin and Nikolaev's (1972). These three evaluations disagree discernibly in the range of 10 to 50 keV and around 1 MeV. In the same energy region, the experimental values also show rather large discrepancies.

In the present work, data other than the fission cross-section, i.e. total cross-section, capture cross-section and scattering cross-section, will also be evaluated consistently with the fission cross-section. The work is not yet completed.

REFERENCES

[1] NAKAGAWA, T., IGARASI, S., oral report to 1972 meeting of Atomic Energy Society of Japan.
[2] MATSUNOBU, H., NISHIMURA, K., to be published as JAERI report.

NEUTRON STANDARD REFERENCE DATA ACTIVITIES IN SWEDEN

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At present, no direct measurements of neutron standard cross-sections are being done in Sweden. However, there are some activities which are related to problems connected with the determination and evaluation of neutron standard cross-sections.

METHODS AND TECHNIQUES OF NEUTRON FLUX MEASUREMENTS

The Monte-Carlo program MULTSCAT for calculations of neutron attenuation and multiple scattering effects has been further developed and tested by Holmqvist et al. [1,2]. The program has recently been used to correct data of fast neutron elastic and inelastic scattering measurements. In these experiments, the cross-sections for the H(n,n) or C(n,n) reactions have been used as reference data.

A proton recoil telescope [3] has also been developed to measure fast neutron fluxes based on the (n,p) scattering cross-section. The telescope is equipped with solid-state detectors.

FISSION AND CAPTURE STANDARDS

Strömberg [4] gives a review of the experimental fission cross-section data for ^{233}U, ^{235}U, ^{238}U, ^{239}Pu, ^{240}Pu and ^{241}Pu. The review points out the deviation between different evaluations and experimental data in the region 10 - 200 keV for the ^{235}U fission cross-section and the structure below about 100 keV.

It is intended to measure fission cross-section ratios at neutron energies above 5 MeV at the pulsed neutron facility, Tandem Accelerator Laboratory, Uppsala [5]. Experiments are planned on ^{236}U, ^{238}U and ^{232}Th, using the ^{235}U fission cross-section as a standard but also referring to the (n,p) cross-section by the use of a proton recoil telescope. The ^{235}U fission foils were prepared at the Central Bureau for Nuclear Measurements, Geel, and a back-to-back ionization chamber with time-of-flight techniques will be used.

Different corrections to the measurement of $\bar{\nu}$ for ^{252}Cf by Asplund-Nilsson et al. [6] have been investigated. The French effect was studied experimentally, and in accordance with results by Soleilhac et al. (see Ref. [7]) a correction of -0.6% in the absolute $\bar{\nu}$-value was found [8].

Axton [9] has calculated the leakage correction for a similar scintillator as was used by Asplund-Nilsson et al. and got a different result. The possibility to re-investigate the correction for the leakage

of neutrons from the liquid scintillator is being discussed. The correction was necessary because the leakage of neutrons from the large liquid scintillator was different in the case of isotropically emitted neutrons from a Cf-source put in the centre of the tank and in the case of neutrons scattered by an anthracene crystal. In the latter case, the neutrons enter the scintillator at certain angles corresponding to certain energies of the scattered neutrons. A Monte-Carlo calculation was made by Asplund-Nilsson et al. which resulted in a leakage correction of (1.3 ± 0.3)%.

Furthermore, experiments by Bergqvist et al. of capture cross-sections in the MeV-region are in progress, comparing results from activation measurements with those from gamma-ray spectrum measurements. A large contribution from thermal neutron capture has been observed near the target in the activation measurements, stressing the importance of a small thermal capture cross-section of the materials to be used as standards in this type of measurements.

Finally, the measurement by Johansson et al. [2] of the ^{235}U fission neutron spectrum has been continued. Accurate results exist at 0.53 MeV of incident neutron energy, which are in better agreement with a Watt distribution than with a Maxwellian distribution. The spectrum has been measured from 0.6 to 15 MeV of fission neutron energy, using time-of-flight techniques and a liquid scintillator detector with pulse-shape discrimination against gamma rays. The relative efficiency of the detector was determined from n-p scattering.

STANDARD REFERENCE DATA REQUIREMENTS

The standard data activities reported above are related to a number of relative neutron data measurements which are in progress, e.g. on neutron elastic and inelastic scattering, fast neutron capture and fission neutron spectra. These measurements are in general initiated as being requested for reactor or shielding calculations.

The last Request List for Neutron Data Measurements from Sweden was presented in October 1971 and is included in WRENDA. The fast neutron data set of the SPENG library has also recently been discussed by Häggblom [10, 11] in two reports about adjustments of neutron cross-section data by a least-squares fit of calculated quantities to experimental results. Integral data obtained for some fast zero-power reactors including the FR-0 reactor were used. The required accuracies of the neutron data were determined from the effect of cross-section errors on the integral data. Häggblom concludes in his report that the sensitivities to changes in the neutron data of the fuel isotopes are very large, leading to very small acceptable errors in these data. Some of the accuracies required can probably not be achieved by differential measurements. In any case, this emphasizes that very accurately determined standard cross-sections are needed.

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PRESENT AND PLANNED WORK ON NEUTRON STANDARDS IN THE UNITED KINGDOM*

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Part I

The present and planned standard neutron reference work in the United Kingdom on the measurement of neutron flux and light-element standard cross-sections is reported briefly. The experiments have been carried out at the Atomic Energy Research Establishment, Harwell, and at the National Physical Laboratory (NPL), Teddington. The work at NPL referred to is condensed from a paper by Axton et al. [1].

First, the flux measurements in the two laboratories are described. At Harwell, there are three experiments of interest.

(1) The Harwell long counter has been recalibrated absolutely on the pulsed Van-de-Graaff IBIS over the energy range 50 keV to 1300 keV, using the associated-activity technique. This work, done by Adams et al., has been published as a Harwell report [2].

(2) The Harwell black detector, which is used to measure the relative flux spectrum on the neutron booster of the 45-MeV linac, has been cross-calibrated against the Harwell long counter on IBIS in the energy range of 64 keV to 2 MeV in order to test its theoretically predicted efficiency.

(3) Rose at Harwell has measured the relative efficiency of a liquid scintillation counter to higher accuracy over the energy region of 100 keV to 13 MeV using neutrons of known primary energy which are scattered from hydrogen.

For the future it is hoped to build a type of proton recoil telescope for use in time-of-flight work above 1 MeV.

At NPL, facilities are provided to establish neutron flux density standards at a number of neutron energies from thermal to 19 MeV. At thermal energies, a standard neutron-flux facility has been successfully commissioned which uses fast neutrons produced by a Van de Graaff accelerator and moderated in graphite. Also an intense slowing-down flux proportional to 1/E has been established using a similar fast-neutron source moderated in water. For a great part of the higher-energy range above about 100 keV and up to about 2 MeV, the primary standards are neutron sources calibrated in a manganese sulphate bath. Work has been

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done to establish long counters as secondary standards by calibration against the standard sources and by direct comparison with the response of a vanadyl sulphate bath over the energy range 150 keV to 630 keV. At 14 MeV, the neutron flux has been measured to higher accuracy by means of proton recoil monitors, and progress has been made in establishing iron and aluminium foils as secondary standards.

For the future it is expected to use the associated-activity technique as well as hydrogen proportional counters to improve the accuracy of absolute measurements up to about 2.5 MeV. At higher energies it is hoped to use the associated-particle technique on the D(d,n) reaction for neutrons between 2.5 MeV and 5 MeV and on the T(d,n) reaction for neutrons between 14 MeV and 19 MeV.

The light-element standard cross-section measurements are all being made at Harwell. Two experiments to determine the $^{6}Li(n,\alpha)$ cross-section have been done. Firstly, Clements and Rickard have used ^{6}Li sandwich detectors to obtain a measurement in the energy range of 160 keV to 3.9 MeV using Van de Graaff accelerators, and secondly, Coates et al. have used ^{6}Li glass scintillators on the electron linac to obtain measurements between about 1 keV and 500 keV. This latter experiment is described in paper IAEA-PL-246-2/17 in these Proceedings. The Clements and Rickard data have been published as a Harwell report but this is not available for general reference.

A preliminary measurement of the $^{10}B(n,\alpha)$ cross-section has been obtained between 1 keV and 300 keV on the electron linac using a B_2O_3 disc in conjunction with NaI scintillation counters. This is described in paper IAEA-PL-246-2/20 in these Proceedings. For the future, more measurements on the $^{10}B(n,\alpha)$ cross-section are planned and also, if necessary, further measurements on $^{6}Li(n,\alpha)$.

All of the work mentioned has direct relevance to the United Kingdom nuclear energy programme. At Harwell, the work is directly geared to meeting the Category-1 and -2 requirements. Although the primary aim of the NPL work is not the same, this part of their work effectively moves towards the same objective.

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Part II

As regards the fast fission cross-section of ^{235}U, the sole activity in the United Kingdom consists of my measurements on the linac. These are in the energy range of 1 keV to 1 MeV and are relative measurements of the cross-section. The measurements are made by detecting prompt fission neutrons with a pulse-shape discrimination system. As flux standards, the black detector of Coates and Hart and the Harwell long counter are used. An accuracy of between 3% and 4%, depending on the energy, has been achieved for these measurements. Although we are dealing here with standards, I think that these data should not be considered completely in isolation. We have measurements of the ^{239}Pu fission cross-section and of its ratio to ^{235}U, but these are not completely analysed yet.

In the future, with highest priority, we hope to improve our knowledge of the lower-energy end of the incident neutron spectrum using a thin ^{6}Li glass detector. Because the neutron beam which is used for these measurements will also be used in the future for important capture cross-section measurements, we hope to do further work on the neutron flux spectrum at higher energies, possibly with the proton recoil detector used by Rose for the fission neutron spectrum work. With less urgency we hope to measure the ^{235}U fission cross-section in the 1- to 5-MeV region on the Harwell synchron-cyclotron using flux measurements with the proton recoil detector mentioned in Part I.

This type of measurement is a priority-1 request in the UK Nuclear Data Request List. An accuracy of ±3% is being asked from 100 eV to 5 MeV and is not presently available in evaluated cross-sections.

Ferguson has done work on the fission neutron spectrum. Currently his group is continuing measurements on the Harwell pulsed Van-de-Graaff IBIS on the ^{235}U fission neutron spectrum at an incident neutron energy of 550 keV. This energy was chosen to be exactly comparable with the measurements of Holmqvist. It was found that a double Watt spectrum is required to fit the data. These measurements were made with a solid cylindrical sample. Measurements above 550 keV with a ^{235}U fission chamber were also made. Finally, high resolution measurements on ^{252}Cf with a 3-m-long flight path are made to investigate data from Russian reports on structure at high energies.

Again the motivation for this work comes from the UK Nuclear Data Request List. Although the request is not priority 1 at the moment, it is considered of great urgency to make the experimental measurements. The request is for a mean energy of the neutron spectrum to better than ±2%.

DISCUSSION

E. J. AXTON: This comment is an extension of what Mr. Coates said about the NPL work. The work at NPL is mainly oriented towards its own problems and not specifically aimed at neutron data measurements. Nevertheless, where NPL techniques can be used to make a significant contribution to the nuclear data field, specific nuclear data measurements are undertaken.

The main items are the ^{238}U capture cross-section in the range 150 keV to 600 keV and the measurement of $\bar{\nu}$ of ^{252}Cf. The $\bar{\nu}$ measurements are described in paper IAEA-PL-246-2/31 in these Proceedings. The ^{238}U activation cross-section measurements are not covered because this is not a standard cross-section. Detailed papers describing the keV measurements with the long counter and vanadium bath and also the 14-MeV work are available. Since these have been submitted to journals, they are not presented here.

H. LISKIEN: You mentioned that it is planned at Harwell to construct a proton recoil telescope for time-of-flight measurements. What type of fast detectors do you plan to use for such a device and what resolution do you expect?

M.S. COATES: We have not yet considered the details of the construction but we would expect it to be a telescope of the type used by Macklin at Oak Ridge which has a hydrogenous foil radiator and semi-conductor detectors to detect the recoil protons.

H. LISKIEN: Also for the dE/dx counter?

M.S. COATES: I do not think we will use a dE/dx counter.

H. LISKIEN: So telescope in this sense does not mean that a coincidence condition is required?

M.S. COATES: The geometry of the detector would be used to define the energy received, so that an essentially monoenergetic pulse would be produced rather as in the Käppeler telescope. Time-of-flight methods would be used to determine the actual energy.

H. LISKIEN: What sources were used in calibrating the Harwell long counter by the associated-activity method?

M.S. COATES: Targets of vanadium and iron were activated by the reactions $^{51}V(p,n)^{51}Cr$ and $^{57}Fe(p,n)^{57}Co$. The activated targets were compared with standard sources of ^{51}Cr or ^{57}Co.

H. LISKIEN: These measurements are extremely difficult, partly because they are so sensitive to background neutrons. The sources are very weak, and the long counter has no directional selectivity.

M.S. COATES: To overcome this difficulty, a second experiment was performed in which a ^{6}Li glass scintillator plus time-of-flight analysis were used to check for false neutron groups.[1]

F. KÄPPELER: What is the energy range in which you plan to use the proposed telescope?

M.S. COATES: It would be used above 1 MeV because of the difficulty of getting a thin enough foil, and we hope to go up to about 5 MeV.

F. KÄPPELER: I am wondering how you plan to discriminate against the background caused by neutron reactions in the silicon of your detector.

M.S. COATES: The detectors would be out of the beam.

L. STEWART: Will this telescope be used in the linac measurements?

M.S. COATES: Yes. Perhaps it will also be used on the synchrocyclotron, certainly for time-of-flight work. This is really very much in the future; we have not the effort at the moment to actually do it. All we have done is to order the foil from Geel.

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[1] Editor's note: In Ref.[2] of Part I it is stated that the angular distributions of neutrons from each of the activating reactions were measured at a number of incident proton energies: "The background angular distributions, measured with a paraffin absorber between the target and detector were essentially isotopic in all cases."

PRESENT AND PLANNED NEUTRON STANDARD DATA ACTIVITIES IN FRANCE

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1. DEVELOPMENT OF NEUTRON FLUX MEASUREMENT TECHNIQUES AT CADARACHE

During the last few years, the reaction cross-section measurements were an essential part of our programme. Therefore it was necessary to have a reliable and precise device for measuring neutron fluxes. The results of this research effort have partly been published, but more work has been done recently with the aim of enlarging the available energy range and obtaining more stringent cross-checks. Especially, associated-particle counting with the $T(p,n)^3He$ reaction was used. These developments are reported in another paper in these Proceedings [1]. It is planned to continue this work during the next year.

Some investigations are also planned at Bruyères-le-Châtel, the aim of which is to cover the energy range of a few hundred keV to 15 MeV. This work will begin with associated particles at low energies.

2. LIGHT-ELEMENT STANDARDS

The $^6Li(n,\alpha)$ cross-section has been measured with the associated-particle technique in the energy range 80 keV to 500 keV, and in the energy range 20 keV to 1700 keV by comparison with the calibrated flat response counter developed at Cadarache. This is discussed in detail in two papers by Fort in these Proceedings [2]. From the experimental point of view, this work is nearly finished, although a few minor checks are still to be done.

The $^{10}B(n,\alpha)$ cross-section has also been measured from 20 keV to 150 keV by Szabo at Cadarache. The reaction products emitted from a boron layer were counted in a multi-wire proportional counter. This layer was made and calibrated at Geel. The geometry of the experiment and the procedure used were as close as possible to those of the fission cross-section experiment, and most of the systematic errors are essentially the same. The aim of this experiment was to compare this technique with the measurements based on the difference between total and scattering cross-sections. The values obtained are still preliminary because the calibration of the foil has to be improved by comparing it with a reference foil in a thermal flux. Presently, the results are in agreement with the evaluation by Gubernator and Moret [3] and slightly higher than the recommendation of Sowerby et al. [4] below 80 keV. The calibration of the foil will be completed and a few more points measured during the next year.

3. FISSION AND CAPTURE STANDARDS

The ^{235}U fission cross-section has been studied extensively at Cadarache by Szabo and his co-workers; for a discussion of these studies, see the paper by Szabo et al. in these Proceedings [5]. As this cross-section is of great importance for the measurement of other fission cross-sections, this programme will be continued in a larger energy range.

The absolute measurement of $\bar{\nu}$ for ^{252}Cf was contemplated at Bruyères-le-Châtel, but this experiment has been delayed because the large liquid scintillator is being used at the 60-MeV Saclay linac for one and a half years. The experiment is a measurement of the variations of $\bar{\nu}$ for ^{239}Pu and ^{235}U over the resonances. The results obtained are interesting and the experiment is still in progress. The decision to undertake an absolute measurement of $\bar{\nu}$ for ^{252}Cf will be reconsidered.

The fast neutron capture cross-section of ^{197}Au has been measured at Cadarache by two different techniques, between 70 keV and 550 keV. This work is described in detail in a paper by Fort et al. in these Proceedings [6]. It is a part of an important programme of capture cross-section measurements which will be continued for several years. The energy range will be extended from a few keV to 700 keV.

4. RELATIONSHIP OF THESE ACTIVITIES TO THE NATIONAL NUCLEAR ENERGY REQUIREMENTS

Accurate standards are useful for cross-section measurements for the development of nuclear energy. For instance, neutron standards can be used in the field of fast reactor physics. The material buckling of a fast reactor can be obtained from neutron flux distributions in a critical assembly like MASURCA. The measured distribution must be corrected for spectral perturbations near the boundary of the medium. The correction can be computed from the spatial distributions obtained using several detectors with different energy response, for instance ^{235}U and ^{238}U fission chambers. An improvement of the method would be to use an extra ^{6}Li detector whose maximum sensitivity corresponds roughly to the peak of the imperturbed neutron energy spectrum. The shape of the ^{6}Li cross-section has to be accurate around the 250-keV resonance.

^{6}Li and ^{10}B samples will be used in the core of the PHENIX fast reactor as integrated flux monitors. After irradiation, a comparison of the burn-up of these materials with the burn-up of the other materials irradiated at the same place will allow effective cross-sections of practical interest for reactor operation to be derived.

The thermal fission and capture cross-sections of fissile elements are used as standards for calibrating the detectors used to measure spectral indices.

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DISCUSSION

A.J.DERUYTTER: You mentioned the evaluations of the $^{10}B(n,\alpha)$ cross-section by Gubernator and Moret and by Sowerby et al. How do the preliminary values measured at Cadarache compare with these evaluations with regard to structure up to about 100 keV?

J.L. LEROY: Pending final calibration of the foil, our values are slightly higher, by about 2% I believe, than those recommended by Sowerby et al. There is no definite structure, but our points are not close enough together to eliminate the possibility.

E.J. AXTON: I would like to know the dimensions of the liquid scintillator tank used in the measurements of $\bar{\nu}$ for ^{252}Cf and ^{235}U and to inquire whether provisional values are available.

M. SOLEILHAC: The large liquid scintillator is spherical with a diameter of 70 cm. Since it is now being used at the Saclay linac, absolute values for the Bruyères-le-Châtel measurements are not yet available.

GENERAL DISCUSSION

B. D. KUZMINOV: At the Institute of Physics and Energetics in Obninsk, reference-data efforts are directed at the present time to research on the fission neutron spectrum from spontaneous fission of ^{252}Cf. We consider californium to be a very good reference for calibrating neutron detectors. We measure neutron fluxes using n-p scattering in a thin stilbene crystal, and we also use a proportional counter containing hydrogen. Briefly, this is our programme for the next few years.

C. D. BOWMAN: I would like to comment on the needs and justifications for some of the measurements currently going on in the United States. Two rather important developments in the past year have caused an emphasis on the region from about 3 MeV to 15 MeV. There are sizable engineering efforts under way in the controlled thermonuclear reactor programme which require higher-accuracy data in this energy range. The second matter is the strong interest in using 14-MeV neutrons for cancer therapy. Both of these relatively new needs, I think, will place a great deal more emphasis, at least in our country, on measurements of cross-sections with greater accuracy in the higher-energy range so that the question of standards in that energy range will become far more important than in the past.

J. J. SCHMIDT: I would like to ask for comments on the impact of WRENDA[1], the World Request List for Nuclear Data measurements for reactors, on national measurement programmes. May I invite comments of speakers who have not dealt with this before. The question I would like to have answered is, "Is WRENDA really used as a guide for national measurement programmes? Or is it just a list about which nobody really cares?"

E. J. AXTON: The two nuclear data items which I mentioned in my summary were both Priority-1 'starred' requests in WRENDA. In addition to these, there are many Priority-1 'unstarred' requests and many Priority-2 and -3 requests. It seems to me that there are so many requests in this document that anything which does not have at least Priority 1 will not attract very much effort.

J. J. SCHMIDT: Would you say that, for example, you yourself would consult this list to see if there is a Priority-1 item which fits into your general field of measurements? Would you then consider it?

E. J. AXTON: Yes.

L. STEWART: Having currently been working on the US Request List, I certainly agree with Mr. Axton. There are so many requests in WRENDA that the only things that have received any attention at all have been, in my opinion, Priority-1 items. For that reason, I think a great deal of effort needs to be extended to reduce the request list or at least to re-judge the priorities. The funding in the USA is such that the Priority-1 items are about the only things that are touched. I think some people have the tendency to use our request list merely to look at.

J. L. LEROY: In France, the WRENDA list is related to the programme of measurements. The number of requests is so large that we must use some criteria to discriminate among them. Our own requests, which of

[1] RENDA was changed to WRENDA when the IAEA assumed responsibility for the publication of the world-wide request list, i.e. at the end of 1972.

course are included in the list, are one criterion. In other cases, for example the neutron capture programme which is expected to cover a large number of isotopes, the consideration of all requests for a particular type of measurement has resulted in the completion of some measurements which might otherwise never have been made at all.

A. J. DERUYTTER: The WRENDA list is not restricted to standards and is used at our laboratory as a guide for other types of measurements. For planning of standard measurements we have relied until now on the International Nuclear Data Committee (INDC) Standards Subcommittee and on the Joint European Nuclear Data and Reactor Physics Committee.

E. MIGNECO: I remember that when I worked in Geel, we considered the WRENDA list very carefully in order to find experiments which we could perform effectively. We noticed that feedback to us, concerning our experimental results, from those people who originated the requests, was extremely slow. This gave us a very poor impression of the effectiveness of the request-list procedure.

J. L. LEROY: I think that what people really need are evaluations; users should not have to ask for measurements. The reason for long feedback times is that new measurements usually are not considered by reactor physicists directly and therefore remain unused until a new evaluation is made. Maybe it would be more effective if requests were made at a meeting of evaluators rather than by the users of the data.

B. D. KUZMINOV: We envisage a means of taking into account the established priorities with an objective theory of the planning of experiments. Usachev has done relevant work in this field. I agree with Mr. Leroy to the extent that requirements established theoretically should be supplemented by an evaluation of the data acquired. In this evaluation, discrepancies and errors should be considered. The evaluation should not give a single figure but should include a representation of the statistical and systematic errors.

E. MIGNECO: If one considers requests for resonance parameters, I think the confusion is complete because of the importance of the formalism used to analyse the resonance. The results depend not so much on the experimental data as on the way in which the analysis is applied. When I see a list of requests for resonance parameters with specified precision, I wonder whether it makes sense to ask for such high experimental precision when we know that we cannot analyse the results with equivalent precision.

J. J. SCHMIDT: Mr. Migneco's comment relates to the credibility of the request, which should be assured by discussions between the requestor and experimentalists who could actually perform the measurements before the request is put on the list.

H. LISKIEN: I want to return to the point which Mr. Leroy has raised. I remember that two or three years ago, there were two problems: first, to make a world-wide list out of the EANDC's RENDA, and second, to agree on the contents of the list, i. e. whether it should contain requests for experiments or for evaluations. At that time it was decided not to attack both problems at the same time but first to attempt to enlarge the geographical scope of the list. I now want to ask what is foreseen, by INDC or others, to solve the second problem, namely the flow of information from experimentalists to evaluators to reactor physicists? I direct this question to Mr. Schmidt in particular.

J. J. SCHMIDT: One part of our programme for co-ordination of the flow of information among measurers, evaluators and users is the new

WRENDA system, which is currently under development by the Agency's Nuclear Data Section with the co-operation of the Nuclear Data Centre in Saclay. This project was begun after recommendation by the European-American Nuclear Data Committee (EANDC) that the scope of its RENDA list be expanded, and following a subsequent recommendation by the Agency's own advisory group, the International Nuclear Data Committee (INDC). I shall not describe the programme in detail, but I would like to mention some features which are directly related to Mr. Liskien's question.

One weakness of the old RENDA list was that it was not updated and edited every year, and we consider yearly updating essential if the list is to be useful. This means that the users, requestors, evaluators and measurers of data must be contacted at much shorter intervals.

Another problem is the credibility of the requests which are put into the list. What are their justifications within the scope of the various national programmes? We feel that the credibility of the requests must be the responsibility of the national nuclear data committees and ultimately of the requestors themselves.

Of course, a request list must be reviewed periodically to determine which requests are filled, or partially filled, and which should remain open. The review procedure could operate either on a national basis or could employ topical reviewers to review all requests of a certain type. This matter continues to be under discussion.

One criticism of the RENDA list is that it contains too many requests; I think the present number is almost 400. Many of the requests refer to essentially the same quantity but differ slightly in detail. We plan to subdivide the new list into blocks which would contain all the requests for the same quantity. For example, there would be one block for the ^{239}Pu fission cross-section which would contain the approximately 15 requests for that quantity which appear in RENDA. This subdivision should make the list more readable and should eliminate duplication of much redundant information.

As you know, there are four neutron data centres in the world who are supposed to compile all experimental neutron physics data and to exchange them as rapidly as possible. Whether these centres do this effectively is not my point, but this is the aim of what we call 'four-centre co-operation'.

Some of these data are then evaluated by more or less national evaluation groups. The evaluated data are used in computer programs for reactor physics, and the reactor physicists compare their calculations with, for example, the results of critical experiments and other integral measurements. In co-operation with the evaluators, they then reformulate their requests for different or more accurate experimental data.

We are devising a scheme where all these requests will be put together in the WRENDA system and published annually. The neutron data centres may participate in the collection and exchange of data-request information as well as in the exchange of experimental data.

At the IAEA, we will use the WRENDA list as a means of promoting and implementing the nuclear cross-section measurement programme which we operate primarily in the developing countries. Presumably, other countries might make similar use of the list to obtain the data which are needed in the context of their own national programmes.

I hope this has explained briefly the kind of information exchange which we envisage and which we are trying to promote and facilitate.

Outside of this close group co-operation there is one further way, which was mentioned in informal discussion, to accomplish maximum dissemination of new experimental information. Anyone who has completed, or who has under way, measurements which directly relate to a WRENDA request should inform the requestor directly.

II. METHODS AND TECHNIQUES OF NEUTRON FLUX MEASUREMENTS

PROGRESS IN THE ESTABLISHMENT OF NEUTRON STANDARDS AT THE NATIONAL PHYSICAL LABORATORY

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Abstract

PROGRESS IN THE ESTABLISHMENT OF NEUTRON STANDARDS AT THE NATIONAL PHYSICAL LABORATORY.

Facilities are provided at the National Physical Laboratory to establish neutron flux density standards at a number of neutron energies from thermal to 19 MeV. A 3-MeV Van de Graaff and a 150-keV Sames positive ion accelerator are used in conjunction with analysing magnets to direct beams of accelerated charged particles along a number of flight tubes to irradiate neutron-producing targets in a number of neutron production and measurement facilities. Standards already available in the thermal, intermediate, and fast neutron energy ranges are described, together with plans for their extension and for calibration in the 1-eV to 1-keV energy range. A kerma standard for neutron therapy is also envisaged.

The full text of this paper has been published in Neutron Monitoring for Radiation Protection Purposes (Proc. Symp. Vienna, 1972) 2, IAEA, Vienna (1973) 431.

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INTERNATIONAL INTERCOMPARISON OF FAST NEUTRON FLUX DENSITY SPONSORED BY BUREAU INTERNATIONAL DES POIDS ET MESURES

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Abstract

INTERNATIONAL INTERCOMPARISON OF FAST NEUTRON FLUX DENSITY SPONSORED BY BUREAU INTERNATIONAL DES POIDS ET MESURES.

An international intercomparison of fast neutron flux density has been undertaken at nine laboratories under coordination by the Bureau International des Poids et Mesures. Neutron energies and transfer instruments related for the intercomparison are discussed briefly.

INTRODUCTION

During the past few years, the Consultative Committee for Measurement Standards of Ionizing Radiations (Section III, Neutron Measurements) of the Bureau International des Poids et Mesures (BIPM) has had under consideration the organization of an international intercomparison of fast neutron flux density. The main difficulty was to reach an agreement on a suitable transfer instrument with which to effect an intercomparison of the absolute flux density measurements in the various countries. Other difficulties were the choice of monoenergetic neutron energies, and whether or not to include pulsed white spectra from linear accelerators. At the April 1972 meeting of the committee, some firm decisions were made with a view to completing an intercomparison during 1973. Since such an intercomparison is considered relevant to the subject matter of this panel it seems pertinent to report the present status of this exercise.

CHOICE OF NEUTRON ENERGIES

To keep the intercomparison to a reasonable size and yet represent energies of interest to nuclear energy, neutron protection, and neutron therapy, three neutron energies were selected. These are shown in Table I, together with the dates at which the participating laboratories expect to complete the measurements.

The committee wishes to adopt a non-exclusive policy concerning the participation of other laboratories and so the chairman was authorized to put additional participants on the list with the proviso that the number of participants should remain small.

TABLE I. SELECTED ENERGIES AND COMPLETION DATES OF INTERCOMPARISON MEASUREMENTS AT NINE LABORATORIES[a]

Energy	CBNM	BIPM	CEN	ETL	IMM	NBS	NPL	NRC	PTB
250 keV	1972	-	1972	1972	-	1973	1972	1972	1974?
2.5 MeV	1973?	1972	1972	-	1972	1974	1972	1972	1974?
14.5 MeV	1972	-	1973	?	1972	1974	1972	-	1974?

[a] CBNM — Central Bureau for Nuclear Measurements, Euratom, Geel, Belgium.
BIPM — Bureau International des Poids et Mesures, Paris, France.
CEN — Centre d'études nucléaires de Saclay, Gif-sur-Yvette, France.
ETL — Electrotechnical Laboratory, Tokyo, Japan.
IMM — Institute for Metrology, D.I. Mendeleev, Leningrad, USSR.
NBS — National Bureau of Standards, Washington, D.C., USA.
NPL — National Physical Laboratory, Teddington, UK.
NRC — National Research Council, Ottawa, Canada.
PTB — Physikalisch-Technische Bundesanstalt, Braunschweig, FRG.

TRANSFER INSTRUMENTS

After considerable discussion it was decided to enter a phase of study of the properties of transfer instruments in order to select an optimum instrument or instruments and then later proceed with the intercomparison. It seemed desirable to adopt one moderation method and one reaction counting method at each energy, and Table II shows the instruments which were proposed for each energy. The long counter was eliminated for a number of reasons. There would be uncertainties in its energy response, no two laboratories possessing counters of exactly the same design. The problem of determining the effective centre is a major operation, and also it was felt that the flat response was not necessarily an advantage. To circulate one long counter to each participating laboratory in turn would extend the time-scale of the intercomparison beyond acceptable limits.

The gold-foil-sphere combination, and the indium and the nickel foils were eliminated because of the low count rates expected from the available flux levels. The ^{235}U fission chambers were rejected for the two higher energies and ^{238}U fission chambers selected instead, the reason being that although the ^{238}U cross-section is lower, there are no problems of thermal neutron background. The sphere-counter combination was eliminated at 14.8 MeV because the sphere size would be very large and would introduce severe scattering problems. Therefore, the following transfer instruments have been selected:

250 keV	Sphere and counter	^{235}U fission chamber
2.5 MeV	Sphere and counter	^{238}U fission chamber
14.8 MeV	Iron foil	^{238}U fission chamber

TABLE II. POSSIBLE TRANSFER INSTRUMENTS

Energy	Moderation methods		Reaction counting	
250 keV	CH_2 sphere (Au foil, BF_3 counter, LiI scintillator, Li glass scintillator)	Long counter	$^{115}In(n,\gamma)^{116m}In$ foil (54-min β-γ activity, irradiated in Cd cover)	^{3}He gas (scintillator or proportional counter) $^{235}U(n,f)$ fission ionization chamber
2.5 MeV	CH_2 sphere (Au foil, BF_3 counter, LiI scintillator, Li glass scintillator)	Long counter	$^{58}Ni(n,p)^{58}Co$ foil (70-d EC activation detector)	$^{238}U(n,f)$ fission ionization chamber $^{235}U(n,f)$ fission ionization chamber
14.8 MeV	CH_2 sphere (Au foil, BF_3 counter, LiI scintillator, Li glass scintillator)	Long counter	$^{56}Fe(n,p)^{56}Mn$ foil (2.6-h β-γ activation detector) $^{27}Al(n,\alpha)^{24}Na$ foil (15-h β-γ activation detector)	$^{238}U(n,f)$ fission ionization chamber $^{235}U(n,f)$ fission ionization chamber

Various thermal neutron counters are being investigated by four laboratories in an effort to find a sufficiently stable detector for the sphere-counter combination:

BIPM	BF_3 and ^3He proportional counters
CEN	^6Li-glass scintillator with photomultiplier
CBNM	^3He gas scintillator
NPL	Solid-state semiconductor with ^6Li layer

The use of iron foils at 14.8 MeV is subject to prior agreement on the beta counting of ^{60}Co foils which will be issued by the National Physical Laboratory.

The reaction counter methods will be investigated by three laboratories as follows:

CBNM	^3He gas scintillator
NRC	^3He proportional counter
NBS	^{235}U fission chamber
	^{238}U fission chamber

It is hoped that the feasibility studies will be completed by late 1972 so that the intercomparison measurements can get under way in 1973.

EXPERIMENTAL TEST OF THE CALCULATED EFFICIENCY OF THE HARWELL BLACK DETECTOR AT HIGH NEUTRON ENERGIES USING THE HARWELL LONG COUNTER

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Abstract

EXPERIMENTAL TEST OF THE CALCULATED EFFICIENCY OF THE HARWELL BLACK DETECTOR AT HIGH NEUTRON ENERGIES USING THE HARWELL LONG COUNTER.

The Harwell black detector has an essentially constant time response in the neutron energy range 10 eV to 700 keV with a fast enough time response to be used in time-of-flight experiments. Recently the theoretical efficiency calculated by Monte-Carlo techniques has been confirmed experimentally. It is considered that the relative efficiency has been established to ± 2%.

The Harwell black detector has been described in detail elsewhere [1,2] and only a brief summary is given here. The detector is designed to have an essentially constant efficiency over a wide neutron energy range (10 eV - 700 keV) and a fast enough time response to be used in time-of-flight experiments on the neutron booster of the Harwell 45-MeV linac.

The device consists of a homogeneous mixture of vaseline (effectively $CH_{2.05}$) and ^{10}B contained in a thin spherical aluminium shell. A radial re-entrant hole allows a parallel neutron beam to fall on the inner end near the sphere centre. Neutrons are moderated and captured in ^{10}B to produce 478-keV gamma rays, which are detected at the surface of the sphere in NaI scintillation counters. By suitably varying the parameters of the detector and calculating the properties with a Monte-Carlo code, a design was chosen in which the efficiency had the required energy characteristics. The neutron lifetime in the system (time to capture 99% of incident neutrons) is ~ 0.7 μs. The calculated properties of the detector were shown to be insensitive to changes in the relevant nuclear data over the region of flat energy response. The parameters of the experimental detector are listed in Table I.

The calculated efficiency is known to an estimated accuracy of ±1 - 2%. This precision can be reached with other detectors only over relatively limited energy ranges, which gives rise to normalization problems in the regions of overlap. Therefore, the wide energy range covered by the present detector will prove particularly useful. It must be demonstrated experimentally, however, that the calculated efficiency is correct. At low energies, measurements with detectors based on standard cross-sections provide a test, and some data are reported elsewhere in these Proceedings. At higher

* On attachment from Imperial College, University of London.

TABLE I. PARAMETERS OF THE EXPERIMENTAL DETECTOR

Sphere radius	12 cm nominal
Re-entrant hole depth	8 cm nominal
Re-entrant hole diameter	2.5 cm nominal
Containment shell	Aluminium, 0.175 cm thick
Boron content [a]	1059 g powder (nominally 90% pure boron with 90% ^{10}B content. Impurities: C (2%), Fe (0.7%), Ni (0.3%), Si (2.4%), O_2 (1%), (Cr+Al+Co+Cu) (1.9%)
Vaseline content	5389 g
NaI crystals	Four crystals symmetrically placed and essentially in contact with the sphere surface. Each crystal is 10.8 cm in diameter and 5 cm thick.

Comments: The re-entrant hole is formed by placing a thin-wall (0.0125 cm) stainless-steel tube across the sphere on a diameter and inserting into this an aluminium cylinder (0.025-cm-thick walls) containing ^{10}B-vaseline. The NaI crystals have 2.5-cm-thick lead collars to ensure that they detect only gamma rays from the required area on the sphere surface.

[a] The isotopic content and the presence of impurities were included in the calculations.

energies, the Harwell long counter has been used as the calibration detector in measurements on the pulsed Van de Graaff IBIS over the energy region from 68 keV to 2 MeV. Recently, the efficiency of the long counter has been re-measured [3] in this energy range to an accuracy of ±1-2%, using the technique of associated activity.

For technical reasons the black detector could not be installed easily on IBIS, so a secondary detector was used for the comparison. This secondary detector is a ^{10}B-vaseline cylinder, 7 cm dia. and 10 cm long, surrounded by four NaI scintillation counters to detect the 478-keV gamma rays following neutron capture in ^{10}B. It had been calibrated previously against the black detector on the 300-m flight path of the neutron booster [2]. In the IBIS measurements the Li(p,n) and the T(p,n) reactions were used to cover the required neutron energy range, with the secondary detector and the long counter set at 20° to the incident proton beam to allow a simultaneous accumulation of counts at a given neutron energy. Nanosecond time-of-flight analysis was used with the secondary detector in order to obtain accurate background determinations. The data and representative error bars are shown in Fig.1, where the continuous curve gives the secondary detector efficiency deduced from the black detector calibration, and the points give the efficiency obtained with the long counter. The two data sets are normalized to give the best eye fit of the continuous curve through the points between 68 keV and 700 keV. The experimental data confirm within the errors of measurement that the theoretical efficiency is correct. It is considered that the relative efficiency has been established to $\sim \pm 2\%$. It is to be noted that above 700 keV the IBIS measurements can be regarded as a calibration of the black detector efficiency in a region in which it cannot be calculated with confidence.

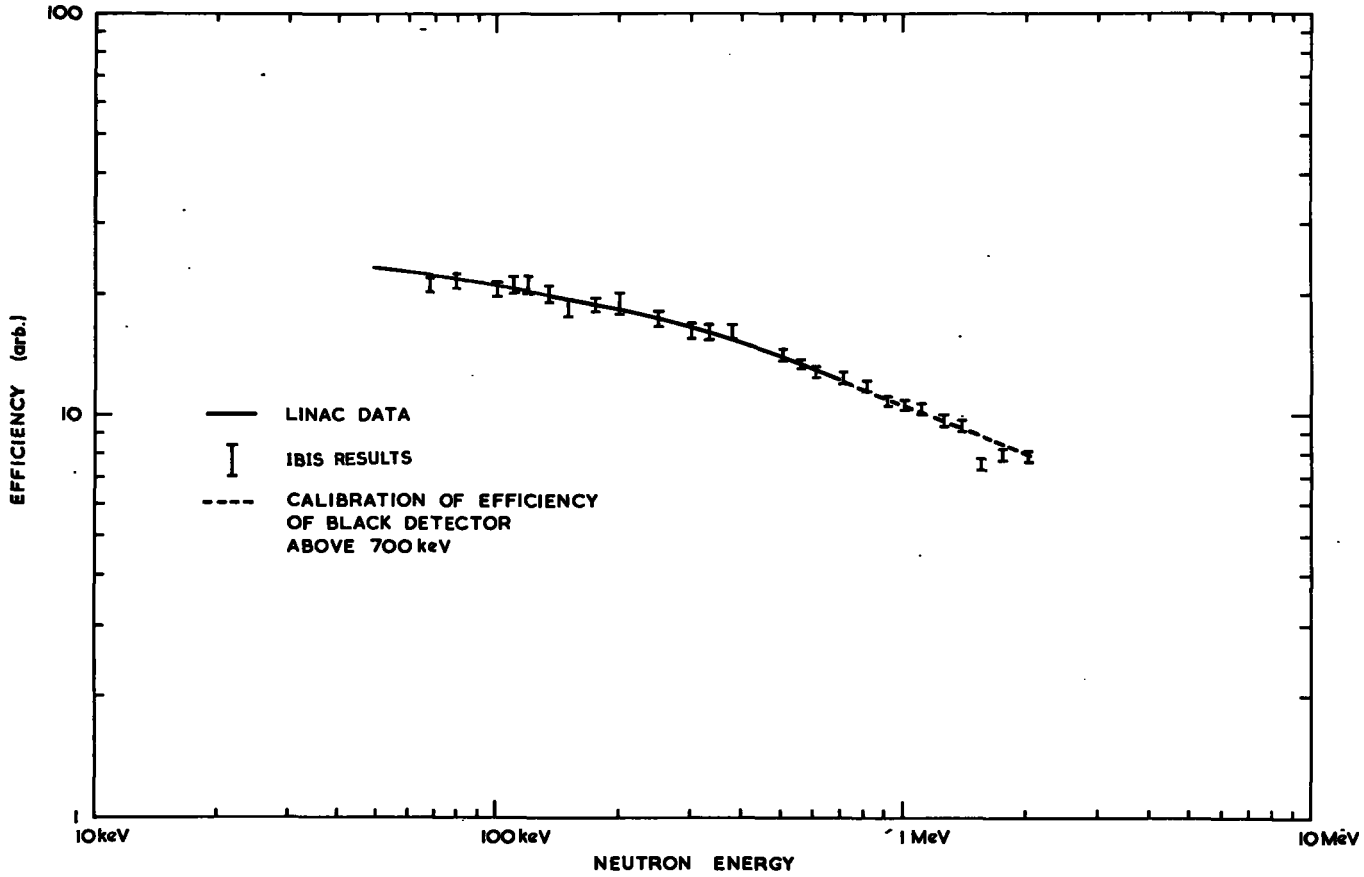

FIG. 1. Calibration of the secondary detector. Comparison of the efficiency of the new boron-vaseline plug using the Harwell black detector on linac and the long counter on IBIS (new results).

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[2] COATES, M.S., HUNT, G.J., UTTLEY, C.A., RAE, E.R., "Experimental tests with the time-of-flight method on a neutron detector with flat energy response", Neutron Standards and Flux Normalization (Proc. Symp. Argonne, 1970) CONF-701002, AEC Symp. Ser. 23 (1971) 401.

[3] ADAMS, J.M., FERGUSON, A.T.G., McKENZIE, C.D., Activation Technique for the Absolute Calibration of a Long Counter, Rep. AERE-R 6429 (1970).

DISCUSSION

W.P. POENITZ: How large were the deviations between the measured efficiency and the calculated efficiency in the higher energy range?

M.S. COATES: We have a calculation of the efficiency above 700 keV which extends to something like 1.5 MeV. As far as I remember, the deviation from the measured efficiency was not more than about 2%, but it was a systematic deviation. I would have to confirm that value. I really would not want to believe the calculated efficiency above 700 keV, partly because the efficiency begins to change more rapidly there and also because one can get gamma rays from inelastic scattering in the sodium iodide crystals at that energy. And so one would tend, perhaps, not to believe the data anyhow.

W.P. POENITZ: But you are really measuring the 411-keV gamma ray from the boron. You should have considerable discrimination against background from sodium iodide.

M.S. COATES: No, this is not true because the gamma rays coming from the sodium iodide are approximately of that energy.

J.L. LEROY: Is it correct that you have no experimental check below 300 keV?

M.S. COATES: No, this extends down to 68 keV.

J.L. LEROY: Do you plan to have more experimental checks below this energy?

M.S. COATES: Below this energy region the standard cross-sections are assumed to be known to better accuracy than we know the calculated efficiency. In the region below 10 keV the results which I shall present on thin lithium glasses tend to confirm that the efficiency is correct there, because the measured values are proportional to 1/V. To bridge the gap between 10 keV and the lowest energy obtainable with a Van de Graaff, one must take the calculated shape as the best available. It is possible to make measurements to lower energies on a Van de Graaff, but I tend to be sceptical of making measurements at backward angles when this sort of accuracy is required. Perhaps one could go down to 20 keV, but that is not much further down.

J.L. LEROY: Is it a question of background?

M.S. COATES: Yes.

J.L. LEROY: Some special arrangements would be required. It is possible, but it is difficult.

M.S. COATES: For the benefit one would get from extending the experimental check from 70 keV down to 20 keV, I think it would require unreasonable effort.

W.P. POENITZ: In addition, in this low energy range the calculated efficiency should be increasingly reliable. There should be essentially no question about the calculations if they are checked at higher energy.

R.W. PEELLE: Did I understand that the calibration of the black detector is via a second detector of similar type, then a long counter and then some source to calibrate the long counter?

M.S. COATES: Yes, that is more or less correct. The long counter was calibrated absolutely. The step in going from the black detector to the secondary detector was done on the linac with very good statistics, better than 1%. Systematic effects enter from the measurement of background and are the major sources of error in transferring the calibration from one detector to the other. This error must then be compounded with the statistical error of the points on the long counter calibration curve considering that there are several points. Finally this error must be compounded with the systematic error quoted for the long counter.

TWO FLAT-RESPONSE DETECTORS FOR ABSOLUTE AND RELATIVE NEUTRON FLUX MEASUREMENTS*

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Abstract

TWO FLAT-RESPONSE DETECTORS FOR ABSOLUTE AND RELATIVE NEUTRON FLUX MEASUREMENTS.

The application of the Grey Neutron Detector at higher neutron energies is considered. The detection of different capture γ-rays provides for a satisfactory check of the evaluated efficiency functions. A new detector, the Black Neutron Detector, was designed for absolute neutron flux measurements in the MeV energy range. The detector has a fast time response and converts the primary neutron energy into proton recoil energy. The energy sum spectra obtained from these proton recoils allows a simple extrapolation to zero energy and thus an accurate absolute count-rate determination. The detector is a promising instrument for accurate cross-section experiments in the MeV energy range.

1. INTRODUCTION

Since the earliest days of neutron physics there is a need for neutron detectors with a smooth and flat efficiency over a large energy range. One of the first detectors designed for this purpose was the long counter, which was applied in a large number of experiments in the past. However, soon it became apparent that this detector had several shortcomings, mainly associated with its heterogeneity. Thus, different concepts were introduced in a variety of detector designs, including among others the Marion counter, the Macklin sphere, the boron pile, and the large liquid scintillator [1]. An exceptional position has the manganese bath [1], which not only has a superior flat efficiency curve over a large energy range but also can be used to measure absolute neutron fluxes.

Here, two detectors are considered, which were designed as relative and absolute neutron flux detectors in neutron cross-section experiments.

2. THE GREY NEUTRON DETECTOR

The Grey Neutron Detector concept combines the flat efficiency curve of a manganese bath with the semi-prompt detection technique of a large liquid scintillator. Indeed, the first design provided a manganese bath surrounded with a thin shell of liquid scintillator for the detection of the capture γ-rays of manganese. In the final stage, the liquid scintillator was substituted by one or more NaI detectors. This provides for a better

* Work performed under the auspices of the US Atomic Energy Commission.

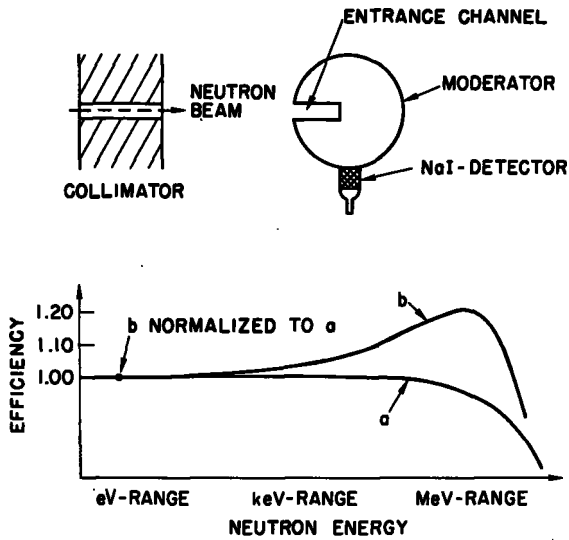

FIG. 1. Schematic of a Grey Neutron Detector and comparison of its efficiency curve (b) with that of a manganese bath (a).

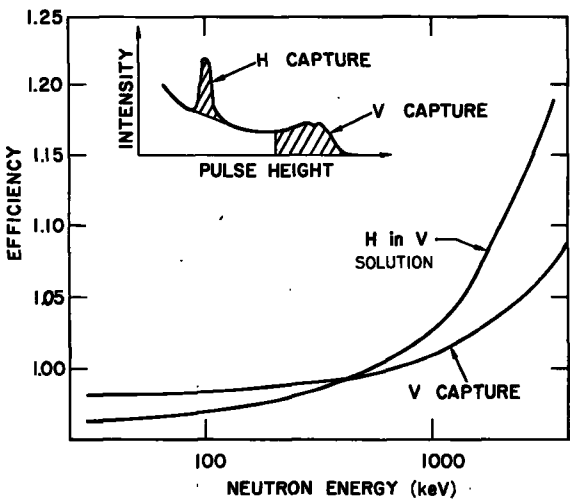

FIG. 2. Schematic energy spectra obtained with a vanadium sulphate solution, and efficiency curves corresponding to different spectra portions.

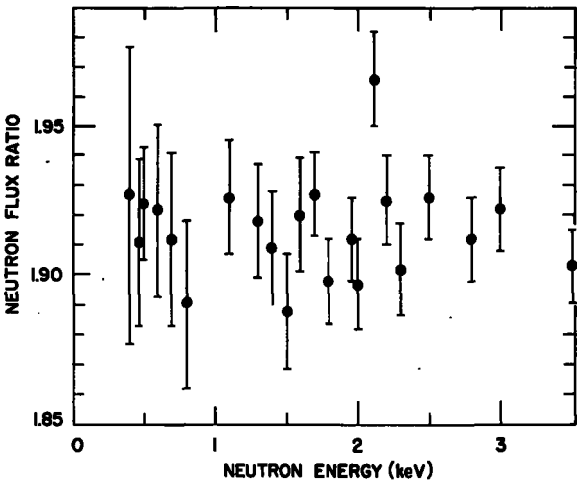

FIG. 3. Ratio of the relative neutron flux as a function of energy determined with the γ-spectra and efficiencies shown in Fig. 2.

background identification. This detector was discussed in detail elsewhere [2-4], and only some additional considerations, mainly concerning the application at a higher neutron energy, are given here.

A Grey Neutron Detector consists of a moderator with an entrance channel for a collimated neutron beam. The neutrons are slowed down in the moderator to thermal energy and subsequently captured in the moderator material. The capture γ-rays are detected at the surface of the moderator. Whereas the capture process corresponds to the capture of the neutrons in a manganese bath and thus has a flat efficiency curve (see Fig.1, curve a), the detection of γ-rays leaking through the surface of the moderator is dependent on the γ-ray absorption within the moderator. This γ-ray attenuation causes a second-order correction to the original energy dependence of the efficiency and a rise of the efficiency with increasing energy (see Fig.1, curve b).

The application of the detector at higher energies requires a larger correction and thus an increased uncertainty. When using the detector up to 3.5 MeV, several effects were considered:

(a) The correct description of the leakage by the flux function used for the efficiency evaluation was checked by comparing the evaluated leakage for a californium source with experimental values reported recently by DeVolpi [5]. The evaluated leakage agreed within ±0.5% with the experimental values.

(b) Different moderators were used, including $MnSO_4$, $VOSO_4$, water and paraffin. Because the 2.25-MeV capture γ-ray of hydrogen is part of the γ-ray spectra obtained with the salt solutions, a direct comparison of the relative neutron flux measured in either way could be made. Because of the different γ-ray attenuation coefficients the efficiency curves are quite different as shown in Fig.2. The same is true for the vanadium bath solution as shown in Fig.3. The ratio of the relative

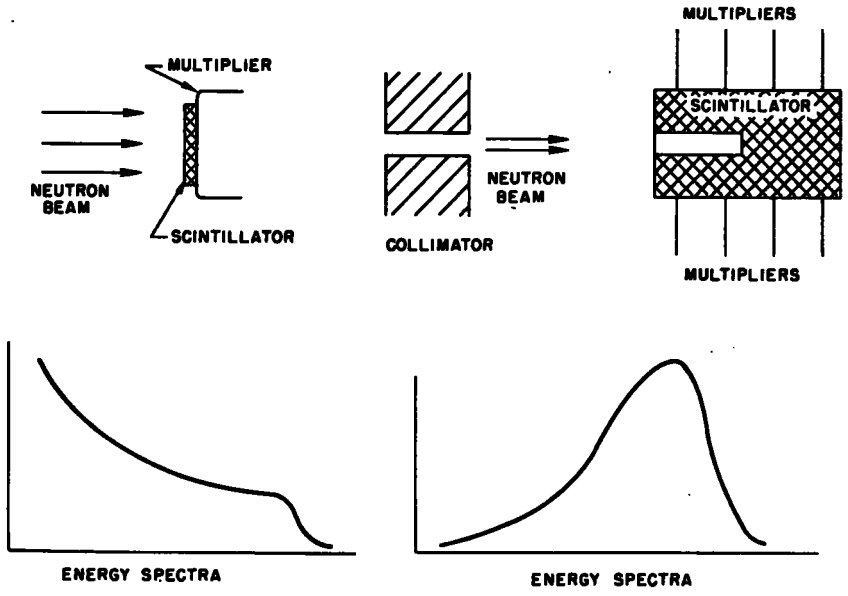

FIG. 4. Schematic comparison of a conventional scintillation detector (left) and the Black Neutron Detector (right).

neutron flux determined with the high-energy capture γ-rays of vanadium to that determined with the hydrogen capture γ-ray is shown in this figure.

A version of the Grey Neutron Detector with a timing in the range of 0.5 ns was recently reported by Coates et al. [6]. As moderator, a mixture of ^{10}B and vaseline was used and the 411-keV γ-rays from the $^{10}B(n,\alpha\gamma)$ conversion reaction were detected. A detector containing a mixture of various salts of elements with large (n,γ) cross-sections in the keV range was suggested [7]. This would allow the construction of a much cheaper and thus larger detector with a flat and larger efficiency in a more extended energy range.

3. THE BLACK NEUTRON DETECTOR

A disadvantage of the Grey Neutron Detector for use in fast-neutron cross-section experiments at higher energies is its slow time response. Several detector types are available which easily achieve the desired fast time response. For example, plastic or liquid scintillators viewed by a photomultiplier have usually a time resolution of few nanoseconds. However, for absolute counting, such a system has several disadvantages. Secondary effects cannot be sufficiently well evaluated because of uncertainties in the carbon total cross-section and especially the angular dependence of the scattering cross-sections. Owing to secondary processes and the noise of the photomultiplier, the spectrum obtained with such a conventional arrangement has a large percentage of low-energy pulses, making it difficult to determine the total count rate.

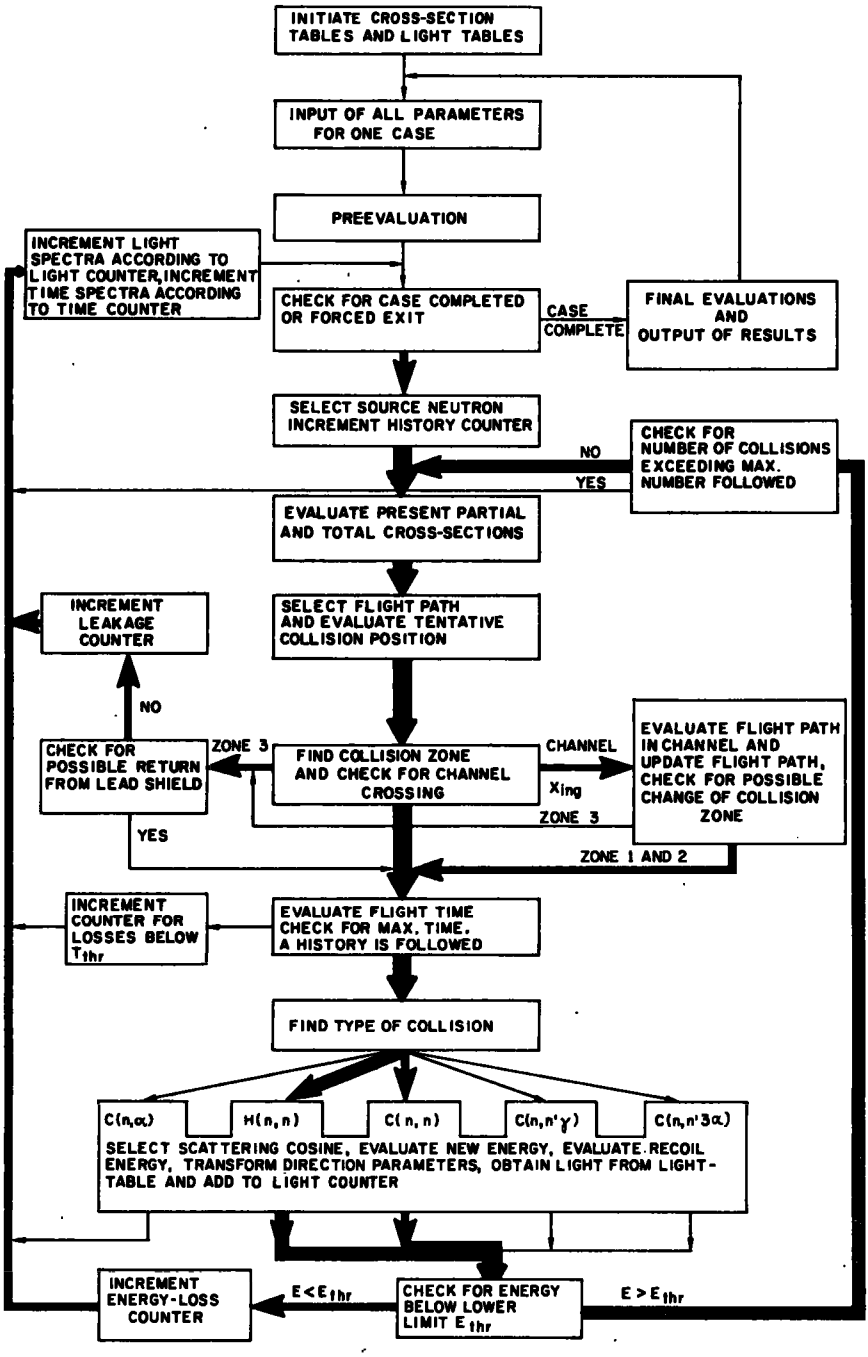

FIG. 5. Flow diagram of CARLO BLACK.

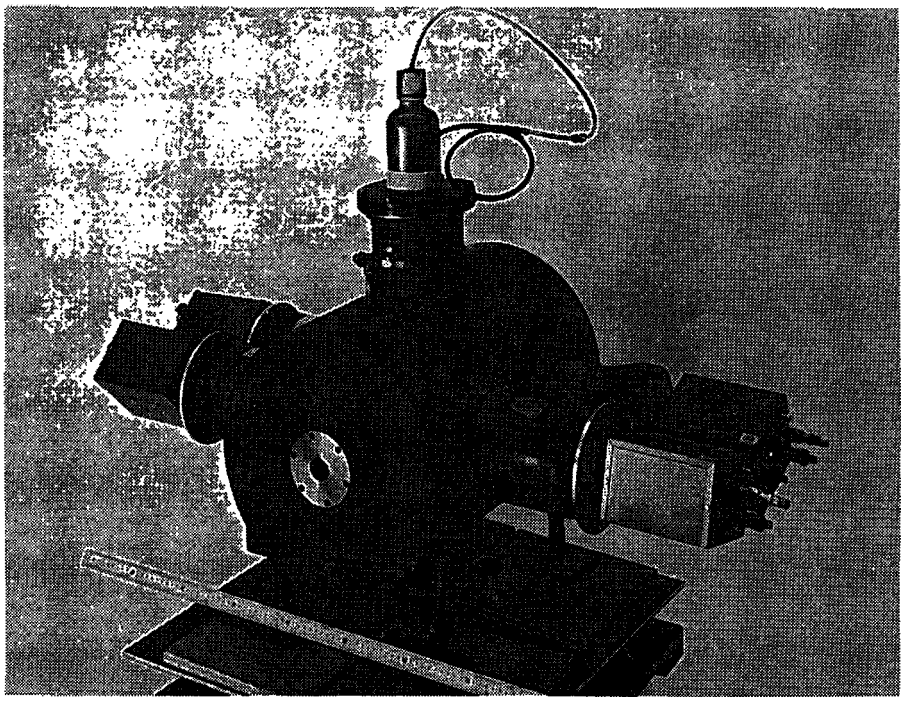

FIG. 6. The Black Neutron Detector.

3.1. Design and principle of the Black Neutron Detector

A detector, called the Black Neutron Detector, is designed to maintain the fast time response of a conventional scintillation detector while overcoming its weaknesses for absolute neutron-flux measurements. The detector consists of a cylinder of hydrogenous scintillator material. The neutron beam enters the detector system through a channel which terminates in approximately the centre of the detector. The system is sufficiently large so that a neutron undergoes several successive collisions before it falls below a low-energy threshold or leaks out of the system. As a result, most of the primary energy of the majority of the neutrons converts to hydrogen and carbon nuclei recoil energy and then to light. The scintillation light is detected with several photomultipliers. The qualitative difference in concept and response of the conventional scintillation detector and the Black Neutron Detector is illustrated in Fig.4.

The advantage of an additive device is appreciable. The extrapolation to zero pulse height can be carried out with high accuracy because there are very few small pulses if the detector system is sufficiently large. The probability for a first collision can be in the range of 99-95% for incident neutrons with energies of 1-10 MeV. Because of the small opening angle of the neutron beam entrance channel with little backscattering escape and the mechanism of multiple collisions, the influence of any involved cross-section is expected to be small.

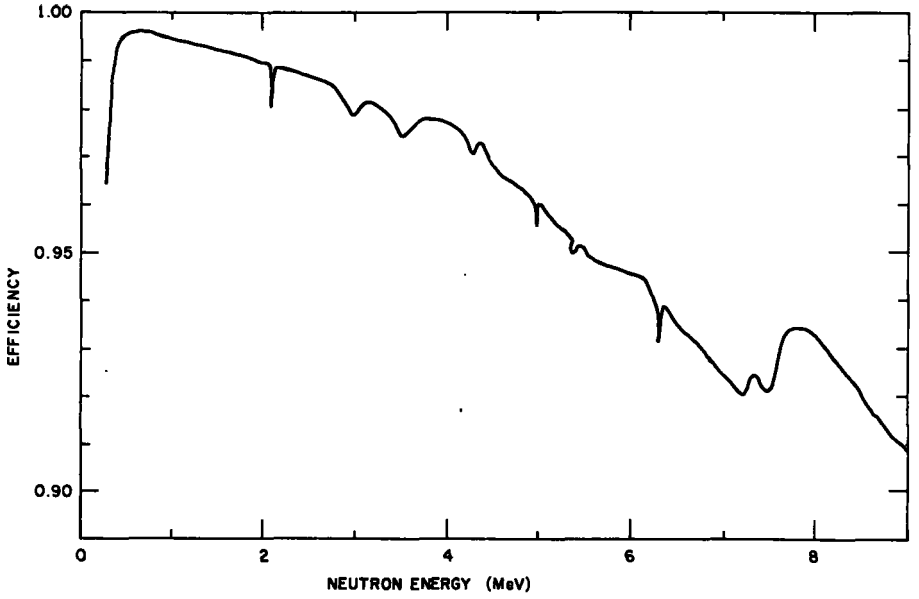

FIG. 7. Efficiency of the Black Neutron Detector versus energy.
H = 40 cm, R = 13 cm, H_C = 15 cm, R_C = 1.26 cm, E_C = 200 keV, $N_C = 4.2 \times 10^{22}/cm^3$, $N_H = 7.04 \times 10^{22}/cm^3$.

3.2. Monte-Carlo evaluation

The use of the detector in absolute-flux measurements of good accuracy requires a precise knowledge of the cross-section effects and of the optimization of the various detector parameters. For these purposes a Monte-Carlo code, CARLO BLACK, has been developed for the evaluation of a Black Neutron Detector of cylindrical shape with an axial neutron-beam entrance channel surrounded by a lead shield. The detector cylinder has a height H and radius R and is filled with a scintillator having the hydrogen and carbon atomic densities N_H and N_C. The neutron-beam entrance channel has a radius R_c and a height H_c. The incoming neutron beam is centred and has a circular, homogeneous distribution with a radius R_n. A neutron is described by its position parameters X, Y, Z, its directional parameters U, V, W, and its energy E. The history of a neutron is followed until it either leaks out of the system, falls below an energy threshold E_{thr}, or exceeds a time threshold T_{thr} before it is lost due to either of the two other reasons. The light produced by the recoil particles or by other charged-particle reaction products during the history of a neutron is summed and recorded at the end of the neutron history. High probabilities for first and second collisions within the detector require a large scintillation system. The size and thus the useful energy range for the application of the Black Neutron Detector are limited by the absorption of light within the scintillator and by the background, which increases with the volume of the detector. In the present considerations, a neutron energy of 10 MeV was chosen as a useful upper energy limit.

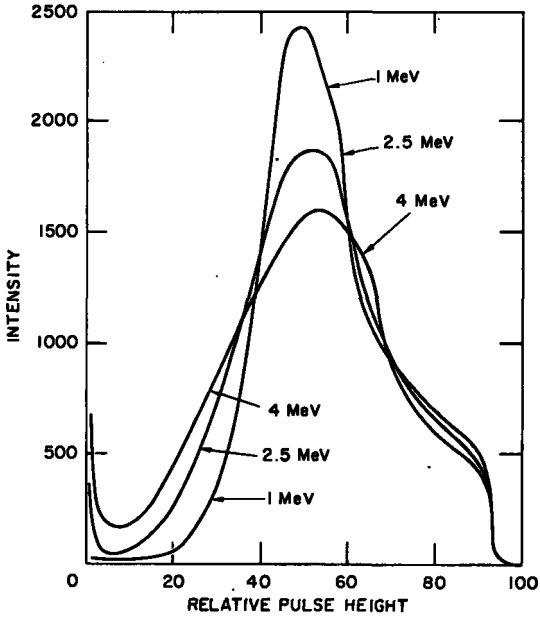

FIG. 8. Pulse-height spectra evaluated for three different monoenergetic neutron beams.

Figure 5 shows the schematic flow diagram for the problem. The frequency of the path a neutron may take in this flow diagram is indicated by the thickness of the lines. The main loop is that in which a neutron has to be followed through successive collisions. The average number of collisions of a neutron before its history is terminated by one of the above limitations is five to eight, depending on the incident energy. Usually two-thirds or more of these collisions involve hydrogen. About 90% of all collisions take place in Zone 1 of the detector which is the cylindrical part without the channel. Most neutron histories are ended by the neutron falling below E_{thr}. 10 000 to 20 000 histories usually give sufficiently low statistical errors. This is a computational reflection of the high physical efficiency of the detector (> 90%). Some parts of the present problem are extensively described along with their solutions in the general literature dealing with Monte-Carlo problems [8-10]. A summary description of the present evaluations is given elsewhere [11].

4. EFFICIENCIES AND SPECTRA

The efficiency of the Black Neutron Detector will depend mainly on the choice of the parameters R, H, R_c, H_c, N_H, and N_C. The detector parameters should be selected in such a way that the efficiency has the least dependence on the accurate knowledge of these parameters. Thus an investigation of the dependence of the efficiency on these parameters is of primary importance. As a result of these investigations [11], a detector with a size fulfilling these requirements was built. This detector is shown in Fig.6.

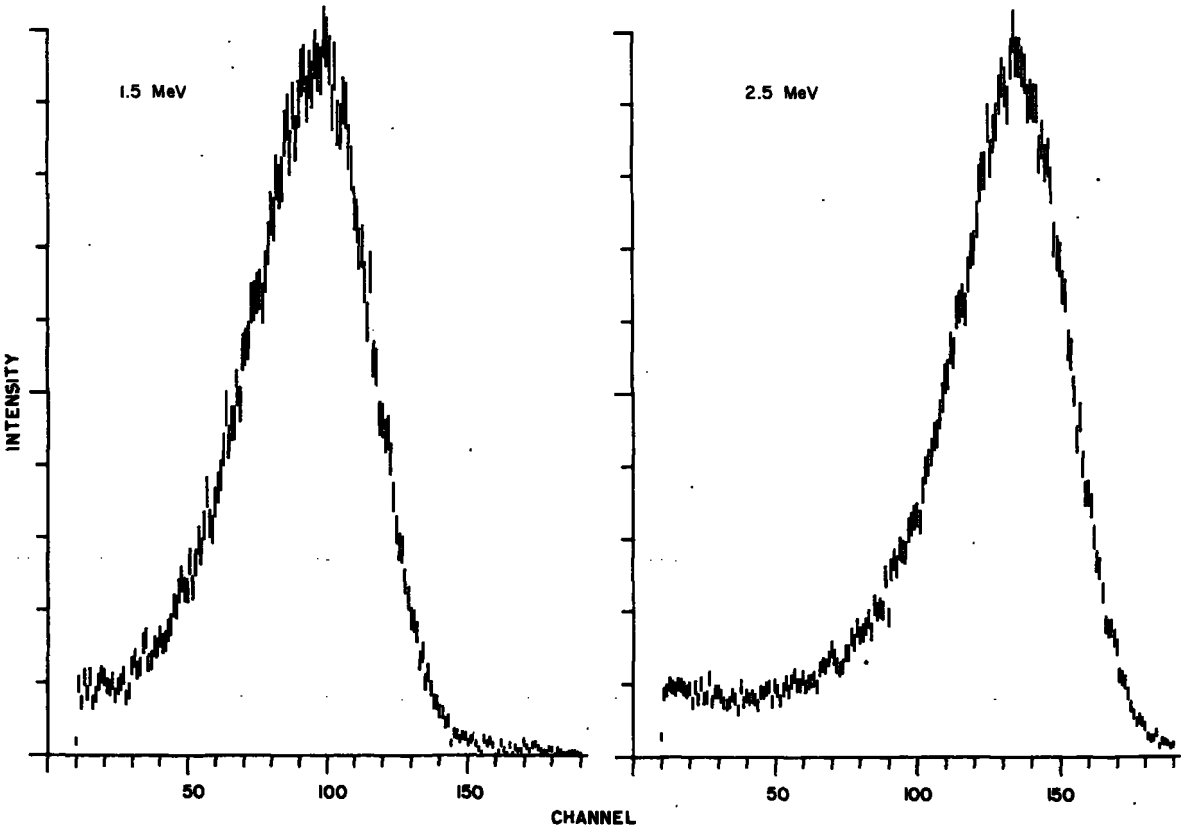

FIG. 9. Experimental pulse-height spectra for 1.5- and 2.5-MeV neutrons.

4.1. Efficiency versus energy

Figure 7 shows the efficiency of the Black Neutron Detector as a function of energy. A first glance at the structure of the efficiency curve appears disturbing if a smooth efficiency is required for the detector. However, the structure is very small and thus of minor importance. The uncertainty of the efficiency curve is small because of the insensitivity even to large changes in the cross-sections and parameters of the detector. The change in the carbon cross-sections around 2.0-2.1 MeV by a factor of 3-4 is reflected in the efficiency only by a change of about 1%.

4.2. Time distribution

The time resolution is mainly determined by the flight-time distribution for the first collision within the system. Although the full width at half maximum is 4 ns, an appreciable tail extends to about 10-12 ns. Still, this time resolution is reasonable for most fast-neutron cross-section experiments with monoenergetic neutron beams.

4.3. Energy spectra

Pulse-height spectra evaluated for three different primary neutron energies are shown in Fig.8. The spectra are normalized to the same maximum pulse height. The spectra still show the first-collision hydrogen pulse-height spectra underlying the more complicated sum spectra which result from the multiple-scattering events. The centre of a spectrum lies closer to the maximum pulse height for higher energies than for lower energies. The spectra shown in Fig.8 were calculated under very simplified assumptions about light attenuation and light reflection. Thus, only the gross structure of the spectra, that is, the peak at larger pulse heights and the minimum for low pulse heights, can be expected to be seen in experimentally obtained pulse-height spectra.

5. CONCLUSIONS

The Black Neutron Detector promises to be a very useful instrument for absolute neutron flux measurements in the MeV energy range. First experimental checks of the design concept showed a time resolution for the detector of 4 ns for the prompt γ-flash. The energy spectra shown in Fig.9 do not show a specific structure but confirm the expected low number of counts of small pulse height.

The Black Neutron Detector appears to be a promising instrument for the MeV energy region. Thus, with the Grey Neutron Detector and the Black Neutron Detector the whole energy range of interest for nuclear data for reactors is accessible.

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DISCUSSION

B.D. KUZMINOV: In calculating the efficiency, did you consider scattering from the lead shielding of the detector?

W.P. POENITZ: We did take into account scattering from the lead shield by making some rough calculations. Changes in these correction factors with neutron energy would cause changes in the efficiency. In this energy range most of the neutrons fall below the threshold for light production before they escape from the scintillator. Therefore changes in efficiency due to changes in reflection from the shield are not expected to be significant. At higher energies, this would be a problem if the size of the scintillator were not increased.

B.D. KUZMINOV: Did you take into account the effect of the passage of the neutrons through the collimator?

W.P. POENITZ: Not at the point where we evaluated the efficiency of the detector itself, because there we assumed that we had an incoming collimated neutron beam. But in the experiment in which the cross-section was measured we have of course corrected for the effect of the collimator.

C.D. BOWMAN: In the measured pulse-height spectra in Fig.9, the intensity drops very much more rapidly on the high side than on the low side. There appears to be a very remarkable difference between these spectra and the calculated spectra of Fig.8.

W.P. POENITZ: I noticed this myself and made a small effort to obtain a more linear spectrum. I was able to observe spectra in which the centre was more on the other side, but still I could not directly make out a bump on the higher-energy side.

I did not investigate further because the important thing is the extrapolation to zero pulse height. We investigated the linearity on the low-pulse-height side using a white source and the computer to isolate different energy ranges. The linearity is satisfactory for extrapolation.

C.D. BOWMAN: But you must rely on your calculated efficiency to take into account the effect of structure in the carbon cross-section, etc.,

on the efficiency. It is important to be able to calculate at least a close approximation of what you measure.

W.P. POENITZ: Yes, but remember that the carbon light is not included. The rise of the pulse-height spectrum at low pulse heights is due to carbon light, and we measure outside this range. The carbon light has only a secondary effect on the rest of the spectrum.

C.D. BOWMAN: But it changes the efficiency.

W.P. POENITZ: Not really. It is only the direct scattering effect from the carbon which changes the efficiency. For example, if you have a large resonance in the carbon cross-section, then it becomes increasingly probable that a neutron will be scattered by carbon rather than by hydrogen. Then maybe the scattered neutron could leak from the scintillator immediately. This is the effect, not the light production from the carbon. The result is a drop in efficiency at the energies of the carbon resonances.

J.L. LEROY: Do you think that a direct experimental confirmation of your efficiency curve would be useful? For instance, one could look for this drop in efficiency at the energy of a carbon resonance.

W.P. POENITZ: It is an excellent idea to look for the effects of the carbon resonances, and there is an experiment to do that. One measures the yield of the ^{7}Li(p,n) reaction using the detector whose efficiency is being studied and looks at the ratio of the second neutron group to the first neutron group. If the ratio is a smooth function of energy, and there is no reason to assume otherwise, there should be a noticeable effect on the ratio when the energy of the primary neutron group passes through a carbon resonance.

When we attempted this experiment, the results were insufficiently accurate statistically, and we did not have time to perfect the experiment. I was using a smaller detector at the time. The carbon resonances are very narrow so that there is a resolution problem in addition. One must use a very thin lithium target, but this causes statistical problems.

J.L. LEROY: Without any experimental check, the shape of your efficiency curve is purely theoretical. Although there are many reasons to believe that the calculations are good, can you guarantee an accuracy of better than 2% without a direct experimental cross-check when so many factors must be considered?

W.P. POENITZ: The basic design of the detector prevents large errors because in principle its efficiency should be 100%. If you vary some of the contributing factors in the calculation, e.g. the total or angular differential scattering cross-sections of carbon or the size of the detector, the changes must become very large before the uncertainty reaches 1% or the efficiency changes by 1%. We have studied these effects extensively.

J.L. LEROY: I agree that if you vary these parameters, the change in efficiency is small. But does this prove that the absolute efficiency is correct? You may have neglected something in your analysis which may cause 1% error.

W.P. POENITZ: For just this reason it is our basic principle to measure cross-sections by as many different techniques as possible. Reasonable agreement among the various results serves as an indirect check of the whole procedure.

F. KÄPPELER: Did you have count-rate problems or pulse pile-up problems because of the high efficiency of the Black Neutron Detector?

W.P. POENITZ: This problem is discussed in detail in my paper on the ^{235}U fission cross-section. To reconcile the efficiencies of the fission counter and the Black Neutron Detector, we used a double-collimator system to limit the number of neutrons reaching the Black Detector.

F. KÄPPELER: Did you have background caused by captured neutrons?

W.P. POENITZ: The time-of-flight spectra are also shown in my paper on the ^{235}U fission cross-section. We eliminated the background caused by capture γ-rays by the time-of-flight method. Of course, time resolution is not so good as with a smaller scintillator because of the time difference caused by the multiple-collision flight path. But it is good enough to get a very narrow peak and to see the capture γ-rays as a broad, flat background underlying the neutron time-of-flight peak. The capture γ-ray background causes only a very small number of counts in the narrow neutron time-of-flight peak although it may contribute something like 20-30% of the counts in the total time-of-flight range (500 ns) which was recorded.

One could try to eliminate the background due to γ-rays by applying pulse-shape discrimination, but this might affect the efficiency. You will see from the time-of-flight spectra that it is unnecessary anyhow.

C.D. BOWMAN: I have a question about the Grey Neutron Detector. I was concerned about the problem which you might have with γ-rays detected in the sodium iodide crystal which do not come from the sphere, for example the 2.2-MeV γ-rays from capture in the concrete in the walls of the accelerator room. The detector response, I think, is perhaps not fast enough to eliminate this kind of background by timing alone. Also with regard to the use of this kind of detector for linac measurements with the black resonance technique, such as Mr. Coates described earlier, I do not think one eliminates, with the black resonance technique, the neutrons that come down the flight path, scatter off the detector, hit the floor and come back within a time less than the width of the resonance.

W.P. POENITZ: Let me answer my part of this question first, and then perhaps Mr. Coates may wish to speak.

We have completely surrounded our Van de Graaff source with shielding. Our detector is behind a very big lead shield, which is part of the large liquid scintillator located nearby, so that the detector is shielded from γ-rays which come directly from the source shield.

Everything which goes into the room and which in some way comes back into the detector would cause some background. It is measured by plugging off the hole in the beam collimator. In addition, the time-independent or machine-independent background is measured.

C.D. BOWMAN: But when you plug off the beam, you also eliminate the neutrons which leak from the detector sphere and are captured in the walls of the room and which therefore contribute to the background.

W.P. POENITZ: Very few neutrons, perhaps 1%, which enter the detector come out of it.

C.D. BOWMAN: Is that true even at 1 or 2 MeV?

W.P. POENITZ: Our detector is much larger than the one which Mr. Coates described. Ours has a 38-cm effective radius sphere.

C.D. BOWMAN: Would the plug also eliminate a neutron scattered off the inner walls of the collimator?

W.P. POENITZ: That raises the question of collimators, which I think is a separate one, but an important one for the use of this detector.

It is difficult to see how the space angles which extend between the collimator, the source and the detector could allow a background effect. To check this, we measured a time-of-flight spectrum at the entrance hole to the Grey Neutron Detector. At Karlsruhe we used a $^{10}B(n,\alpha)$ detector with about 1-ns resolution. The timing equipment was available because it was anyway used in the measurement of fission or capture cross-sections. We could make sure that the known prompt or known monoenergetic components were less than 1%.

M.S. COATES: Mr. Bowman inquired about the black-resonance technique's not picking up neutrons which were scattered within the time band of the 'black' channels.

(1) The time band of the black channels is very short. It is difficult to think how neutrons could be scattered and then enter the detector again within this time limit.

(2) With the Black Detector only a very small fraction of the neutrons is scattered out of the detector.

(3) In the transfer detector, where many neutrons are scattered, it was possible to inject a monoenergetic pulse of neutrons produced with the Van de Graaff (IBIS) and to check the time distribution of the detector's response. When we did this, we had exactly the same shielding around the detector as when we used it on the linac. Very few neutrons are detected in the period of time to which you refer.

C.D. BOWMAN: The time scale is not necessarily short. You proposed to use a flight path of 100-200 m, so the time range might be of the order of 1-10 μs for an appropriate black resonance. Therefore you cannot rely on timing alone to eliminate problems unless you use a sufficiently wide time gate.

M.S. COATES: In the Van de Graaff work, the measurement of background depends on how infrequently the machine can be pulsed. Normal operation was 1 μs between pulses, and we increased this to 10 μs. If the fraction of neutrons in the tail of the distribution does not change as the repetition rate is changed, one can have some confidence in the results.

E. MIGNECO: With regard to the use of the black-resonance technique in linac experiments, I share the same fears as Mr. Bowman, especially for measurements in the lower-energy region. The only way to be sure of the black-resonance technique is to keep the background so low that any error introduced in the analysis is negligible. Some other way should be found to verify at least once that the background really is low. Unless this can be done in a rather obvious and simple way, I would be afraid to assign high precision to an experiment which uses this technique.

M.S. COATES: I would agree with the point of keeping the background low, and that is what one tries to do. I think it is certainly true that the black-resonance technique is not perfect, but it is one of only two which can be applied. The other way to tackle the background problem at higher energies is to measure a well known cross-section, for example carbon or hydrogen, where the cross-section is varying rapidly with energy. Then by suitable combination of the results for different sample thicknesses, one can arrive at some conclusion about what the background is and how it is attenuated. One actually tries to feed in background neutrons. In the case of the carbon cross-section, which increases with decreasing energy, if the background is caused by faster neutrons colliding with collimators of flight tubes or whatever, there will be relatively more of them when

there is a sample in the beam. By varying the sample thickness, you can work it out.

In fact, the black-resonance technique is the only one we used in this particular set of experiments, although we are using the other one as well.

W.P. POENITZ: This discussion of background problems is relevant for all data which come from white source measurements; with a pulsed, monoenergetic neutron source, one has a completely different background problem which is much more difficult to check.

PRECISE NEUTRON FLUX MONITORING BY T(p, n)^{3}He ASSOCIATED-PARTICLE COUNTING BETWEEN 0.25 MeV AND 1.3 MeV

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Abstract

PRECISE NEUTRON FLUX MONITORING BY T(p, n)^{3}He ASSOCIATED-PARTICLE COUNTING BETWEEN 0.25 MeV AND 1.3 MeV.

The associated-particle method has been used to monitor neutron flux produced by the T(p, n)^{3}He reaction in the energy range 0.25 MeV to 1.3 MeV. $^3He^{++}$ particles were counted with a solid-state detector after unwanted charged particles had been eliminated with an analyser which employed both electric and magnetic fields. Associated-particle counting has been used independently to confirm a previous calibration of a long counter by the $MnSO_4$ bath method within experimental errors of both methods.

1. INTRODUCTION

When the T(p, n)^{3}He reaction is used to produce neutrons, a ^{3}He particle is associated with each emitted neutron. This property can be used in two different ways:

(1) When establishing coincidences between ^{3}He particles and neutrons, the ratio of the number of coincidences to the total number of counts given by the ^{3}He counter is equal to the neutron detection probability. This probability is also the efficiency of the neutron detector if the arrangement is such that every neutron associated with a counted ^{3}He particle impinges on the neutron detector and if the detection probability is independent of the impact point. The associated-particle method has very often been used with the D(d, n)^{3}He and D(T, n)^{4}He reactions. It has been used for the first time with the T(p, n)^{3}He reaction by Fort and Leroy [1, 2] for measuring the efficiency of a ^{6}Li glass scintillator.

(2) An absolute counting of the ^{3}He particles can be made over a definite solid angle. From this measurement and the kinematics of the reaction, it is possible to calculate the neutron flux in the associated direction. This method was used for the first time with the T(p, n)^{3}He reaction by Liskien and Paulsen [3, 4].

This paper describes a new version of the second associated-particle method.

2. DESCRIPTION OF THE METHOD (Fig. 1)

The ^{3}He particles are detected at 10° or 30° from the direction of the beam. The target is a 200 $\mu g/cm^2$ TiT layer on a 200 $\mu g/cm^2$ aluminium foil. As the beam passes through the target, scattering of protons from titanium, aluminium and tritium sends a large number of protons and tritons towards the detector. An analyser combining electric and magnetic fields is used to eliminate these unwanted particles.

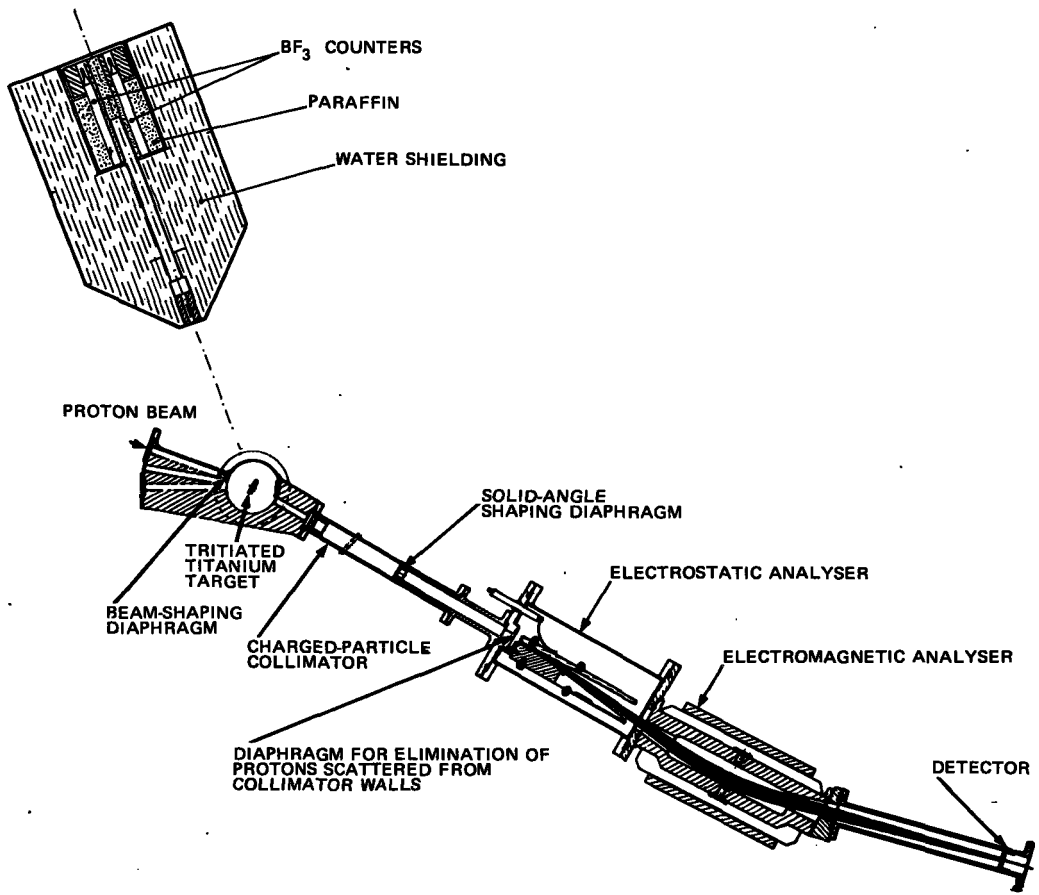

FIG. 1. Arrangement of the experimental apparatus showing the charged-particle analyser and the reference detector to be calibrated.

2.1. Description of the analyser

As an absolute counting is to be made, all the unwanted protons and tritons have to be eliminated by the analyser without loosing any of the ^{3}He particles emitted at the solid angle defined by the entrance collimator. The trajectory deviations caused by the analyser are small, and therefore the small-angle approximation is applicable. In this approximation, the deviation given by the electric field is:

$$\frac{ZV\ K_1 \ell_1}{E}$$

The deviation given by the magnetic field is:

$$\frac{ZH\ K_2 \ell_2}{\sqrt{mE}}$$

As these deviations are of opposite direction, the total distance between the centre of the detector and the collimator axis is:

$$a_0 = - \frac{ZV\ K_1 \ell_1}{E} + \frac{ZH\ K_2 \ell_2}{\sqrt{mE}} \qquad (1)$$

where

Z = charge, E = energy, m = mass } of the particle
V = voltage applied to the electrostatic analyser
H = magnetic field
ℓ_1, ℓ_2 = distances between the detector and the centre of the electrostatic (1) or magnetic (2) field
K_1, K_2 = constants depending on the geometry

As the magnetic field is proportional to the current I in the coils, from Eq. (1) the following equation can be obtained:

$$\frac{I}{\sqrt{mE}} = \frac{\alpha}{Z} + \beta \frac{V}{E} \qquad (2)$$

where α and β are constants of the apparatus. This relation is used as a guide to make the necessary adjustments of the analyser for a given energy (see Fig. 2). The measured count rate is plotted as a function of I in Fig. 3. The width of the plateau of the curve is defined as the 'no-loss domain'; this domain is also plotted in Fig. 2. As the thickness of the target is often a significant fraction of the range of the ^{3}He particles, the energy spectrum of the particles is often broad.

The analyser should be adjusted in such a way that the energy dispersion of the particles does not produce any spatial dispersion at the detector

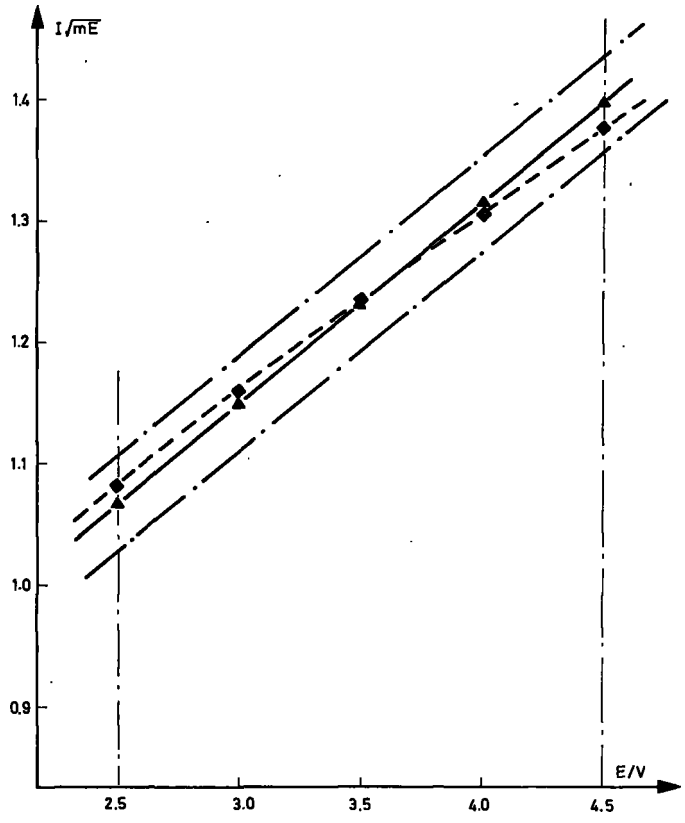

FIG. 2. Current supplied to the magnetic analyser as a function of the ^{3}He energy, E, for fixed electrostatic-analyser voltage, V, and also as a function of V for fixed E, in the 'no-loss domain' for particles of charge 2+. ▲ Function of V for fixed E; ■ Function of E for fixed V.

location. This condition is achieved in first order if the deviation due to the magnetic field is twice the deviation due to the electric field. In this case, $da_0/d_E = 0$.

The compensation obtained in this way is valid as long as the value of I obtained for a fixed V from Eq. (2) remains inside the 'no-loss domain' defined above. The dashed curve in Fig. 2 shows an example of I as a function of E for a defined V. It can be seen from Fig. 2 that a variation of E by a factor 1.8 is allowed without a significant counting loss.

2.2. Measurement of the analyser transmission

It is important to verify directly that all the ^{3}He particles passing the entrance collimator are collected at the detector. This was done with a proton beam elastically scattered from an aluminium foil. The scattered

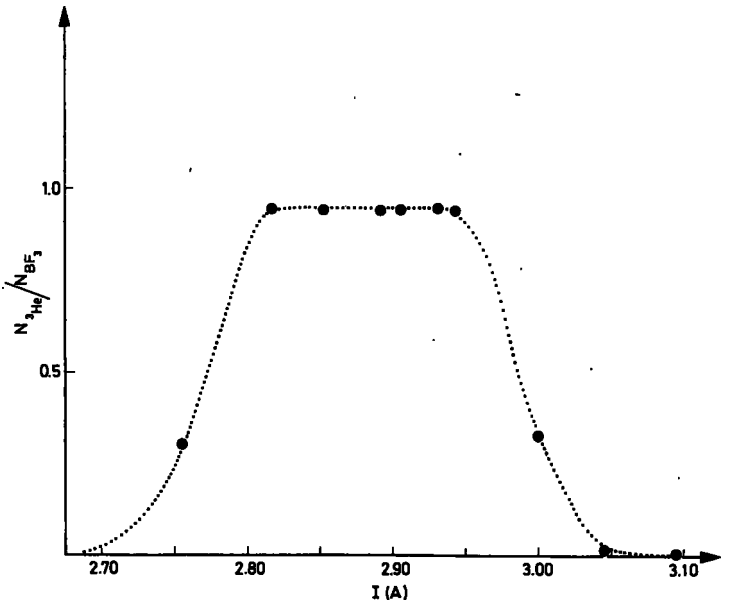

FIG. 3. Variation of the ^{3}He count rate of the charged-particle analyser as a function of current supplied to the magnetic analyser for a given ^{3}He energy and voltage applied to the electrostatic analyser. The plateau in the curve is the 'no-loss domain'. $E_{^3He}$ = 1.95 MeV, $V_{analyser}$ = 6 kV.

protons passing through the collimator were counted in the usual arrangement and directly without the analyser. Another solid-state detector directed at the target was used as a monitor. The ratio of the two count rates was 1.004 ± 0.01. This result shows that there is no significant loss of particles. The experiment was repeated with several energies of the incoming beam without any change to the analyser adjustment in order to check the energy variation allowed without loss. The results were in agreement with section 2.1.

2.3. Charged-particle collimator

The useful solid angle of the analyser was defined by a rectangular slit of 2 mm × 1 mm, placed at 1 m from the target. The surface of the slit was measured using an optical magnifier. The diameter of the incoming proton beam was limited to 2 mm by a diaphragm. If proton scattering occurred at microscopic irregularities of the defining slit, some of these protons could reach the detector. To eliminate this background, a second slit, 3 mm wide, was placed at 20 cm from the defining slit.

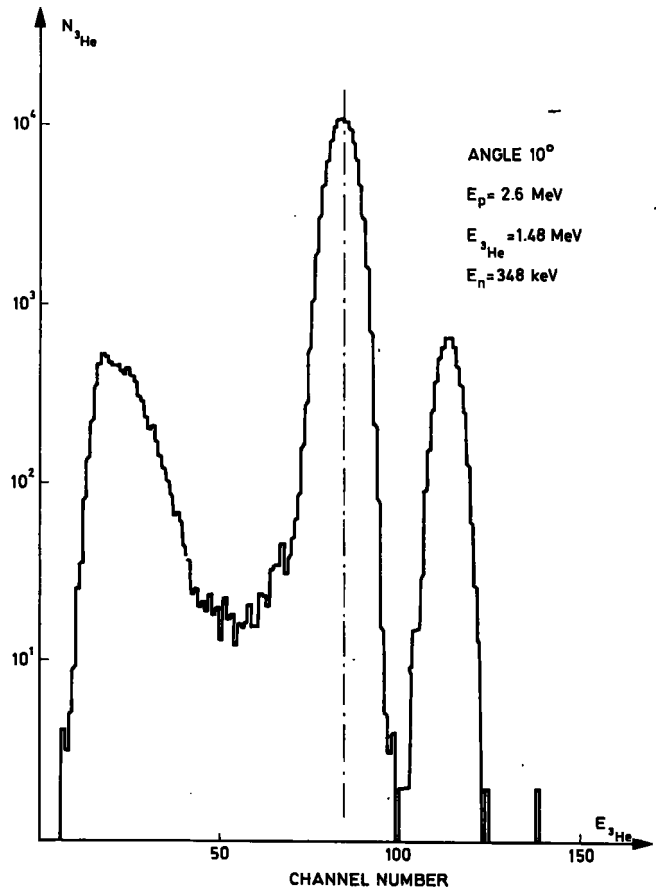

FIG. 4. ^{3}He pulse-height spectrum from the solid-state detector of the charged-particle analyser. Note logarithmic ordinate.

2.4. ^{3}He absolute counting

Figures 4, 5 and 6 show two examples of the pulse-height spectrum obtained from the solid-state detector. The main peak corresponds to the $^3He^{++}$ associated with the neutrons. The smaller peak at higher energy is due to ^{3}He recoil produced by proton scattering on the small ^{3}He content of the target from tritium decay.

When a non-tritiated target was used, the count rate at the place of the main peak was divided by at least 1000. The peak-to-valley ratios of the spectra were never worse than 50 and are generally between 100 and 1000.

The low-energy side of the main peak had a tail which extended to lower energies than could be expected from the thickness of the TiT target.

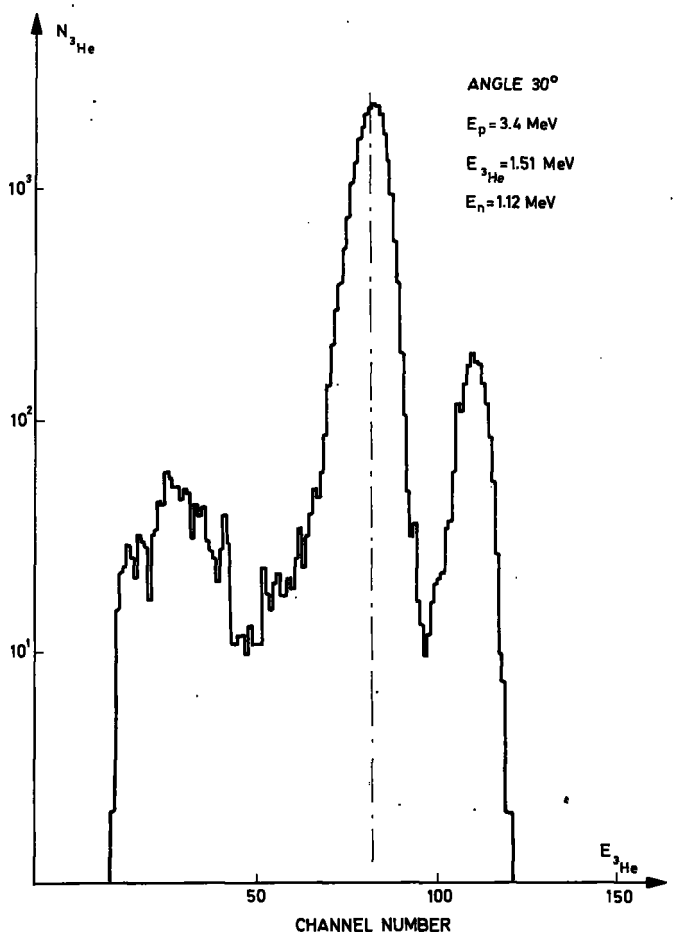

FIG. 5. ^{3}He pulse-height spectrum from the solid-state detector of the charged-particle analyser. Note logarithmic ordinate.

The area under this tail was 2-3% of the total number of counts. When the spectrum of the ^{3}He particles in coincidence with neutrons was measured in order to analyse the meaning of this tail, the tail was reduced to 0.5% of the peak area. The low-energy ^{3}He particles probably came from scattering inside the target and should not have been counted. Therefore, the lower limit of the peak was obtained from an extrapolation of the low-energy side of the peak.

The target could tolerate a beam of 1-2 μA. A typical value of the ^{3}He count rate was 25 counts/μC.

2.5. Charge-exchange correction

Because of charge-exchange phenomena, some of the ^{3}He particles emerging from the target have captured one or two electrons. In the range

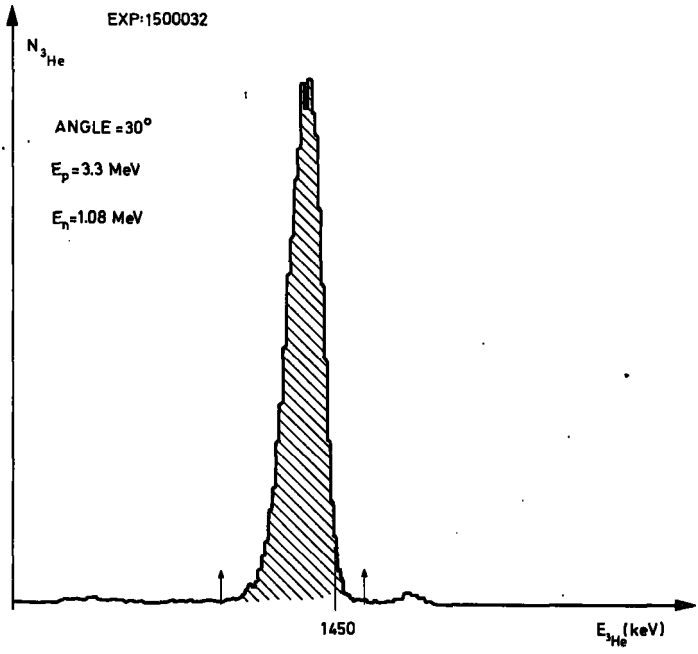

FIG. 6. ^{3}He pulse-height spectrum from the solid-state detector of the charged-particle analyser. Note linear ordinate.

of interest (1-3 MeV), the proportion of neutralized ^{3}He is very small. The proportion of $^3He^+$ can be measured directly when the electric and magnetic fields used during the $^3He^{++}$ counting are multiplied by two. The ratio $^3He^+/^3He^{++}$ measured in this way is in good agreement with the values given by Armstrong et al. [5]. According to them, the charge equilibrium fraction for ^{4}He is given by:

$$\frac{^4He^+}{^4He^{++}} = (0.273 \pm 0.005)\, E^{-(2.00 \pm 0.02)} \quad (E \text{ in MeV})$$

This result was transposed for ^{3}He, assuming that the effect depends only on the speed and charge of the particle. The charge equilibrium fraction for ^{3}He is then:

$$\frac{^3He^+}{^3He^{++}} = 0.154\, E^{-2} \quad (E \text{ in MeV})$$

3. APPLICATION OF THE METHOD TO THE CALIBRATION OF A REFERENCE DETECTOR

The reference neutron detector, a collimated long counter, has been described elsewhere [1]. It has been calibrated using the manganese bath technique [2]. This experiment was intended as a check.

The efficiency of the neutron counter is

$$\epsilon = \frac{C_n (1-a)\, \Omega_\alpha}{C_\alpha\, \Omega_n} \cdot R \cdot f_{air}$$

where

C_n = count rate of the neutron counter
C_α = count rate of the ^{3}He detector
a = fraction of single-charged ^{3}He particles
Ω_α = solid angle of the analyser
Ω_n = solid angle of the neutron counter
R = laboratory to centre-of-mass conversion factor
f_{air} = air-scattering correction factor

The solid angle of the neutron counter Ω_n is computed according to the method described in Ref. [2]. It includes the edge-effect correction factor. The neutrons scattered from the target holder and its surroundings are eliminated by the collimator of the neutron counter. Shadow-bar measurements are made to determine the remaining background.

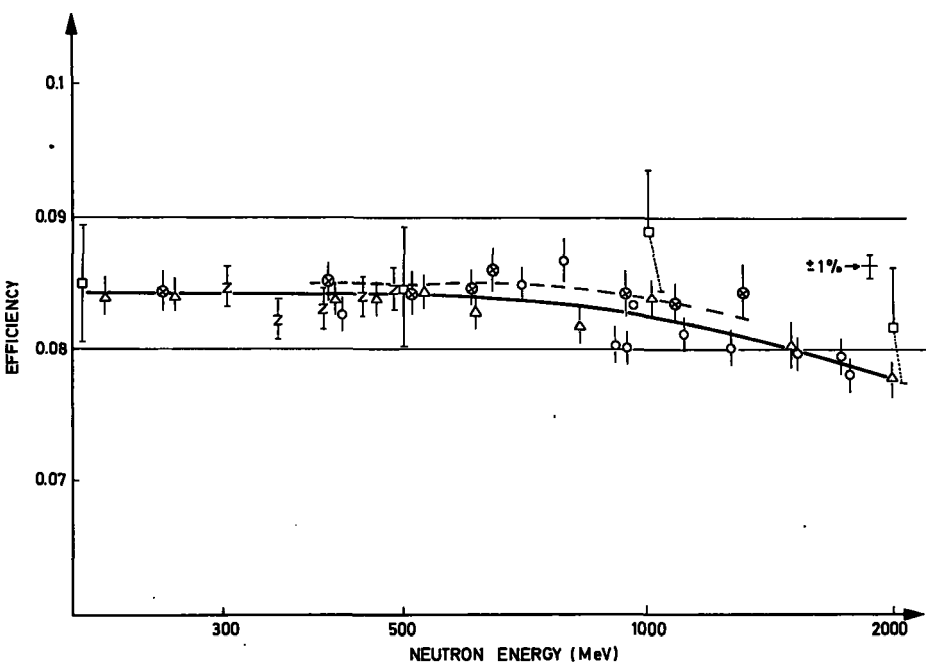

FIG. 7. Comparison of the efficiency of a long counter determined by various experimental methods and by Monte-Carlo calculation. □ Monte-Carlo calculation, △ Manganese bath (Oct. 1969), ○ Manganese bath (Dec. 1970), ⊗ Associated-particle (June 1972), Z Associated-particle (Nov. 1972).

TABLE I. ESTIMATED ERRORS IN THE ASSOCIATED-PARTICLE METHOD (%)

	At present	Possible after further development
Solid angle { neutron counter	0.5	0.5
Solid angle { ^{3}He collimator	1.0	0.5
Analyser transmission	1.0	0.5
Contribution of $^3He^+$	0.5	0.3
Tail of $^3He^{++}$ peak	1.0	0.5
Background	0.1	0.1
Air scattering	0.3	0.3
Statistics	0.5	0.5
$\left(\sum_{i=1}^{n} \sigma_i^2\right)^{\frac{1}{2}}$ =	≈2.0	≈1.3

4. RESULTS

Measurements were made for neutron energies between 0.250 MeV and 1.3 MeV. For the lower part of this range the ^{3}He particles were detected at 10°, for the higher part at 30°. The range could be extended to cover the range 0.2 MeV to 1.75 MeV. The results are shown in Fig. 7, together with the previous results from the $MnSO_4$ bath method [2]. Both experiments are in agreement, within the error bars, although the values from the associated-particle method are 1% higher on the average. The estimated error from the $MnSO_4$ bath was ±1.8%. The typical errors of the present experiment are given in Table I together with the errors expected after further development.

5. CONCLUSION

Previous calibrations of a long counter by the $MnSO_4$ bath method have been confirmed by the present measurements using the associated-particle technique, a fully independent method.

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DISCUSSION

C.D. BOWMAN: How is the long counter used?

J.L. LEROY: It is used to measure the neutron flux, for example in a fission cross-section measurement.

C.D. BOWMAN: Then the question of background becomes very important since the long counter is a slow time-response system. The calibration technique seems very clean without significant problems of background.

J.L. LEROY: We put a shadow bar in front of the collimator. With the collimator plugged like this, the background was 0.5% to 3% depending on energy. Of course the background is greater at higher energies because it is due mainly to neutrons going through the shielding, and this effect is greater at 2 MeV than at 300 keV.

C.D. BOWMAN: You have just described the background during calibration of the long counter. My point is that when you measure a fission cross-section using the long counter for measuring the flux but without the 'associated particle', the background conditions are different.

J.L. LEROY: No, the background measurement technique is the same during a fission cross-section measurement. Neutrons may be produced by the ^{7}Li(p, n) reaction. The flux is measured at 20°, for instance, and the fission chamber is at 20° in another direction. It is easy to put a shadow bar between the target and the neutron detector to make the background measurement. The device has a good signal-to-noise ratio; it is not sensitive to neutrons scattered by the wall.

E.J. AXTON: Was the water shield in your detector loaded with a thermal absorber?

J.L. LEROY: At present it is not. Maybe this would be an improvement.

E.J. AXTON: Then you get some thermal neutrons leaking from the detector into the counter?

J.L. LEROY: Yes, but these are taken into account in the calibration.

ABSOLUTE NEUTRON FLUX DETERMINATION IN THE ENERGY REGION BETWEEN 0.4 MeV AND 2 MeV

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Abstract

ABSOLUTE NEUTRON FLUX DETERMINATION IN THE ENERGY REGION BETWEEN 0.4 MeV AND 2 MeV.

A detector for absolute neutron flux determination in the energy range between 0.4 MeV and 2 MeV is described. It is based on the hydrogen scattering cross-section as a standard and works similarly to a proton recoil telescope. The main advantage is that corrections are almost entirely avoided. To establish its reliability, the influence of all parameters which determine the accuracy has been checked carefully. In this way, an overall uncertainty of the determined neutron flux of about ± 2% was reached.

1. INTRODUCTION

For the experimental determination of any partial neutron cross-section, it is essential to know the value of the absolute neutron flux. The neutron flux can be determined by counting the events of a neutron reaction, the cross-section of which is well known as a function of energy and therefore can be used as a standard. In the energy range above a few hundred keV, the only standard cross-section with an uncertainty of less than 1% is the hydrogen (n, p) cross-section [1]. In the case of thin samples, the neutron flux can be determined from the simple relation

$$\phi = \frac{Z}{\sigma_H \cdot N_H \cdot \epsilon_H} \qquad (1)$$

where ϕ = neutron flux
Z = detector count rate
σ_H = hydrogen scattering cross-section
N_H = number of hydrogen atoms
ϵ_H = detector efficiency

The advantage of the hydrogen scattering cross-section as a standard led to the use of the well known proton recoil detectors [2] in a variety of experiments. However, except for energies above 2 MeV where telescope counters can be used, the accuracy of the proton recoil detectors is limited. Their low-energy background requires an extrapolation to zero pulse height in the energy distribution of the recoil protons and hence a substantial correction of ϵ_H. Moreover, the efficiency is dependent on threshold stability as well as on neutron energy. Other problems arise from the determination of the number of atoms in the case of a plastic scintillator or from volume effects and scattered neutrons in the case of gas-filled counters.

To avoid these difficulties, various other methods have been developed which are not based on the hydrogen scattering cross-section. Examples are the associated-particle method [3], activation methods [4] or the use of flat response counters [5], which allow a shape measurement that may be calibrated at some well known points. But in most cases the accuracy achieved was hardly better than that of the proton recoil detectors.

In this paper, the application of a method [6] is described which extends the advantages of the proton recoil telescope counter to lower energies. Thus it has been possible to use the hydrogen scattering cross-section as a standard without the above-mentioned uncertainties of the ordinary proton recoil detectors.

2. DESCRIPTION OF THE DETECTOR

The principal features of the detector can be seen in Fig. 1. Recoil protons are produced in a thin layer of hydrogenous material (the radiator) and are detected by a solid-state detector placed at a certain distance from the radiator. By this arrangement of radiator and detector, only those recoil protons hit the detector which have been scattered through small angles θ. Because of the kinematics of the scattering process, which can be described by

$$E_p = E_n \cos^2 \theta$$

these protons have the highest possible energies. Figure 2 shows the pulse-height distribution of the recoil protons measured with this arrangement

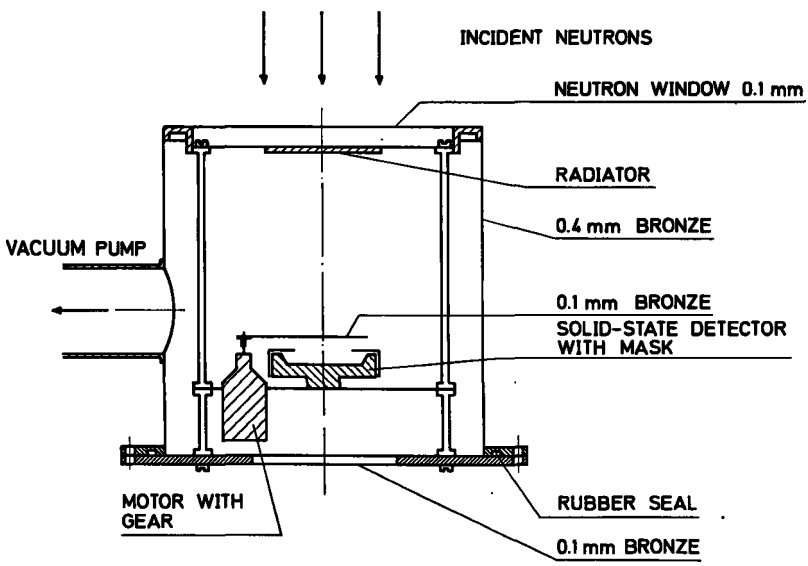

FIG. 1. Schematic view of the proton recoil detector.

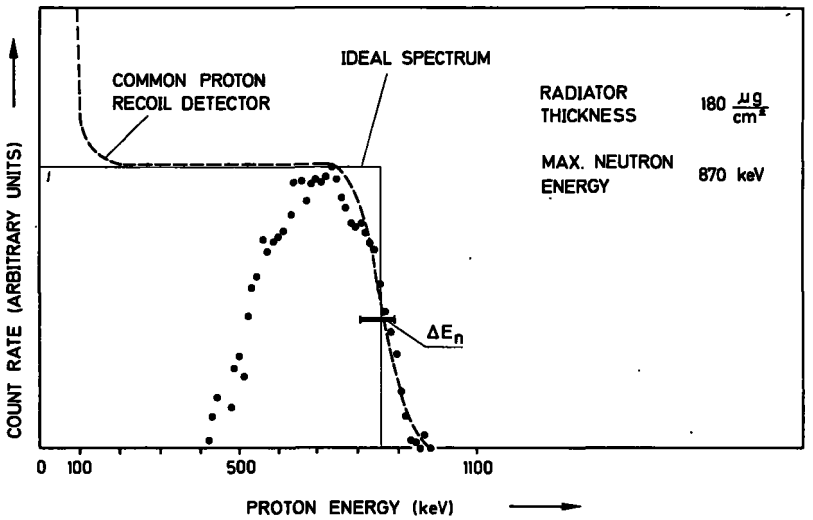

FIG. 2. Comparison of pulse-height distributions of recoil protons measured with the experimental detector and of those measured with a common proton recoil detector.

and, for comparison, a schematic spectrum of all possible recoil protons that would be expected for a gas-filled or plastic scintillation detector. It is clear that the new detector shows the advantages of the telescope counter concerning threshold and extrapolation problems. Some disadvantages are incurred, however, i.e. coincidence measurements for background suppression are not possible because of the low proton energy, and the efficiency is relatively small since the radiator thickness is limited.

The background determination was performed by shielding the recoil protons from the solid-state detector. A thin sheet of bronze was moved by a small motor between definite positions, thus covering the solid-state detector during every second cycle of an automatic sample changer. In this way, the background spectrum could be measured directly. Test runs without radiator ensured that there were no differences greater than a statistical uncertainty of 0.5% between the spectra measured with covered and uncovered solid-state detector. These runs also showed that there were no hydrogenous contaminations on the counter walls. During operation the counter was evacuated to better than 10^{-4} torr. A cooling baffle with liquid nitrogen prevented the diffusion of oil vapour into the counter.

The recoil protons were registered in a surface barrier counter of 100 μm thickness and 450 mm^2 sensitive area (Ortec A-030). It had an entrance window of 40 μg/cm^2 gold and an energy resolution of 30 keV for 5.5-MeV α-particles.

The detector thus described was used in an absolute measurement of the neutron fission cross-section of ^{235}U, which is reported in paper IAEA-PL-246-2/27 in these Proceedings. In section 3, the characteristic features of the flux measurement are described in detail. All further statements refer to the energy region of this experiment, between 0.5 MeV and

1.2 MeV, although the whole useful energy region of the counter ranges from 0.4 MeV to 2 MeV. It is limited at low energies because of background problems and at about 2 MeV by the onset of neutron reactions in the solid-state detector.

3. EXPERIMENTAL DETAILS

3.1. Neutron source

The measurement was performed at the Karlsruhe 3-MeV pulsed Van de Graaff accelerator with a pulse-width of 1 ns and a repetition rate of 2.5 MHz. Neutrons were produced by the $^{7}Li(p, n)^{7}Be$ reaction using metallic lithium targets on thin tantalum backings. The target thickness was between 40 and 50 keV.

The neutron spectrum was measured with the time-of-flight method by a lithium glass detector at 3.20 m distance from the target and a time resolution of better than 1 ns/m. This allowed the energy determination with an uncertainty of somewhat less than ± 5 keV. The width of the neutron energy distribution was determined by the target thickness. It was calculated from the time-of-flight spectra for each run separately with an uncertainty of ± 10%.

3.2. Samples

The high hydrogen content makes substances of the type $(CH_2)_x$ especially suited as radiator samples. The radiators must be thin enough so that the energy loss of the protons is small compared with their energy. Therefore, evaporated layers of stearic acid ($C_{18}H_{36}O_2$) as well as of glycerol tristearate ($[C_{17}H_{35}COO]_3\ C_3H_5$) on thin stainless-steel backings have been used as radiator samples. These substances are available in a very pure chemical form. Their low melting point allows a clean evaporation process free from contaminations.

TABLE I. THICKNESS, MASS AND UNCERTAINTY OF THE RADIATOR SAMPLES

Substance	Radiator thickness ($\mu g/cm^2$)	Total mass (mg)	Uncertainty (%)
Stearic acid	218	5.775	0.5
	230	3.191	0.9
	229	3.170	0.9
	115	1.596	1.8
Glycerol tristearate	198	5.242	0.5
	152	4.012	0.7
	140	3.685	0.8
	102	1.888	1.4

The samples were weighed with a microbalance before and after evaporation. In this way, the sample masses were determined with an uncertainty of about ± 30 μg. To exclude systematic uncertainties, several different radiators were used throughout the experiment. Their masses are listed in Table I.

After the measurements had been finished, the hydrogen and carbon content of the radiator samples was determined by a standard chemical microanalysis [7]. For an analysed amount of 4 mg the standard deviation was ± 0.18% for the carbon content and ± 0.13% for the hydrogen content. Deviations of between 1.9% and 3.0% from the stoichiometric amount were found for the hydrogen content, dependent on how long the samples had been in vacuum. An explanation may be that small amounts of gaseous carbon-hydrogen compounds split from the large molecules during the evaporation process and in vacuum. For these losses a correction was made assuming a linear time dependence. The resulting uncertainty was estimated to be 0.8%.

3.3. Efficiency

In the following, the counter efficiency, ϵ_H, is defined as the ratio of detector events to all events which happened in the counter. As only the forward-peaked recoil protons are detected, ϵ_H is determined by the inner geometry of the counter. From the radii of radiator and entrance aperture of the solid-state detector and the distance between them, the efficiency was calculated using the Monte-Carlo program described in Ref. [6]. The values of ϵ_H for the different counter geometries used are shown in Table II. In the investigated energy range, the dependence of ϵ_H on the neutron energy was negligible (see also Ref. [8]).

The overall efficiency of the proton recoil detector depends on ϵ_H, on the sample thickness and on the hydrogen scattering cross-section. Under the conditions of this experiment, the overall efficiency was roughly 4×10^{-6}.

TABLE II. VALUES OF THE COUNTER EFFICIENCY ϵ_H FOR THE DIFFERENT COUNTER GEOMETRIES USED DURING THE EXPERIMENT

Diameter of solid-state detector mask (mm)	Efficiency, ϵ_H ($\times 10^{-2}$)	Uncertainty (%)
16.75	1.999	0.8
17.92	2.277	0.8
19.00	2.537	0.8

3.4. Electronics

After passing through the pre-amplifier and timing amplifier, the pulses of the solid-state detector were divided into a pulse-height and a time branch. The pulse-height and time signals of each event were coded in analogue-to-digital converters (ADC) and stored in the memory of a 16-K computer as a two-dimensional spectrum. Independently, a second pulse-height spectrum

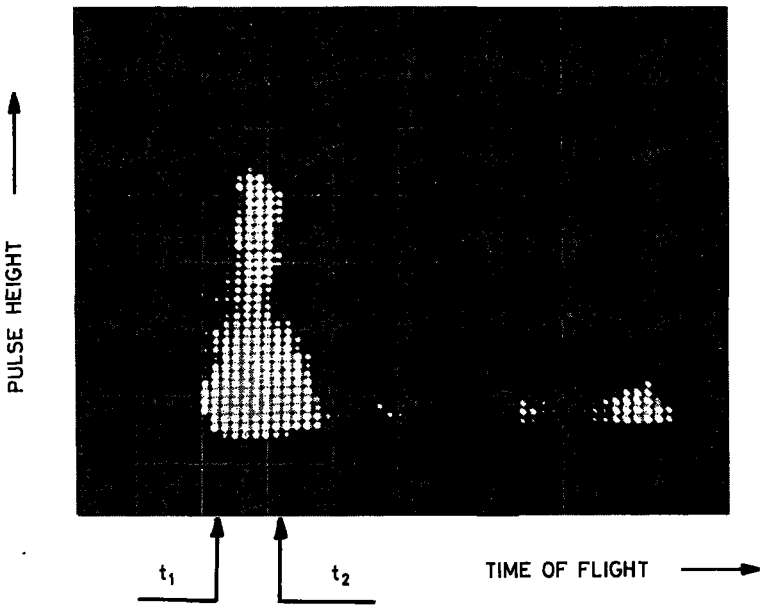

FIG.3. Horizontal cut of the two-dimensional spectrum of the proton recoil counter.

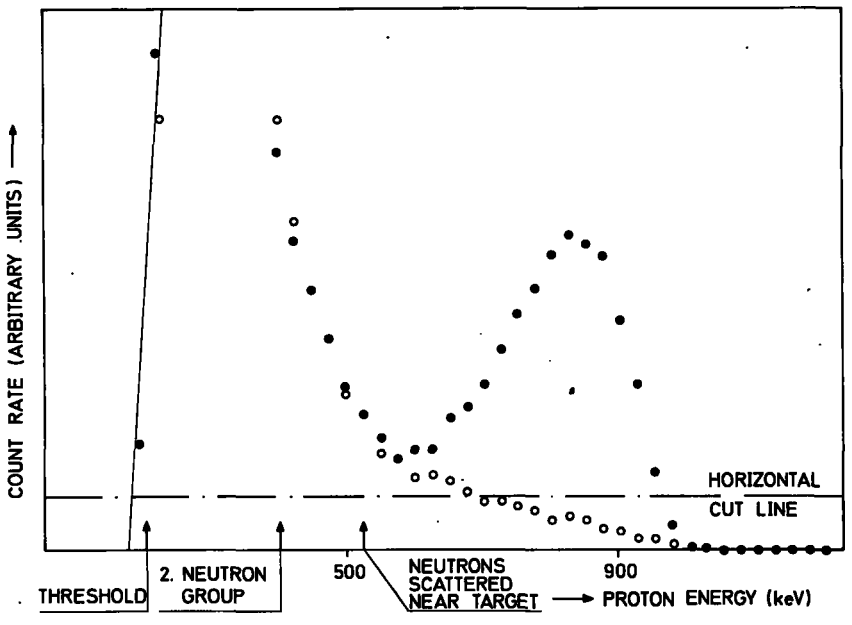

FIG.4. Pulse-height spectrum of the recoil protons between the flight times t_1 and t_2 (E_n = 909 ± 22 keV).

was accumulated in a multi-channel analyser. This was used for control and for a better survey. Every ADC was coupled with the automatic sample changer mentioned above, so that the spectra, with the solid-state detector covered and uncovered, were stored in two different fields of the respective memories.

Figure 3 shows a horizontal cut through the two-dimensional spectrum of the proton recoil counter at a neutron energy of 909 ± 22 keV. At low pulse heights, there is an overlap between the band-like recoil proton distribution and the broader background of the solid-state detector. In Fig. 4, a cut along the pulse-height axis is shown, containing all events with flight times between t_1 and t_2. The corresponding distribution with the solid-state detector covered is shown by the open circles. The horizontal cut line of Fig. 3 is drawn as a dashed-dotted line in Fig. 4.

The background at low pulse heights is somewhat dependent on the neutron energy. It generally limits the useful energy region of the counter for low neutron energies. At about 400 keV, background and recoil protons become comparable and this causes a very high statistical uncertainty. The time resolution of the counter is about 12 ns in the energy range of this experiment.

3.5. Corrections

Besides the correction concerning the deviation from the stoichiometric amount of hydrogen in the samples, there is another correction necessary which accounts for the interaction of scattered neutrons with the detector. Several different types of scattered neutrons may falsify the detector count rate. The wall-scattered neutrons lead to a time-independent background and can easily be subtracted in the time-of-flight spectra. Two other types of scattered neutrons can be discriminated in the pulse-height distribution because of their low energies. The maximum energies of these groups are indicated in Fig. 4 by arrows. The first type are neutrons which have been scattered in the target region. Their low energy is due to the kinematics of the neutron-producing reaction, which causes a strong dependence of the neutron energy on the angle with respect to the incident protons. The second type originates from the $^7Li(p, n)^7Be^+$ reaction in which the beryllium nucleus is left in its first excited state. For primary neutrons below 1.2 MeV, this second group always has energies lower than the first type of background neutrons. Therefore, the only correction necessary is for neutrons scattered in the counter or by the nearby surroundings. In this case, the working principle of the proton recoil counter restricts drastically the region from which neutrons can be scattered and produce recoil protons with correct energies. This region contains only the plane end of the cylindrical counter. The contribution of these scattered neutrons to the count rate was calculated by a Monte-Carlo program for a slightly simplified geometry from the differential scattering cross-sections of the corresponding materials. The result was that in the whole energy region from 400 keV to 1.2 MeV the scattering correction is smaller than 0.5%.

4. CONCLUSIONS

It was shown by experiments that the method described for absolute neutron flux determination can be used in the energy range between about

TABLE III. EXPERIMENTAL UNCERTAINTIES

Source of uncertainty	Uncertainty (%)
Hydrogen (n,p) cross-section, σ_H	0.5
Number of hydrogen atoms, N_H	
Sample mass	0.5 - 1.8
Stoichiometric correction	0.8
Efficiency, ϵ_H	0.8
Count rate, Z_H	
Statistical uncertainty	1.0
Correction for scattered neutrons	0.2
Overall uncertainty	1.7 - 2.4

400 keV and 2 MeV. The reliability of the measurement was carefully evaluated and checked by a systematic variation of all important parameters. For this purpose, the experiment was performed with eight radiator samples of two different materials and with a threefold varied counter geometry leading to three different counter efficiencies. In this way, the results of the sample mass determination, the stoichiometric correction and the determination of the efficiency were proven to be consistent with the experimental uncertainties summarized in Table III.

The achieved accuracy of the neutron flux determination was possible because corrections could be reduced considerably. Extrapolation corrections and corrections for volume effects have been avoided, the scattering correction is almost negligible and the remaining correction for the deviation of the stoichiometric hydrogen content is small and therefore causes a relatively small uncertainty.

All these features make the described method suitable for absolute neutron flux determinations with an overall uncertainty of about 2%.

ACKNOWLEDGEMENTS

The author wishes to thank A. Ernst and D. Roller for their help in data handling and sample preparation, as well as Dr. R. R. Spencer for his reading of the manuscript. Also, thanks are due to the Van-de-Graaff crew for their contribution to this work.

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DISCUSSION

C.D. BOWMAN: Why are carbon recoils not observed?

F. KÄPPELER: Recoil carbon atoms have about 1/12 of the energy expected for protons and therefore do not appear in the observed energy range.

W.P. POENITZ: Can you explain the background in your experiment? At the 1970 Argonne Symposium you showed proton-recoil time-of-flight spectra in which the ratios of the real count rates to background count rates were approximately 1:1, and you could not explain the origin of the background.

F. KÄPPELER: The background depends on the neutron energy. At Argonne, I showed spectra taken at 440 keV and at about 500 keV in which the background was 1/3 to 1/2 of the proton-recoil contribution. The spectrum presented in this paper was taken at about 900 keV, and therefore the background was much lower. I still cannot explain its origin.

NEUTRON FLUX MEASUREMENTS AT THE LLL LINAC

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Presented by C.D. BOWMAN

Abstract

NEUTRON FLUX MEASUREMENTS AT THE LLL LINAC.

Three detector systems to cover three overlapping energy ranges will be used in high-accuracy measurements of the ^{235}U fission cross-section with the LLL linac: (1) Thermal to 100 keV — thin ^{6}Li glass scintillator; (2) 50 keV to 2 MeV — gas-filled proportional counter; (3) 1 to 20 MeV — proton recoil telescope. The status of each detector system is reported.

A programme of neutron-flux measurements is being carried out at the Livermore linac in preparation for high-accuracy fission cross-section measurements on ^{235}U.

It is planned to cover the neutron energy range from thermal to 20 MeV in three overlapping energy regions using the following detector systems: (1) thermal to 100 keV — thin ^{6}Li glass scintillator, (2) 50 keV to 2 MeV — gas-filled proportional counter, (3) 1 to 20 MeV — proton-recoil telescope. The present state of development of these three systems is described below.

We have obtained and are currently using a 1-mm-thick ^{6}Li glass scintillator which is viewed by two RCA-4525 phototubes optically coupled to the scintillator edges as shown in Fig. 1a. The phototubes are 15 cm apart and are placed outside of the neutron beam to minimize scattering. Pulse-height resolution has been optimized by adding the pulses from both phototubes and by surrounding the assembly with an aluminium reflector foil. Figure 2 shows the resolution obtained with a 10-cm-dia. neutron beam centred on the scintillator. The full width at half maximum is less than 20% under these conditions. (Full irradiation of the 10 cm by 15 cm scintillator yields a resolution of 22%.)

Improvements in proportional counter response have concentrated on two areas — decreasing the range of recoil protons and eliminating the effects of recoil carbon nuclei inherent in methane-filled counters. Time-response measurements indicate that a suitable time resolution can be obtained with a broad range of methane-krypton concentrations. The goal of this study is to extend the useful energy range above 1 MeV to afford a suitable overlap with the proton recoil telescope. A search for high-drift-velocity gases other than methane is in progress in an effort to eliminate the carbon recoil problem.

A proton-recoil telescope currently in use with a ^{7}Li glass scintillator as the proton detector is shown in Fig. 1b. A 10-cm-dia. hydrocarbon radiator is placed at the downstream end of a 6-cm-dia. by 50-cm-long lead cylinder and is viewed by the scintillator from a distance of 10 cm.

The 4-cm-dia. scintillator and its attached phototube are shielded from the direct beam by the lead cylinder. Figures 3, 4 and 5 show the pulse-height response obtained at 2.9 MeV, 8.0 MeV and 17.5 MeV.

For all three detector systems, the data are recorded in a two-dimensional mode to allow signal-to-background optimization and to reduce systematic errors caused by base-line shifts.

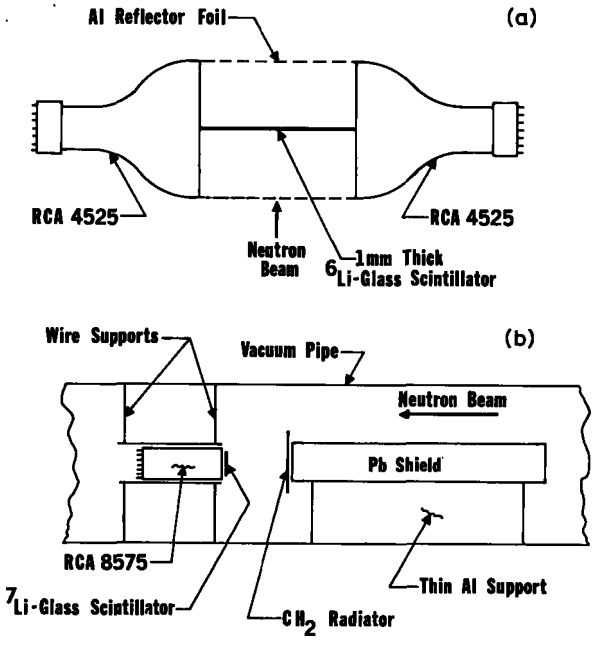

FIG. 1a. The 1-mm-thick ^{6}Li glass scintillator.
b. The geometry of the proton-recoil telescope.

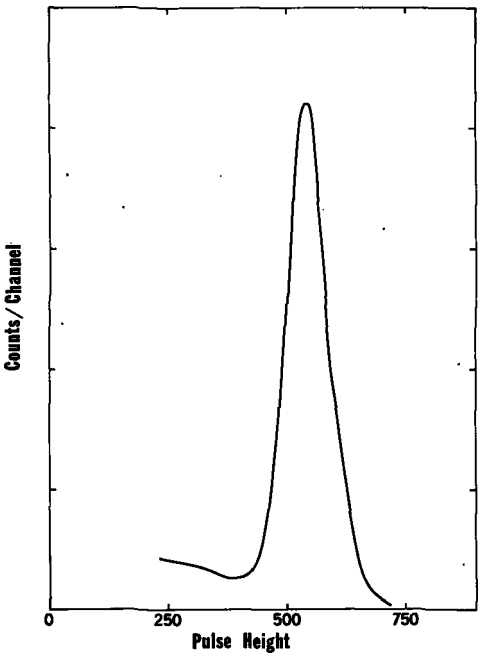

FIG. 2. ^{6}Li glass scintillator pulse-height distribution. Full width at half maximum is 19.5%.

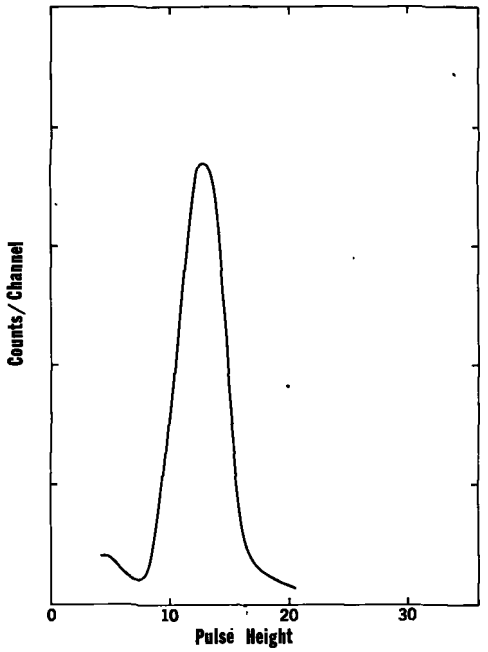

FIG. 3. Proton-recoil telescope pulse-height distribution. The neutron energy was 2.9 MeV with an energy spread of 0.16 MeV.

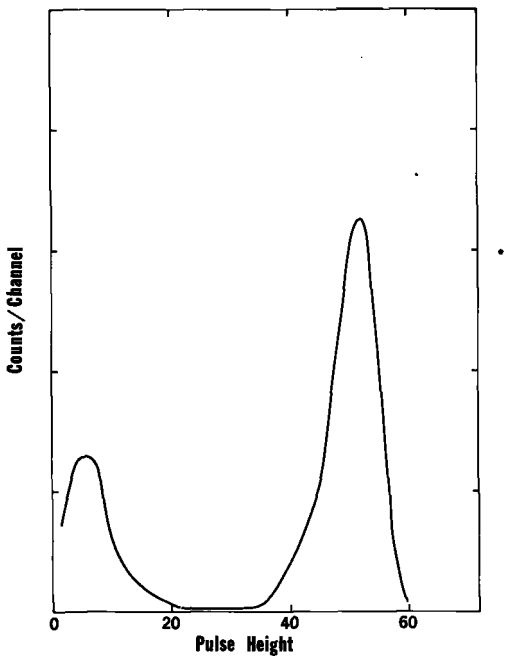

FIG. 4. Proton-recoil telescope pulse-height distribution. The neutron energy was 8.0 MeV with an energy spread of 1.2 MeV. The pulse-height separation between the proton and carbon events is evident.

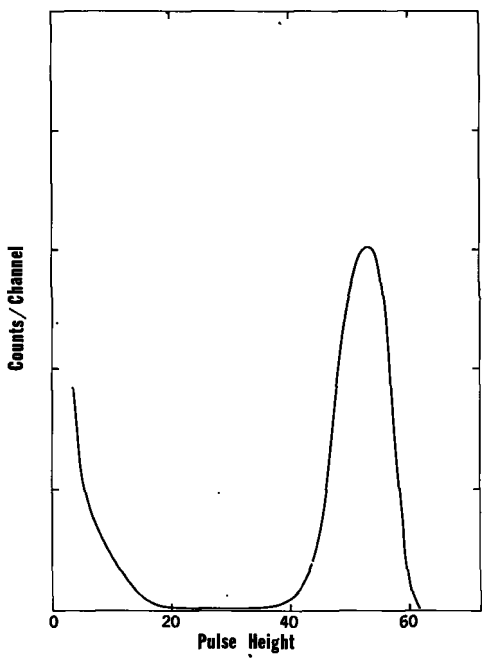

FIG. 5. Proton-recoil telescope pulse-height distribution. The neutron energy was 17.5 MeV with an energy spread of 4.0 MeV. The data for Figs 3, 4 and 5 were not obtained with the same gain conditions.

GENERAL DISCUSSION

H. LISKIEN: I would like to describe briefly some work done at Geel which will appear very soon in Nuclear Instruments and Methods. At the 1970 Argonne Symposium, I described our programme to compare different flux-measuring techniques at various energies in order to discover hidden systematic errors. We have used the conventional recoil telescope above 1 MeV, hydrogen- or methane-filled proportional counters below 3 MeV and the associated-particle method with the T(p, n) and D(d, n) reactions. We had made comparisons at 15 MeV, 2.5 MeV, 1 MeV and 0.5 MeV.

Since the 1970 Argonne Symposium, we have also made a comparison at 250 keV, which is exactly the lowest energy in the range which Mr. Leroy has studied. Our methods were very similar to his; we used associated-particle counting with the $T(p, n)^3He$ reaction. As a second method we used a methane/hydrogen-filled proportional counter with pulse-shape discrimination against gamma rays.

Our associated-particle method differs from the Cadarache arrangement in that, in addition to pulse-height discrimination, we also use an electrostatic field and charged-particle time-of-flight methods for discrimination among the various particles.

We claim 2% accuracy for the associated-particle method, which is exactly the figure Mr. Leroy quoted earlier, and about 3% for the proportional counter.

Although it is always good to have a new device to check consistency, I do not agree with Mr. Käppeler that extrapolation to zero pulse height is such a great problem. We claim 3% for our proportional counter. Combining this accuracy with that of the associated-particle method should give an expected accuracy for the ratio of the two methods of 3.6%. We have made six comparisons of the two methods at 250 keV and we find a precision of 0.7%; for the ratio of the two methods, both of which are in themselves absolute, we find 0.996. We would have expected the deviation of the ratio from unity to be larger than this 0.4%.

J.L. LEROY: Down to what energy do you expect to be able to use the associated-particle method?

H. LISKIEN: The next point where we plan to compare flux-measuring techniques is 100 keV. A comparison at, say, 200 keV would not yield much new information. We do not believe that the associated-particle method can be used at 100 keV. Therefore we made preliminary tests with the associated-activity method using vanadium but found difficulties with background neutrons because the source was so weak. For comparison of the proportional counter with other methods, we found that measurements with the proportional counter were unreliable because of the poor foreground-to-background counting ratio.

Some other type of detector is required to make the measurements at 100 keV and then to make comparisons with the other methods at higher energies. Such a counter either would have to be more efficient or would have to be insensitive to scattered, background neutrons. However, with a long counter and the vanadium source we found that the ratio of neutrons coming directly from the target to those coming from the beam tubes and other places was very unfavourable. This was the reason for my earlier questions to Mr. Coates about calibration of a long counter at Harwell using a vanadium source.

M.S. COATES: This is the work of Adams et al. (UKAEA Report AERE-R 6429). The actual measurements on vanadium were d.c. measurements using proton currents of up to 50 μA. The background was studied with a ^{6}Li glass scintillator and time-of-flight equipment with the Van de Graaff operated in a pulsed mode. It was assumed that if the background were clean and the neutron groups were monoenergetic in the pulsed mode, then the same conditions would appertain in the d.c. mode.

W.P. POENITZ: Mr. Liskien mentioned that he did not think extrapolation of the recoil-proton pulse-height spectrum to zero pulse height was a significant source of uncertainty with the proton-recoil counter which he used. What uncertainty was actually assigned to the extrapolation?

H. LISKIEN: This extrapolation is always made with the aid of a theoretical spectrum calculated with a Monte-Carlo program. The experimental spectrum is compared with the theoretical spectrum step by step. The experimental spectrum is rather flat down to the point where the gamma-ray contribution becomes very strong. Using gamma discrimination shifts this point nearer to zero pulse height so that the gap which must be extrapolated becomes smaller. There is a point where gamma discrimination no longer works, and there extrapolation must begin. The parameters of the Monte-Carlo program are then varied to determine reasonable limits and uncertainties for the extrapolation. (Editor's Note: For details, see LISKIEN, H., PAULSEN, A., Nucl. Instrum. Methods 69 (1969) 70.)

F. KÄPPELER: How did you determine the effective volume of the proton-recoil detector?

H. LISKIEN: The sensitive volume is defined by the 'field tubes' which are inserted in the two ends of the counter to compensate for distortion of the electric field because of end effects. The sensitivity of the counter is then uniform over its entire length and falls off sharply at each end of its effective volume. The effective volume was calculated from the geometry of the counter and, in addition, was checked by scanning along its length with an X-ray beam. The deviations of these methods were included in the uncertainty of the total volume of the detector.

J.L. LEROY: In a proton-recoil counter a small number of lower-energy neutrons can give an incorrect shape to the proton-recoil pulse-height spectrum and thereby cause an incorrect extrapolation to zero pulse height. For instance, a low-energy neutron group might come from scattering of neutrons in the target backing, which is sometimes quite thick. Since the scattering cross-section of hydrogen decreases with increasing energy, the proton radiator has a higher efficiency for low-energy neutrons. If the shape of the theoretical spectrum calculated by the Monte-Carlo code is not exactly the shape of the experimental spectrum, it is very difficult to make the correct extrapolation. We tried to use a proton-recoil counter and had trouble for these reasons.

W.P. POENITZ: We also tried a proton-recoil counter. The Monte-Carlo evaluation could be adjusted in so many ways that we were a little dissatisfied with the extrapolation. That is why I prefer a counter such as Mr. Käppeler described. Incidentally, was not the counter used by White of the latter type? (WHITE, P.H., J. Nucl. Energy 19 (1965) 325.)

F. KÄPPELER: Yes, but White's counter had a very short distance between the radiator and detector so it measured essentially the same spectrum as a gas-filled counter.

W.P. POENITZ: So that was why its spectrum was much broader.

H. LISKIEN: I do not know what Monte-Carlo programs each of you has used, but I should point out one problem with the original Aldermaston program. When the energy of a neutron does not correspond closely with an energy of the program's data tables, the program makes a slight error. The result is a theoretical spectrum with a slope which differs from the experimental spectrum. When this error is corrected, the theoretical spectrum is flatter.

E.J. AXTON: What is the energy dependence of this effect?

J.L. LEROY: We made an overall background measurement and did not measure components due to individual causes. However, I think the effect of thermal neutrons would be small because our Monte-Carlo calculations show that the efficiency does not vary much, at least below 1 MeV; for higher incident energies, thermal neutrons may have greater effect on the background.

M.S. COATES: Was the 4% accuracy of individual points from the Monte-Carlo calculations due to the complicated structure of the detector?

J.L. LEROY: No, it was because of statistics. To obtain an accuracy of 4% for a single point requires 10 minutes of IBM 360/65 computer time. Of course one could achieve better accuracy, but it would be expensive.

W.P. POENITZ: You have shown energy spectra of the ^{3}He particles which you detected. There was one peak due to recoil of ^{3}He particles following scattering of protons on ^{3}He which was present in the tritium target from β-decay of the tritium. As the primary (proton) energy was changed, was there any region where the separation of the recoil peak and the peak due to the T(p, n)^{3}He reaction was so poor as to make the technique useless?

J.L. LEROY: The positions of the peaks change with incident proton energy, but they were always sufficiently well separated. With decreasing proton energy the relative size of the recoil peak becomes larger.

W.P. POENITZ: Why did you measure fission cross-sections using the calibrated detector as a flux monitor instead of using your associated-particle apparatus to determine the flux directly?

J.L. LEROY: There are two reasons: (1) intensity; (2) the charged-particle counting apparatus required a large amount of material – such as the target assembly, the analysers, etc. – to be near the target. In cross-section measurements with thin-walled, relatively unshielded fission chambers, scattering from all this material would be troublesome. However, in the calibration experiment this scattering is not a problem because the collimator looks at the target only.

F. KÄPPELER: The experimental proton distribution in a gas-filled counter can be fitted very accurately in the upper energy range above the threshold. My feeling is that most difficulties arise at low proton energies where the adsorption of gas at the walls and other effects make it very difficult to describe the processes which are occurring and hence to make an accurate correction.

H. LISKIEN: Of course, we try to correct for adsorption on the housing of the detector. We have compared the proton-recoil proportional counter at 2.5 MeV with a counter telescope, at 1 MeV both with the telescope and with associated-particle counting, and at 500 and 250 keV with associated-particle counting. In all cases we have applied the same correction procedures, and the results of the measurements have agreed within estimated uncertainties. If we have underestimated some sources of error, the consequences have certainly not made our results useless.

III.. LIGHT-ELEMENT STANDARDS

A. The $^{6}Li(n,\alpha)T$ cross-section for fast neutrons

THE $^{6}Li(n,\alpha)$ CROSS-SECTION*

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Abstract

THE $^{6}Li(n,\alpha)$ CROSS-SECTION.

Measurements of the $^{6}Li(n,\alpha)$ cross-section were carried out in the 30- to 600-keV energy range using the Grey Neutron Detector. Of major interest was the ratio between the peak of the resonance and the minimum between 80 keV and 100 keV for which a value of 4.96 was obtained. The present $\sigma_{n,\alpha}$ values were combined with recent σ_{tot} measurements in an attempt to fit both cross-sections with one set of resonance parameters. Both the fitting results and the absolute cross-section values are preliminary and will be supplemented by additional work.

INTRODUCTION

The $^{6}Li(n,\alpha)$ cross-section is used as a reference cross-section for the low keV energy region with increasing frequency. The cross-section is sufficiently large and has a smooth energy dependence. Moreover, the utilization of this reference cross-section via lithium glass detectors provides for powerful neutron flux monitors in white neutron source flux measurements. However, the possible extension of the usable energy range to the higher keV energy range, as well as the accuracy of the cross-section in the 50-100 keV range are restricted by large uncertainties in the resonance energy range around 245 keV. Most notable are differences in the order of 20% for the peak value between recent measurements by Fort [1] and Uttley et al. [2].

The present measurements were carried out in the energy range from 90 to 600 keV with the majority of the measurements in the resonance region. The $\sigma_{n,\alpha}$ values were combined with recent σ_{tot} measurements in an attempt to fit both cross-sections with one set of parameters.

MEASUREMENTS

The schema of the set-up is shown in Fig.1. A collimated neutron beam passes through a lithium glass detector and is captured in a 'beam-catcher' type neutron detector. The $^{7}Li(p,n)$ reaction was used as a neutron source. Thin lithium metal targets yielding a resolution of 7 keV were used. Below 120 keV primary neutron energy, a second low-energy neutron group exists which is, however, small in the case of the present measurements.

The lithium glass detector was positioned at the end of a 1.88-m flight path. Besides passing through the 0.09-cm-thick lithium glass (NE 905), the neutron beam passes through two thin aluminium windows, thus neutron

* Work performed under the auspices of the US Atomic Energy Commission.

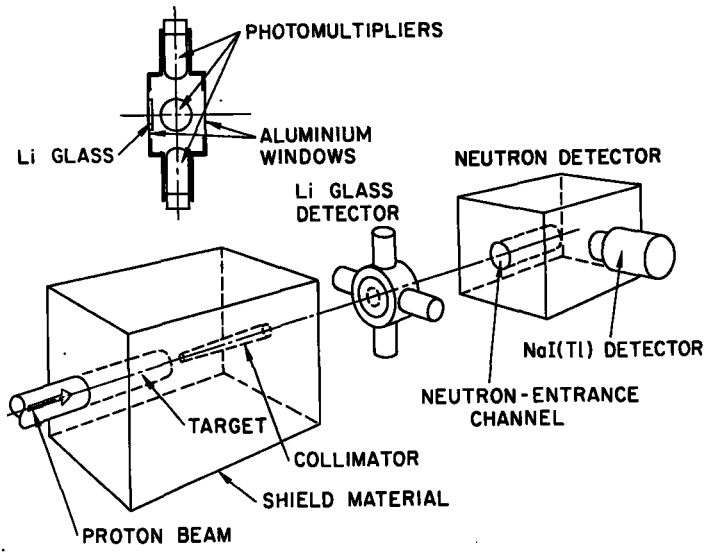

FIG. 1. Schematic arrangement of the target, collimator, shielding, ^{6}Li glass detector and neutron detector.

scattering in the detector is negligible except for scattering in the lithium glass. The lithium glass was chosen sufficiently thin to keep the correction for scattering in the lithium glass mostly below 5% and always below 10%. Four photomultipliers view the scintillation light in the lithium glass through air. A 'free' mounting of the lithium glass was chosen in order to best approximate the assumptions of the Monte-Carlo evaluations for the efficiency of the glass.

The Grey Neutron Detector [3] was used as a neutron flux monitor. Both water and vanadium-bath solution were used as moderators. Over the energy range of the present measurements the efficiency rises with increasing energy by about 3.5% for the water moderator and 1.5% for the vanadium bath. The Grey Neutron Detector should be extremely well suited for this energy range.

Measurements were carried out with a pulsed and bunched beam of about 1-2 ns duration. The time-of-flight spectra, the energy spectra of the lithium glass detector, and the energy spectra of the NaI(Tl) detector were recorded with an on-line computer system. The same input terminal was used for both signals, thus eliminating dead-time corrections.

The alpha count rate, C_α, is given by

$$C_\alpha = N_6 \cdot \sigma_{n,\alpha} d \cdot \Phi \cdot \frac{1 - \exp(-\Sigma_{tot}\, d)}{\Sigma_{tot}\, d} + C_\alpha^{Sc}$$

$$= N_6 \cdot \sigma_{n,\alpha} d \cdot \Phi \cdot k$$

where N_6 is the number of ^{6}Li atoms per cm^3, d is the thickness of the glass, Φ the neutron flux per cm^2, Σ_{tot} the macroscopic total cross-section

of the lithium glass and C_α^{Sc} the contribution to the alpha count rate from neutrons scattered in the glass. The correction factor

$$k = k_1 + k_2$$

with

$$k_1 = \frac{1 - \exp(-\Sigma_{tot} d)}{\Sigma_{tot}\, d}$$

and

$$k_2 = C_\alpha^{Sc} / \Sigma_\alpha d \Phi$$

consists of two parts, the first of which corrects for the attenuation of the primary neutron flux (loss) and the second of which corrects for the count-rate gain from scattered neutrons. The factor k_1 could be determined in a transmission experiment which yields directly $\exp(-\Sigma_{tot} d)$. This should be done for thicker glasses, but was not necessary for the present experiment where $\Sigma_{tot} d$ was usually smaller than 0.05. The factor k as obtained from a Monte-Carlo evaluation is shown in Fig.2. The figure suggests that the energy range of the oxygen resonance for which the correction is largest should be avoided. For the existing uncertainties in the input data the correction is undesirably large in this range.

The neutron count rate, C_n, is

$$C_n = \eta(E, E_0) \cdot \eta(E_0) \cdot \Phi$$

where $\eta(E, E_0)$ is the energy-dependent efficiency, normalized at E_0 to 1.0, and $\eta(E_0)$ is the absolute efficiency at this energy. $\eta(E, E_0)$ was considered extensively in recent reports [3]. $\eta(E_0)$ was obtained by comparing the activation of the vanadium bath from a 500-keV neutron beam with that of a ^{252}Cf source.

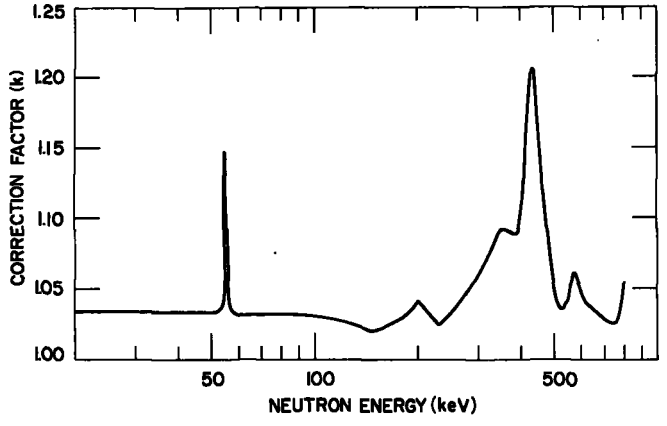

FIG. 2. The correction factor k versus neutron energy obtained by Monte-Carlo calculation.

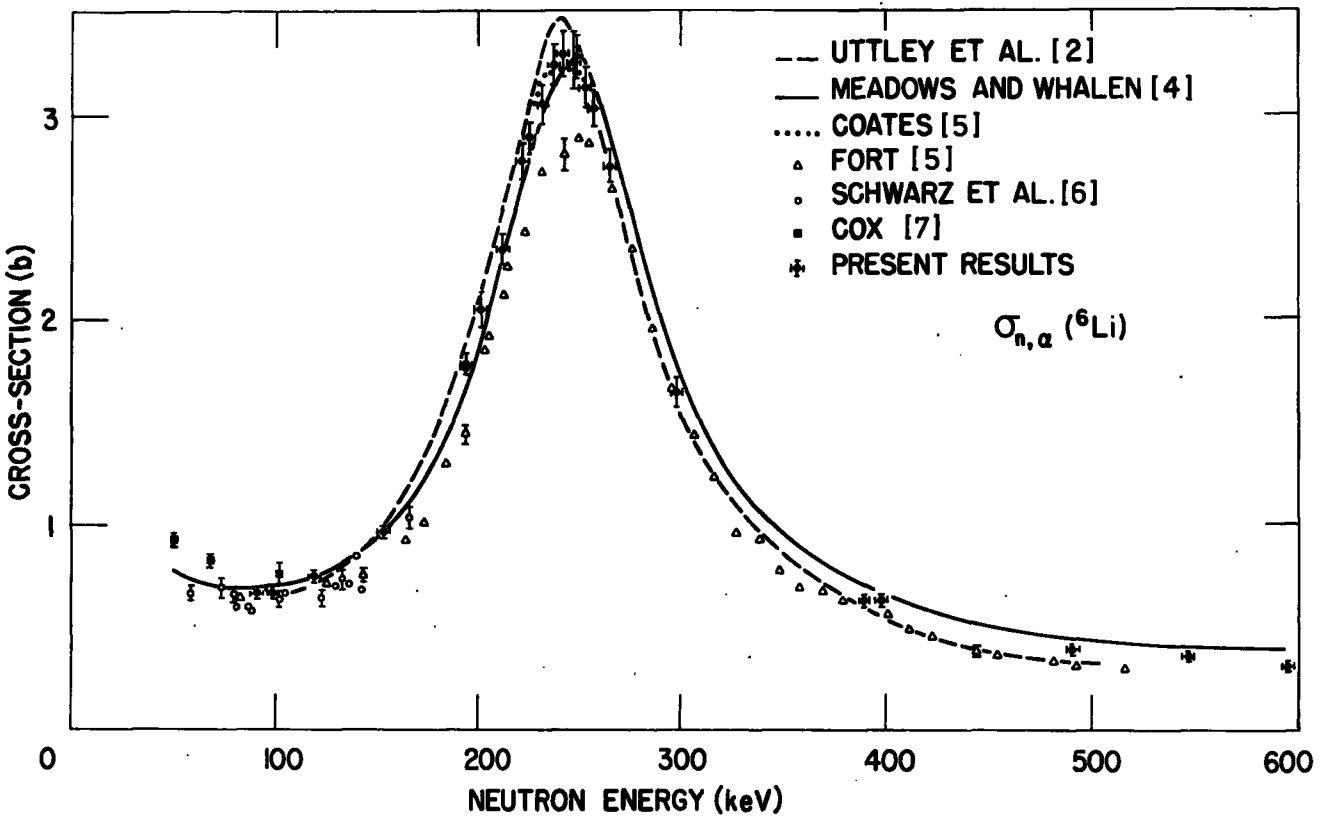

FIG. 3. Present measurements of the ^{6}Li(n, α)T cross-section compared with other experimental and derived values.

Corrections were made for the attenuation of the neutron beam in air and in the lithium glass, which again causes the largest correction at the oxygen resonance. Non-monoenergetic components in the neutron spectrum were well monitored with the time-of-flight method applied to the lithium glass detector. Corrections of usually less than 3% were applied. A major source for these neutrons is the in-scattering from the neutron source target backing.

The maximum neutron energy was determined from the magnetic field of an analysing magnet measured by a Hall probe. The calibration was made with the threshold of the $^{7}Li(p,n)$ reaction. The energy spread was determined once from the known target thickness and the spread due to bunching of the primary proton beam, and again from the spread of the time-of-flight spectra. Both values agreed within 1-2 keV. The energy uncertainty was estimated to be 3 keV.

RESULTS

The experimental values are shown in Figs 3 and 4. The energy uncertainty of 3 keV is shown but not the resolution of about 7 keV. The present values are compared in Figs 3 and 4 with other recent absolute measurements and values obtained from fits of the total cross-section and other data. At 100 keV, good agreement was obtained with recent measurements by Fort and Marquette [10], Condé et al. [8], and Uttley et al. [2].

At the resonance peak, the present values are somewhat lower than those by Uttley et al. [2] and higher than those by Fort and Marquette [10]. They agree best with the peak values obtained by Meadows and Whalen [4] and Coates et al. [5]. The energy of the resonance is slightly lower than that obtained by Meadows and Whalen [4], but not as low as that obtained by Uttley et al. [2] and Coates et al. [5].

The present values for $\sigma_{n,\alpha}$ were combined with previous measurements of the total cross-section [4]. A preliminary fit to the data was made by minimizing the function

$$E = \sum_{\alpha i} d_{\alpha i}^{2} W_{\alpha i}$$

where $d_{\alpha i}$ is the difference between the calculated and experimental cross-sections for reaction α at energy E_i, and $W_{\alpha i}$ is the weight. In this instance, where the number of total cross-section data points was much larger than the number of (n,α) points, the weights for each reaction were normalized, so that

$$\sum_{i} W_{\alpha i} = 1$$

The fit was made using the same basic assumptions as previously used in fitting the total cross-section only [4]:

(1) The sole contributor to the $5/2^{-}$ channel is potential scattering plus a single isolated resonance near 250 keV. The explicit equations are

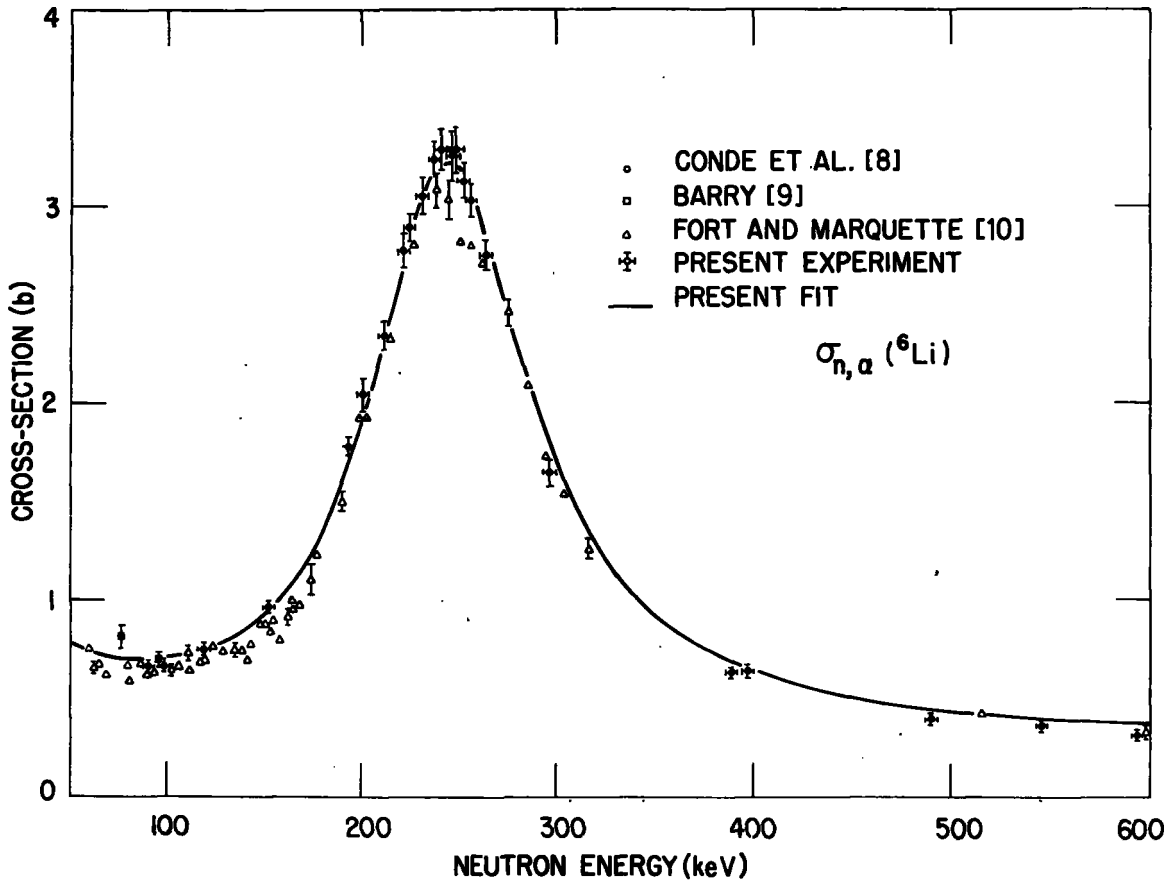

FIG. 4. Other experimental measurements of the ^{6}Li(n, α)T cross-section which are in good agreement with the present measurements and theoretical fit.

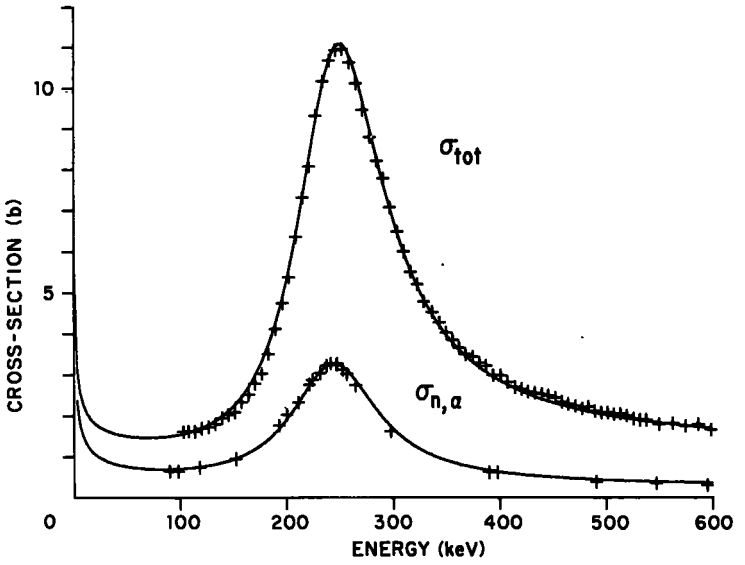

FIG. 5. Results of simultaneous fit of the present measurements of the $^{6}Li(n,\alpha)T$ cross-section and the experimental total cross-section data of Meadows and Whalen [4].

given in Section XII.1 of Ref. [11]. The variable parameters are the neutron channel radius, R_n, the reduced alpha and neutron width, γ_α^2 and γ_n^2, and the energy eigenvalue, E_λ. The alpha channel radius was fixed at 2.5 fermi for reasons given elsewhere [4].

(2) The reactions in the other channels are described by complex phase shifts of the form

$$\delta = \xi + i(a P_{\ell n}^2 + b P_{\ell n} P_{\ell' \alpha})$$

where $P_{\ell n}$ and $P_{\ell \alpha}$ are neutron and alpha-particle penetrabilities and have hard-sphere scattering. The parameters are a and b, and the channel radii R_n and R_α. However, these are fixed by other experimental data outside the range of this experiment such as the thermal (n,α) cross-section, the low-energy elastic scattering cross-section and the angular distribution of the tritons. The actual values are given in Ref. [4]. Figures 4 and 5 show the result from this fit.

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DISCUSSION

L. STEWART: The data of Meadows and Whalen, which you used in the fits of your data, apparently have an oxygen contamination above 400 keV which has not been subtracted. Perhaps this affects the consistency of your analysis of the total and (n,α) cross-sections.

W.P. POENITZ: We also have observed the oxygen resonance in our experimental data; it appears as a very small bump around 440 keV. The inconsistency between our (n,α) data and various total cross-section data exists over the whole 300- to 600-keV range, and I think the size of the oxygen cross-section is insufficient to account for it.

A.J. DERUYTTER: How was the ^{6}Li content of the glass determined?

W.P. POENITZ: Our Chemical Division has analysed three samples of the lithium glass, and the results of their chemical analyses for the total lithium content agree well with the analysis supplied by the manufacturer. Analysis of the isotopic composition is not yet complete so we have relied on the manufacturer's report.

A.J. DERUYTTER: What error do you put on a typical (n,α) cross-section point in the peak of the resonance?

W.P. POENITZ: The final error of course includes the question of the mass analysis. At present I have assigned something like 3% to 3.5% total error to the values which have been shown. On the side of the resonance, the effective error is much larger because of the uncertainty in the energy. I should mention that we have prepared an experiment to determine the energy of the resonance by a completely different technique. We plan to measure the total cross-section using a neutron spectrum which is white over a restricted energy range which covers the 250-keV resonance. We will shift the detectors in such a way that the gamma peak appears at exactly the position of the resonance in the transmission curve. The repetition frequency of the accelerator is then the only time-determining element besides the flight path so that we do not have to determine a time scale for a wide range of neutron energies in order to measure the absolute resonance energy.

A.J. DERUYTTER: With what white source do you intend to perform this experiment?

W.P. POENITZ: With our tandem-dynamitron. By white source I meant a source which gives a flat neutron spectrum over the restricted energy range from about 200 to 300 keV, just covering the resonance.

C.D. BOWMAN: Would you describe the pulse-height spectrum obtained from the ^{6}Li glass. I am interested in the effects of the coincidence requirement and of the bias settings.

W.P. POENITZ: The photomultiplier outputs are added in pairs and a coincidence is required between those pairs. The resolution is not so good as if the glass were in contact with a photomultiplier. We prefer to put the photomultipliers at the side in order to reduce the scattering problem.

We always recorded two-dimensional (pulse-height and flight-time) spectra, although we did not attempt to achieve very high time resolution. The gamma peak is not very great because of the coincidence requirement and because of the small size of the glass. We set a window over the time-of-flight peak and an equally wide window on an adjacent period. In each of these windows we obtain an energy spectrum. From one window we obtain a background spectrum, which is small and mostly due to scattered thermal neutrons.

C.D. BOWMAN: Did you make a correction for the small-amplitude, but genuine, (n,α) pulses which are lost because of the bias setting and the coincidence requirement?

W.P. POENITZ: We made an extrapolation which amounted to 1.2% - 1.5% of the total counts. In the range where the noise was such that the spectra could be compared with and without coincidence, the coincidence condition did not seem to change the shape of the spectra sufficiently to cause an error greater than 0.5% which was assigned to the extrapolation anyhow. I believe this uncertainty is small compared to the uncertainties in the flux determination and in the ^{6}Li content.

E. FORT: Would you explain a little more about the determination of the ^{6}Li concentration? Roughly speaking, are the results of your own chemical analysis higher or lower than the analysis supplied by the manufacturer?

W.P. POENITZ: The manufacturer's analysis did not specify a range of uncertainty. If one assumes that the uncertainty is roughly the range of the last decimal place which he gives, then the agreement between our analysis and his is within 2%. I do not recall the direction of the discrepancy.

M.S. COATES: In measurements with which I am familiar, where people have measured the concentration of lithium in glass, it seems that the manufacturers overestimate the amount of lithium in the glass sometimes by several per cent. I believe this was the case in a measurement with a thick glass at Gulf General Atomic, and I think Mr. Fort had the same experience.

E. FORT: Regarding the Monte-Carlo calculation of the correction for multiple scattering in the glass, I am surprised by the size of the peak due to caesium in view of the rather low caesium content of the glass. Otherwise, I think most of the divergences between your calculation of this correction and our calculation can be explained by differences in the data library used in the Monte-Carlo program.

W.P. POENITZ: I was surprised about the caesium peak myself. The data library is from Macklin, and I have not checked where he obtained the data. I do not consider the caesium peak to be very important for our results because it is due to a very sharp, narrow resonance, and one should simply avoid making measurements near the resonance energy.

I do not think that differences in the input data can explain why the silicon resonance does not show up in Mr. Fort's calculation; it appears in our correction factor and in the one calculated by Macklin.

E. FORT: We should compare our Monte-Carlo program.

J.L. LEROY: Are you sure that the composition of the glass is the same?

W.P. POENITZ: I think the compositions cannot account for the differences at this resonance. The composition and channel levels might vary a little but should not affect the general features of the resonance. These comments are even more true for the oxygen resonance.

MEASUREMENTS OF THE RELATIVE $^6Li(n, \alpha)$ CROSS-SECTION IN THE ENERGY RANGE 1 keV to > 500 keV

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Abstract

MEASUREMENTS OF THE RELATIVE $^6Li(n,\alpha)$ CROSS-SECTION IN THE ENERGY RANGE 1 keV to > 500 keV.

The $^6Li(n,\alpha)T$ cross-section has been measured between 1 keV and 500 keV using the neutron booster – 45-MeV linac system and the Harwell black detector as flux monitor. Data have been obtained for two 6.35-cm dia. ^6Li-loaded glasses, one 0.5 mm thick and the other 9.5 mm thick. The background was determined using the black-resonance technique, and corrections for multiple scattering were made using Monte-Carlo calculations. In contradiction to preliminary results, the cross-sections determined for the thick and thin glasses agree. The new results agree well with those obtained by Fort and Marquette if systematic energy shifts of approximately 5 keV are introduced. The peak cross-section value is about 10% lower than the peak value of the (n,α) cross-section derived by Uttley and Diment from their measurements of the ^6Li total cross-section.

Some preliminary measurements of the relative $^6Li(n, \alpha)$ cross-section using thin ^6Li glass scintillators have been reported earlier [1]. Since then, further data have been obtained with glasses of different thickness and, in addition, a more accurate determination has been made of the neutron flux spectrum used to derive the cross-section. The experiments were carried out on the 300-m flight path of the neutron booster – 45-MeV linac system at the Atomic Energy Research Establishment, Harwell. The ^6Li glass yields were obtained at a 120-m station and the neutron flux spectrum was measured with the Harwell black detector [1] at the 300-m station.

New data were obtained with two ^6Li-loaded glasses (Type GS20). One glass was 0.5 mm thick, 6.35 cm dia., and the other was 9.5 mm thick, 6.35 cm dia. The phototube was separated from the glass in each set of measurements by 5.25 cm to reduce the effects of multiple scattering of neutrons [1]. Collimators of boron-loaded wax (60 cm long) and lead (15 cm long) limited the neutron beam to 5 cm dia. at the detector position. The background, which was never greater than ~10%, was determined with filters of Al, SiO_2 and Mn using the black resonance technique. Discriminator bias conditions were such that no pulses from the $^6Li(n, \alpha)$ reaction were missed. Measurements were made also with ^7Li glasses (Type GS30) of the same dimensions as the two ^6Li glasses to see if there were any effects attributable to neutron capture in the constituents of the glass other than ^6Li. No measurable effect was observed.

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* On attachment from Imperial College, University of London.

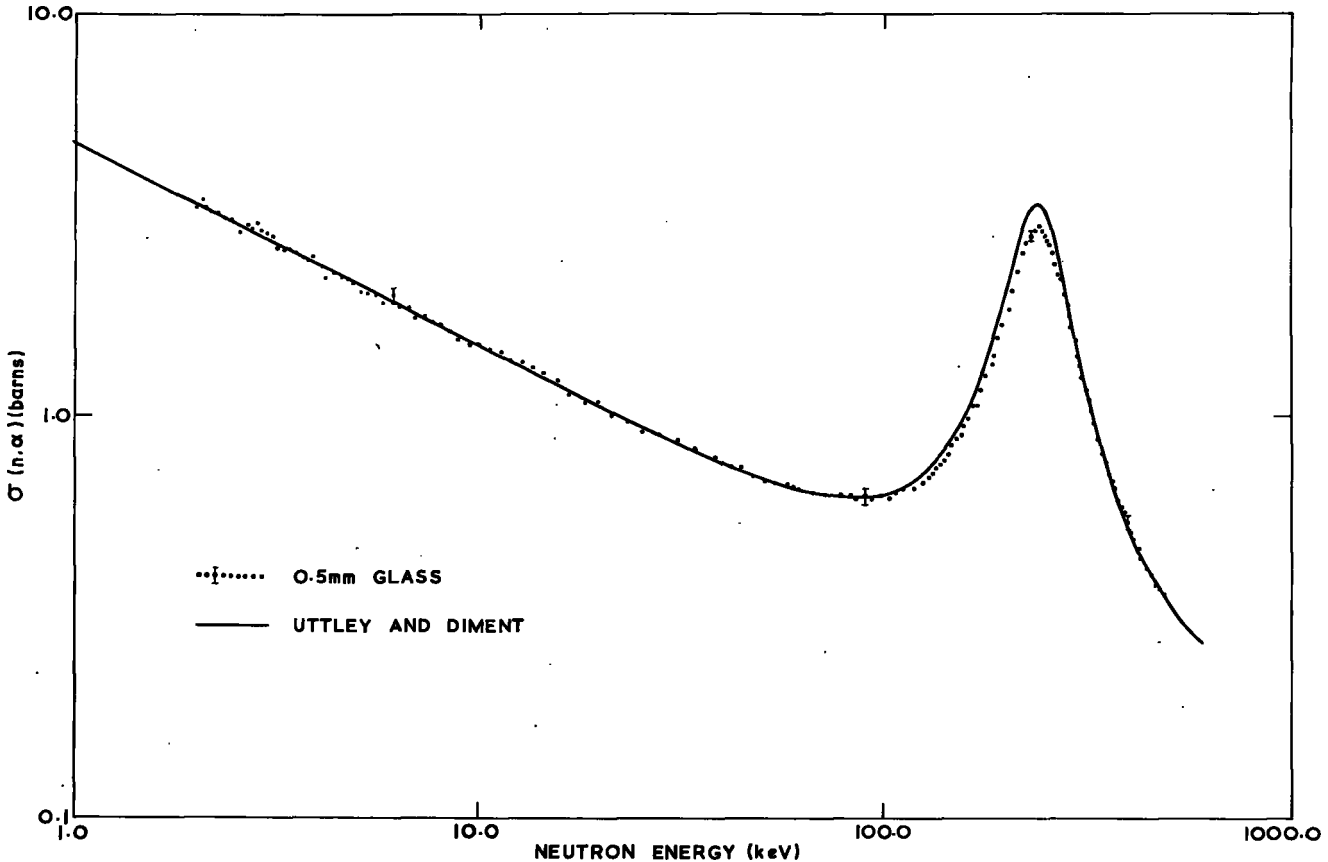

FIG. 1. Measurement of the $^{6}Li(n,\alpha)^{3}H$ cross-section.

TABLE I. THE $^{6}Li(n, \alpha)$ CROSS-SECTION OBTAINED WITH A 0.5-mm-THICK GLASS SCINTILLATOR

Neutron energy (keV)	$\sigma_{n\alpha}$	Neutron energy (keV)	$\sigma_{n\alpha}$	Neutron energy (keV)	$\sigma_{n\alpha}$	Neutron energy (keV)	$\sigma_{n\alpha}$	Neutron energy (keV)	$\sigma_{n\alpha}$
399.8	0.552	231.4	2.785	83.1	0.632	7.29	1.758	2.16	3.229
392.6	0.571	225.1	2.630	78.2	0.637	6.94	1.756	2.10	3.285
385.6	0.603	219.1	2.454	72.27	0.630	6.62	1.855	2.05	3.459
378.8	0.628	213.3	2.239	66.98	0.645	6.32	1.856	2.00	3.295
372.2	0.638	207.7	2.030	62.25	0.658	6.04	1.974	1.946	3.279
365.7	0.692	202.4	1.826	58.00	0.674	5.78	1.919	1.898	3.368
359.4	0.727	197.2	1.658	54.18	0.679	5.53	1.971	1.851	3.289
353.3	0.773	192.3	1.548	50.72	0.689	5.30	2.017	1.806	3.493
347.3	0.819	187.5	1.420	47.58	0.710	5.09	2.044	1.762	3.539
341.5	0.857	182.9	1.318	44.72	0.743	4.88	2.126	1.721	3.599
335.8	0.872	178.5	1.233	42.11	0.750	4.69	2.180	1.680	3.697
330.3	0.964	174.2	1.141	38.68	0.791	4.51	2.220	1.641	3.725
324.9	1.015	170.1	1.065	34.64	0.835	4.34	2.245	1.604	3.806
319.6	1.096	166.1	1.043	31.20	0.879	4.18	2.203	1.567	3.823
314.5	1.183	162.3	0.980	28.25	0.900	4.03	2.362	1.532	4.075
309.4	1.256	158.6	0.927	25.69	0.918	3.89	2.469	1.498	4.013
304.5	1.328	155.0	0.891	23.47	0.973	3.75	2.438	1.465	3.886
299.7	1.417	151.6	0.871	21.53	1.019	3.62	2.480	1.433	3.894
295.1	1.502	148.2	0.839	19.82	1.081	3.50	2.535	1.403	3.823
290.5	1.634	145.0	0.792	18.30	1.087	3.38	2.576	1.373	3.767
286.0	1.732	141.9	0.770	16.95	1.135	3.27	2.600	1.344	4.024
281.7	1.882	138.8	0.756	15.75	1.240	3.17	2.619	1.316	4.245
277.4	2.033	135.9	0.756	14.67	1.272	3.07	2.809	1.289	4.000
273.3	2.189	133.0	0.719	13.70	1.315	2.97	2.847	1.263	4.132
269.2	2.232	130.3	0.695	12.82	1.359	2.88	2.878	1.237	4.303
265.2	2.376	125.7	0.682	12.02	1.362	2.79	3.038	1.212	4.164
261.3	2.504	119.5	0.660	11.29	1.430	2.71	2.915	1.189	4.305
257.5	2.656	113.8	0.657	10.63	1.467	2.63	3.005	1.165	4.388
253.8	2.704	108.5	0.642	10.03	1.503	2.55	2.853	1.143	4.424
250.1	2.778	103.5	0.625	9.47	1.508	2.48	3.030	1.121	4.514
246.6	2.870	98.8	0.635	8.96	1.544	2.41	3.077	1.099	4.763
243.1	2.943	94.5	0.625	8.49	1.617	2.35	3.139	1.079	4.527
239.7	2.857	90.5	0.627	8.06	1.688	2.28	3.127	1.059	4.565
236.3	2.913	86.7	0.621	7.66	1.724	2.22	3.223	1.039	4.586

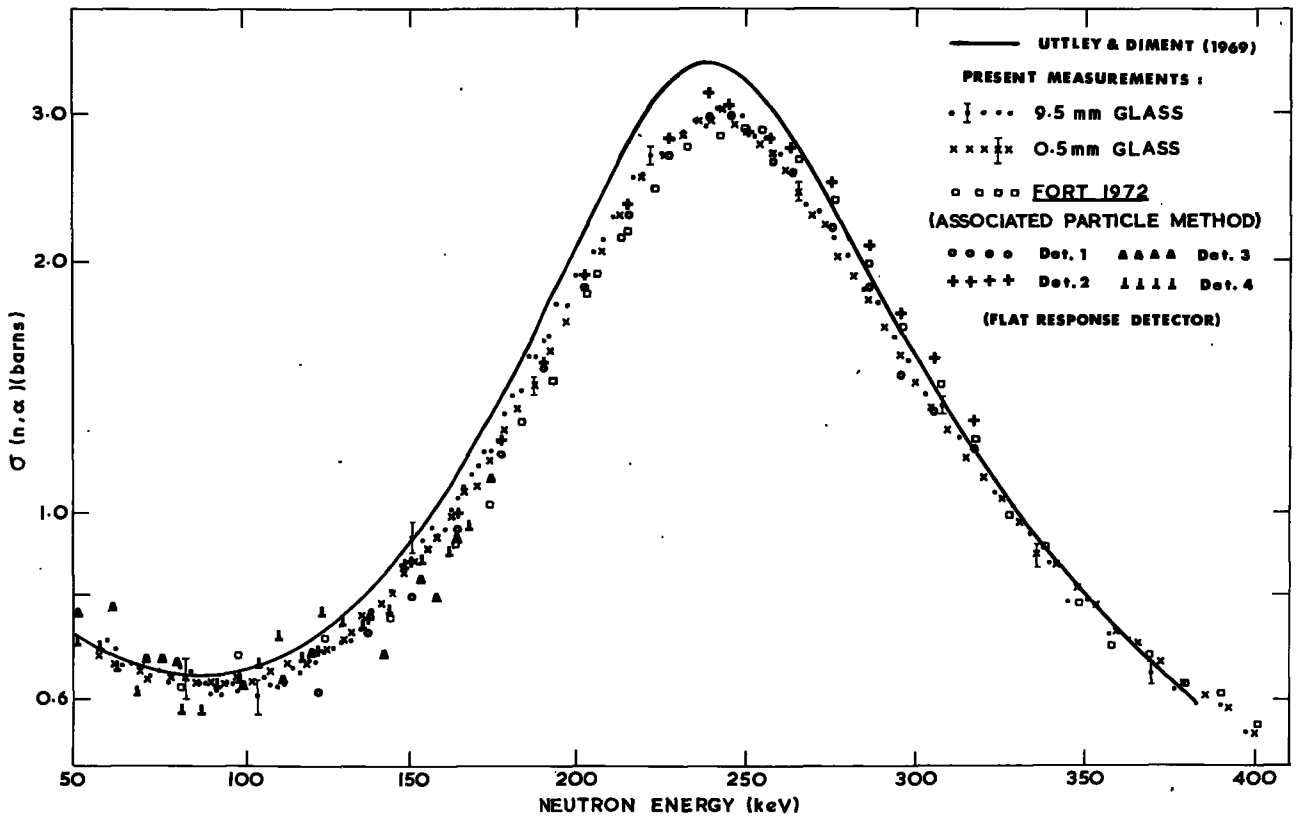

FIG. 2. Comparison of $^{6}Li(n, \alpha)$ cross-section determinations.

Corrections have been made for multiple scattering effects of neutrons in the ^{6}Li glasses using Monte-Carlo calculations made by Fort [2] (9.5-mm glass) and Macklin [3] (0.5-mm glass). The relative corrections between ~1.5 keV and 300 keV are 30% for the 9.5-mm glass and ~3% for the 0.5-mm glass. Above this energy, the correction factors increase because of the effect of the oxygen resonance at 440 keV, to a maximum value of ~1.7 for the thicker glass and ~1.1 for the thinner glass. Some of our earlier measurements, which have been published as a progress report [4], indicated that there was a significant difference between the cross-sections determined from the thin and thick glass measurements over the region of the resonance at ~250 keV. Further measurements proved that this result was incorrect owing to a count-rate-dependent instrumental fault in the timing equipment which invalidated the thick-glass data. Our present results show good agreement between the cross-sections determined from the thick- and thin-glass data. The analysis with the more accurately determined flux spectrum referred to above has resulted in a lower cross-section value over the resonance peak than reported in the preliminary measurement of Ref. [1]. The results for the 0.5-mm glass are shown in Fig. 1, normalized between 1.5 keV and 10 keV to the ^{6}Li(n, α) cross-section deduced by Uttley and Diment from total cross-section measurements [1]. The data are listed in Table I. The Uttley and Diment cross-section has been recommended as the most accurate one available in a recent evaluation [5]. There is a discrepancy of ~10% between the peak cross-section values.

Recently, Fort and Marquette [6] have published new absolute measurements of the ^{6}Li(n, α) cross-section over the resonance region using several glasses of different thickness. The experiments were made on a Van de Graaff accelerator at Cadarache using a flat-response detector of the super-long counter type for the flux determination. Our glass results agree well with these new data if an energy shift of ~5 keV is introduced between the two sets. The Van de Graaff data lie systematically higher in energy than the linac values. The results with error bars are shown in Fig. 2 without any energy normalization. At present, experiments are being carried out at Cadarache and Harwell to see if any systematic energy error can be traced.

The reason for the discrepancy between the latest glass data sets and the Uttley and Diment cross-section determination in the resonance region is not clear. Further ^{6}Li total cross-section measurements are clearly very desirable to test the validity of the basic data used by Uttley and Diment to calculate the (n, α) cross-section.

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DISCUSSION

M.S. COATES: I want to add some remarks on planned future work on the ^{6}Li cross-sections.

(1) We intend to make more total cross-section measurements to increase the accuracy of the Uttley and Diment data in the high-energy region and around the minimum near 100 keV where there could be systematic errors in their data. They used too thin a sample to get highly accurate data in these regions.

(2) Using the Van de Graaff, we intend to investigate the possibility of an (n, γ) contribution to the peak of the total cross-section. We do not expect any, of course.

(3) In a recent discussion, Mr. Uttley raised the possibility that the theoretical analysis is based on a false premise. The p-wave resonance at 250 keV has been analysed in terms of a contribution from a single level on top of an s-wave contribution. However, there is the possibility of interference between a just-bound level, which has previously been taken as an F-state, and the 250-keV level. On the basis of a rough calculation, such interference might qualitatively explain the fact that observed (n, α) cross-sections seem to be low compared with values derived from measurements of the total cross-section.

Previously at Harwell we had been considering differences in experimental techniques which might account for the discrepancies among the various (n, α) and total cross-section data. In the last few months the experimental situation has crystallized considerably. I think that our results are now in agreement with those presented by Mr. Fort and by Mr. Poenitz in these Proceedings if a normalization factor is allowed. Normalization may be permissible since the ^{6}Li contents of the various glasses have not been finally established. We might therefore conclude that all the experimental data from ^{6}Li glasses are in agreement within 3-4%.

This conclusion contradicts Uttley's analysis based on his measurements of the total cross-section. In his analysis, if the peak of the total cross-section is raised, then the (n, α) cross-section is lowered. To represent the experimental (n, α) data correctly would require a peak total cross-section of over 11.1 b. Uttley believes strongly in his peak value of 10.8 b, which agrees well with the value of Hibdon and Mooring apart from very small energy shifts, and thinks he could raise it at the most to 10.9 b unless some very serious error in his data were discovered. If one accepts the value of 10.8 b, then one is forced to conclude either that some other reaction is taking place or that the theoretical analysis is too simple minded.

I think one must accept the experimental evidence that the (n, α) and total cross-section data are discrepant according to the present analysis and must therefore consider carefully the possibility that the theoretical analysis is incorrect and that there is interference from another resonance. This is the opinion of both Uttley and myself.

R.W. PEELLE: J.A. Harvey at Oak Ridge National Laboratory is measuring the total cross-section. He has obtained a peak value of over 11.2 b. This number is preliminary and I mention it at this time only to suggest that there may possibly be total cross-section data which support the experimental (n, α) results.

A.J. DERUYTTER: Uttley and Diment derive from their measurements of the total cross-section the value of 940 ± 6 b for the (n, α) cross-section at thermal energy. How was it derived, and what is the meaning of the error?

M.S. COATES: Their measurements extend down to 70 eV where the total cross-section is essentially all (n, α). The thermal value was derived from a fit of the low-energy data which included a small correction for scattering.

The quoted error is due mostly to uncertainty in the ^{6}Li content of the enriched lithium metal samples.

EXPERIMENTAL METHODS USED AT CADARACHE TO DETERMINE THE $^{6}Li(n,\alpha)T$ CROSS-SECTION BETWEEN 20 keV AND 1700 keV

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Abstract

EXPERIMENTAL METHODS USED AT CADARACHE TO DETERMINE THE $^{6}Li(n,\alpha)T$ CROSS-SECTION BETWEEN 20 keV AND 1700 keV.

The $^{6}Li(n,\alpha)T$ cross-section has been measured absolutely in the energy range 20 keV to 1700 keV using ^{6}Li glass scintillators and the Cadarache Van de Graaff. The ^{6}Li content of the scintillators was determined from low-energy transmission measurements made with the 60-MeV Saclay linac and by comparison with other glass scintillators whose ^{6}Li content had been determined absolutely by the phase oscillation technique. The efficiencies of the glass scintillators were measured by two methods: (1) associated ^{3}He particles from the $T(p,n)^{3}He$ reaction were detected in coincidence with neutrons counted in the glass scintillators; (2) using time-of-flight techniques, the responses of the glass scintillators were compared with the response of a BF_3 counter previously calibrated by both the associated-particle and $MnSO_4$ bath techniques. The $^{6}Li(n,\alpha)T$ cross-sections derived from the two efficiency measurements agree within experimental error, except for an unexplained discrepancy of about 5% at the peak of the approximately 250-keV resonance.

The $^{6}Li(n,\alpha)T$ cross-section can be determined in two ways:

(1) Relative measurements can be normalized at an energy where the absolute cross-section is well known. At present, the cross-section is sufficiently well known only below 10 keV. High fluxes of neutrons extending to the energy range below 10 keV cannot be produced with electrostatic accelerators of the Van de Graaff type but can be conveniently produced with electron linacs.

(2) Absolute measurements can be made more conveniently with Van de Graaff accelerators.

The second method was used in the present experiments.

If the (n,α) cross-section is determined from the efficiency of ^{6}Li-loaded glass scintillators, an exact knowledge of the ^{6}Li content is necessary. The cross-section can be expressed as follows:

$$\sigma = \frac{\epsilon}{N \times F_C}$$

where ϵ = efficiency of the glass scintillator
N = ^{6}Li content in atoms/cm^2
F_C = all correction factors.

The efficiency of the ^{6}Li glass scintillator was measured with two different methods (Fig. 1). The first was an 'associated-particle' experiment

of the coincidence type, using the $T(p,n)^3He$ reaction [1, 2]. The scintillator efficiency is equal to the ratio of the number of coincidences, N_C, to the number of ^3He particles detected, $N_{^3He}$:

$$\epsilon = \frac{N_C}{N_{^3He}}$$

In the second method the glass scintillator was compared to a calibrated detector of known efficiency. The time-of-flight technique was used to distinguish direct neutrons from background neutrons. In this way we could explain and directly measure the effect of scattering in the target, which has been a source of uncertainty in measurements with a continuous neutron beam. Our reference detector was a flat-response directional BF_3 counter calibrated with the $MnSO_4$-bath technique [3] and an associated-particle technique [4].

The ^6Li content, N, was determined by two non-destructive methods.

The first method is a comparative method. The scintillator employed was compared to a 'secondary-standard' scintillator in a monoenergetic beam. The 'secondary-standard' scintillator was compared to a 'primary-standard' scintillator in a monoenergetic neutron beam. The ^6Li content of the 'primary-standard' scintillator was measured by the phase-oscillation method [5].

In the second method, the transmission of the glass scintillator is measured in the energy range between a few eV and about 100 eV. The transmission varies according to $a/\sqrt{E} + b$. As a function of $t = a/\sqrt{E}$, the transmission curve becomes a straight line whose slope determines the ^6Li concentration. Neutrons were produced with the 60-MeV Saclay linac [6].

In the 'associated-particle' experiment, the correction factors involve multiple scattering of neutrons inside the scintillator and multiple scattering of the ^3He particles inside the tritiated target. The spectrum of emission angles and the energy distribution depend on these two phenomena. These correction factors were calculated using a Monte-Carlo method and were particularly large in the energy range between 350 keV and 500 keV because of the resonance in ^{16}O at 425 keV. To prove that they were correctly calculated, the computations were compared with the experiment in three ways, with neutron scattering and ^3He scattering occurring separately and then simultaneously. The results were conclusive within the experimental errors.

The comparative experiment was repeated with glass scintillators of different thickness so as to verify again our calculation of neutron multiple scattering. By comparing theoretical calculations and experimental results, we were also able to show that the effect of scattering in the target was correctly calculated with the Monte-Carlo method [7].

The air-scattering corrections were calculated with a Monte-Carlo method and the usual transmission formula. Both calculations gave the same result.

The values of the $^6Li(n,\alpha)T$ cross-section obtained by the two experimental methods agree very well, except for the region at the peak of the resonance where an inexplicable discrepancy of about 5% occurs.

It must be noted that the two experimental techniques involve either different parasitic effects or identical effects with different magnitudes.

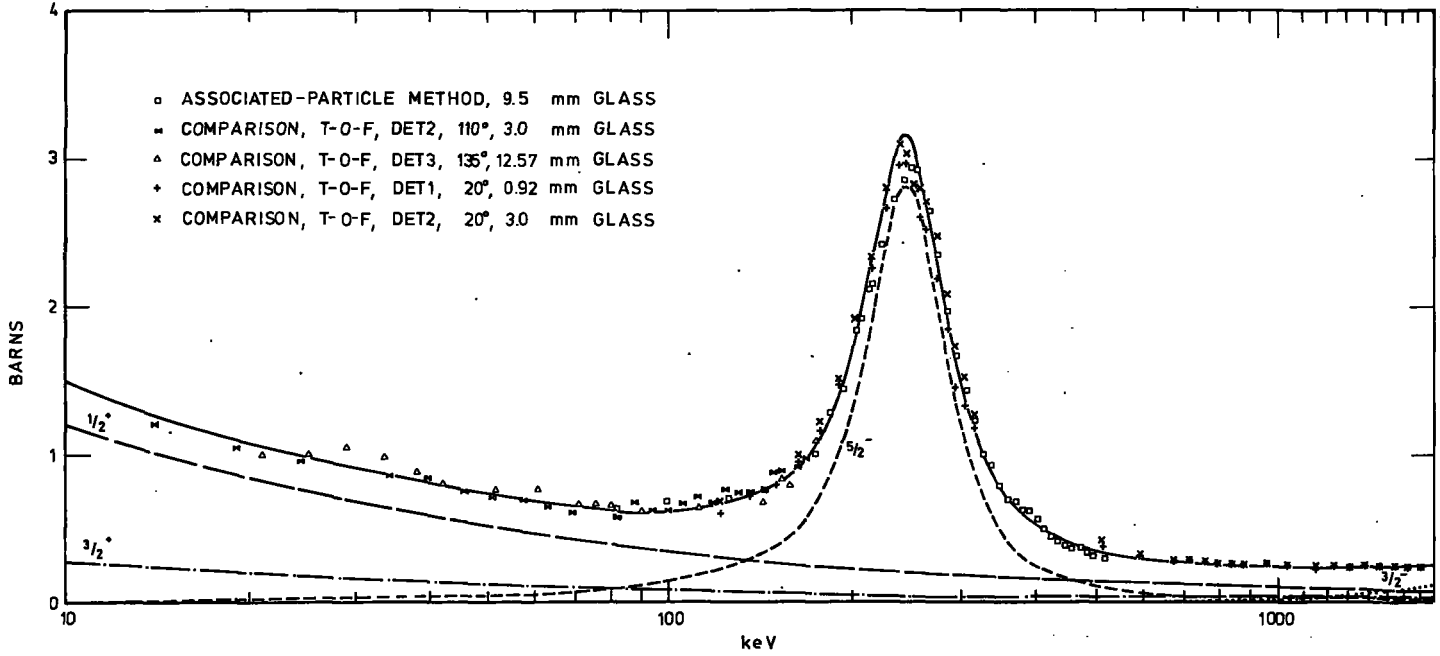

FIG. 1. $^{6}Li(n,\alpha)T$ *cross-section. The efficiencies of the* ^{6}Li *glass scintillators of different thicknesses were determined by the associated-particle method or by comparison with a calibrated detector of known efficiency.*

The experimental values are compared with previously published values by different authors as follows:

(a) In the region between 80 keV and 500 keV

There is good agreement between the absolute values of Condé et al. [8], the renormalized relative values of Schwartz et al. [9] and our values (see Fig. 1 in Fort's paper IAEA-PL-246-2/19 in these Proceedings). We consider this convergence as significant.

The discrepancy between the preliminary values of Coates et al. [10], given at the Argonne EANDC symposium, and our values is about 20% for the range of low-energy resonances. There is good agreement otherwise. The discrepancy decreases to about 7% to 8% if a relative shift of 5 keV is made [11].

Presently, there is no reason to renormalize our values, but a further check of the ^{6}Li glass content will be made in the near future.

(b) In the region between 500 keV and 1700 keV

Our absolute values obtained by a time-of-flight comparison are in agreement with the absolute values of Ribe [12]. They are consistent with the total cross-section values of Hibdon and Mooring [13], Meadows and Whalen [14] and Uttley and Diment [15] on the one hand, and with elastic cross-section values of Lane et al. [16] and Knitter and Coppola [17] on the other (see Fig. 2 in Fort's paper IAEA-PL-246-2/19 in these Proceedings).

We fitted our experimental results using an R-matrix formalism [18]. We used Le Rigoleur's program [19] with the one-level and two-open-channel approximation. Our reference values were as follows:

$\sigma_T(E)$: values of Uttley and Diment, and of Hibdon and Mooring: 70 eV < E < 1700 keV.

$\left(\frac{d\sigma}{d\Omega}\right)_{n,\alpha}(E)$: values of Deets et al., Robaye et al., Darlingtone et al. and Baudinet-Robinet et al.

$\left(\frac{d\sigma}{d\Omega}\right)_{n,n}(E)$: values of Lane et al.; 50 keV < E < 1700 keV

$\sigma_{n,n}(E)$: values of Lane et al; 500 keV < E < 1700 keV

$\sigma_{n,\alpha}$: 941.5 b at thermal energy

$\sigma_{n,n}$: 0.72 b at thermal energy

The $3/2^+$ level contribution to the 1/v law was found equal to 19%, and the Γ_α and Γ_n partial widths of the $5/2^-$ resonance were equal to 43 keV and 107 keV, respectively, giving a ratio $\Gamma_\alpha/\Gamma_n = 0.4$.

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DISCUSSION

A.J. DERUYTTER: In the second method (transmission measurement) which you used to determine the ^{6}Li content of your sample, some value of the total cross-section must be introduced. What value did you use?

E. FORT: We used the value 941.5 b at thermal energy.

M.S. COATES: What are the reasons for the discrepancy between your associated-particle results and the flat detector, time-of-flight results in the region around 300 keV?

E. FORT: Figure 1 of my paper shows only data obtained at Cadarache from all the various methods. Note that although many different thicknesses of glass were used, the overall agreement is quite good. I do not have an explanation for the disagreement between the associated-particle and comparative methods around 400 keV. Perhaps we should attempt to extend the energy range of each measurement in order to determine whether the disagreement is systematic or purely accidental.

There is also disagreement, of perhaps 5%, between the two methods at the resonance. I cannot explain the deviations; perhaps the correction for finite resolution of the neutrons was not correctly calculated in the associated-particle results.

R.W. PEELLE: Can you demonstrate that all neutrons which are produced in the tritiated target in association with ^{3}He-particles do indeed enter the ^{6}Li glass scintillator?

E. FORT: Yes, we can demonstrate this. The glass scintillator was precisely 5 cm away from the neutron-producing target so as to intercept all associated neutrons in the allowed range of emission angles. In fact, a criticism which might be brought against this method is that the neutron spectrum incident upon the glass is rather spread out. The spread of the neutron distribution is about 15 keV at half height but becomes very wide at the base, roughly 40 keV.

To prove that the glass really did intercept all neutrons we varied the distance between the scintillator and target; up to about 10 cm, the efficiency did not change. In addition, we measured the variation in the coincidence count rate as a function of angle to check for multiple scattering of ^{3}He in the target.

ANALYSIS OF EXPERIMENTAL METHODS AND PROPOSAL OF RECOMMENDED VALUES FOR THE $^{6}Li(n,\alpha)T$ REACTION BETWEEN 20 keV AND 1700 keV

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Abstract

ANALYSIS OF EXPERIMENTAL METHODS AND PROPOSAL OF RECOMMENDED VALUES FOR THE $^{6}Li(n,\alpha)T$ REACTION BETWEEN 20 keV AND 1700 keV.

Upper and lower bounds for the (n,α) and elastic scattering cross-sections of ^{6}Li have been calculated from the best known properties of the $^{6}Li+n$ system. These limits were obtained by keeping the total width constant but varying the ratio of the alpha and neutron partial widths. Measurements of the (n,α) and elastic cross-sections which fell outside these limits and measurements which were uncorrected for extraneous and multiple scattering effects were eliminated. The remaining experimental data appeared to converge and could be fitted with a mean deviation of 4% by an R-matrix formalism in the one-level and two-open-channel approximation. Recommended values and recommendations for further experimental work are given.

After a critical study of the many measurements and evaluations which have been made on the $^{6}Li(n,\alpha)T$ reaction, it can be concluded that the following cross-sections are well established:

(a) The total cross-section over the whole energy range [1].
(b) The (n,α) cross-section in the energy range where it varies according to $149.5/\sqrt{E} - 0.024$, which gives a value of 940 b at thermal energy [2]. This value agrees very well with the values given by all other investigators and evaluators within 0.5%.
(c) The elastic scattering cross-section from 500 keV up to 1700 keV.
(d) With reservations, the elastic scattering cross-section from thermal energy to about 70 keV. In this energy range, elastic scattering can be approximated by potential scattering. Thus, the cross-section is nearly constant and equal to about 0.7 b.

In addition, there are angular distribution measurements for (n,α) reaction and for elastic scattering which, although inaccurate (especially concerning the (n,α) process), are very useful because they can be used to define the respective contributions of $1/2^{+}$ and $3/2^{+}$ levels to the 1/v law relatively well [3].

In the studied energy range, only one resonance, a $5/2^{-}$ resonance, is known near 250 keV. Besides, the cross-section results from a reduced number of open channels ((n,α) and (n,n) channels; the (n,γ) channel is neglected). We therefore believe that the theoretical values can be proposed as recommended values on condition that they are calculated taking into account the spectroscopic structure of ^{7}Li and the known values noted above.

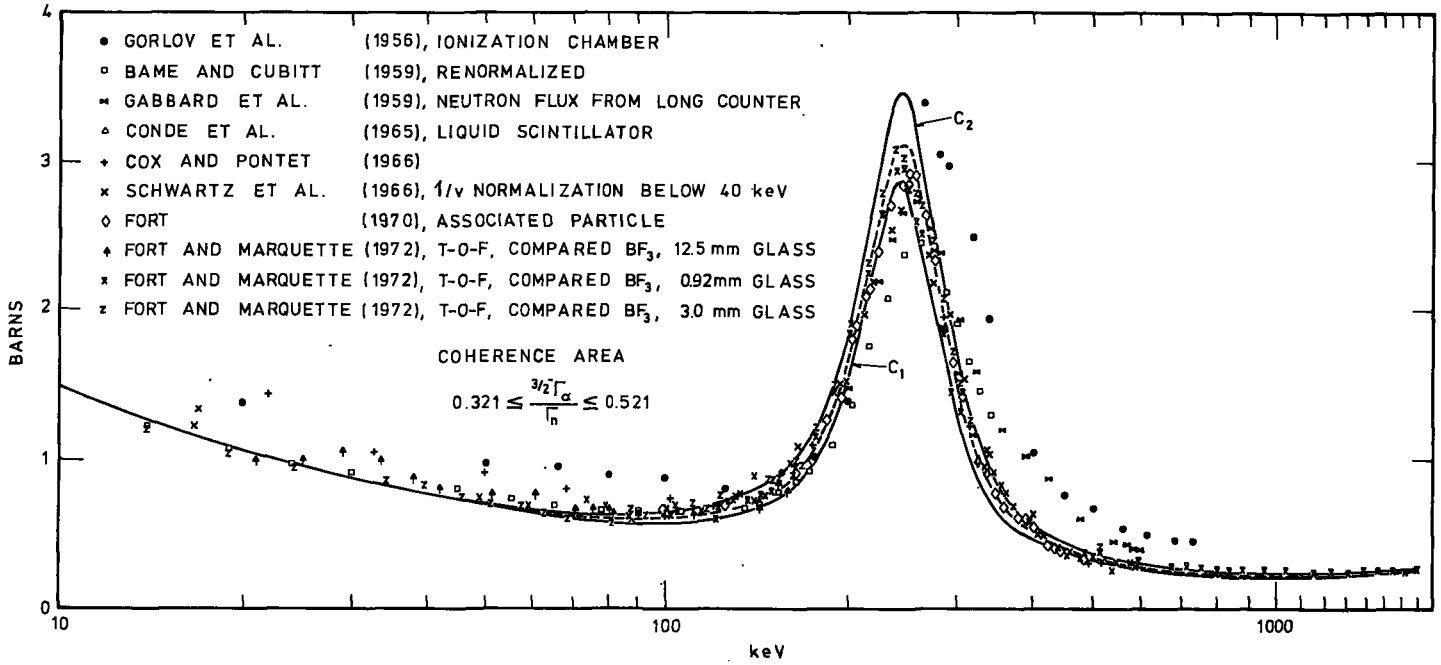

FIG. 1. ^{6}Li(n, α)T cross-section.

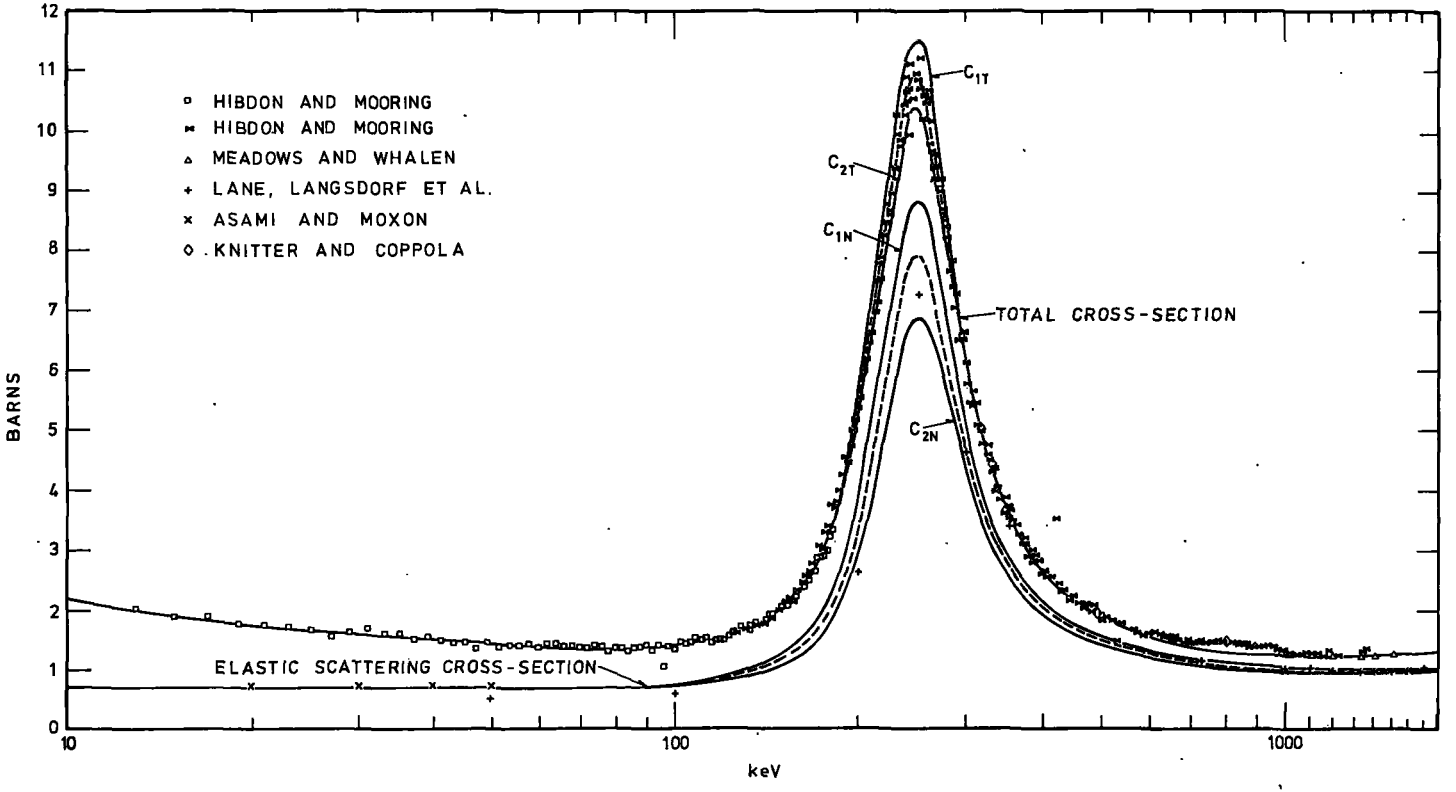

FIG. 2. Elastic scattering and total cross-section of ^{6}Li. The curves C_{1T} and C_{2T} bound the 'coherence area' for the total cross-section, and the curves C_{1N} and C_{2N} bound the 'coherence area' for the elastic scattering cross-section.

TABLE I. PARAMETERS USED IN THE R-MATRIX CALCULATION
Reaction channel: 2.98 fermi; elastic scattering channel: 4.32 fermi

	J^{π}_{λ}	s	ℓ	$\gamma^{2\lambda}_{\alpha s \ell, J}$ (MeV · fermi)	$\Phi_{\alpha s \ell, J}$ (rad)	$E^{\lambda}_{(C.M.)}$ (MeV)	$E^{\lambda}_{(Lab)}$ (MeV)	$E\,(^{7}Li^{*})$ (MeV)	Γ_n (MeV)	Γ_{α} (MeV)	Γ (MeV)
$^{6}Li+n$	$1/2^{+}$	1/2	0	3.250	0	2.547	2.973	9.80	6.300		
$\alpha+T$	$1/2^{+}$	1/2	0	0.147	0					0.946	7.246
$^{6}Li+n$	$3/2^{+}$	3/2	0	2.303	0	3.88	4.256	11.17	5.513		
$\alpha+T$	$3/2^{+}$	1/2	2	0.092	3.600					0.499	6.012
$^{6}Li+n$	$5/2^{-}$	3/2	1	2.570	0		0.252	7.48	0.107		
$\alpha+T$	$5/2^{-}$	1/2	3	0.0268	1.650					0.043	0.150
$^{6}Li+n$	$3/2^{-}$	1/2	1	1.390	0	1.92	2.240	9.17	0.970		
$\alpha+T$	$3/2^{-}$	1/2	1	0.058	2.600					0.321	1.291

TABLE II. THEORETICAL VALUES

Energy (keV)	(n, α) reaction			Elastic scattering		
	σ (b)	B_1 (mb/sr)	B_2 (mb/sr)	σ (b)	B_1 (mb/sr)	B_2 (mb/sr)
Thermal energy	941.5	0	0	0.71816	0	0
0.0001	470.68	0	0	0.71814	0	0
0.001	148.83	0	0	0.7180	0	0
0.01	47.047	0	0	0.7178	0	0
0.1	14.862	0	0	0.7169	0	0
1	4.688	0	0	0.7143	0	0
10	1.488	0.460	1.013	0.706	-1.658	0.0193
20	1.066	0.770	1.579	0.703	-3.418	0.0841
70	0.646	2.820	5.060	0.725	-14.378	1.677
80	0.629	3.438	6.106	0.739	-17.15	2.450
100	0.623	5.048	8.852	0.790	-23.548	4.877
123	0.664	7.857	1.703	0.903	-32.664	10.107
150	0.799	13.591	2.779	1.189	-46.60	23.146
162	0.909	17.633	30.980	1.412	-54.104	33.562
178	1.134	5.457	45.069	1.909	-65.037	55.644
200	1.676	43.425	77.986	3.189	-77.845	113.14
233	2.963	84.079	155.41	6.881	-44.509	278.52
242	3.145	89.336	166.68	7.693	-14.208	314.82
252	3.095	87.110	164.35	7.998	23.196	328.35
270	2.517	68.063	131.26	7.117	72.844	288.66
300	1.518	36.380	73.202	4.838	92.31	186.57
322	1.095	23.346	48.701	3.735	87.145	137.20
350	0.793	14.183	33.964	2.888	76.936	99.258
363	0.703	11.649	28.531	2.625	73.000	87.874
390	0.573	8.087	22.726	2.233	65.275	70.049
420	0.482	5.000	14.303	1.873	58.200	56.926
454	0.415	3.444	10.162	1.734	52.429	47.900
493	0.364	2.118	7.481	1.567	46.86	40.490
565	0.308	0.696	4.572	1.377	39.267	31.984
800	0.235	-1.132	0.761	1.117	25.13	20.02
1000	0.216	-1.912	-0.919	1.028	18.111	15.191
1200	0.213	-2.609	-2.573	0.940	13.104	11.971
1400	0.224	-3.357	-4.698	0.975	9.547	10.539
1600	0.250	5.671	-5.001	1.010	7.618	13.174
1700	0.268	6.033	-6.101	1.050	7.489	17.461

When the well-established values for the various cross-sections are taken into account, the calculated ^{6}Li(n, α)T cross-section cannot be represented by a unique curve but, considering the experimental errors, should be represented by a curve system limited by the two curves, C_1 and C_2. Each curve is characterized by the same value for the sum $\Gamma_\alpha + \Gamma_n$ (of the (n, α) and (n, n) partial widths of the $5/2^-$ resonance) and by a different value for the ratio Γ_α/Γ_n. The curves C_1 and C_2 determine a 'coherent area' (Fig.1). In the same way a 'coherent area' can be determined for the elastic scattering cross-section (Fig.2).

We made a critical analysis of experimental methods concerning the ^{6}Li(n, α)T reaction between 20 keV and 1700 keV. Values which were not corrected for neutrons scattered by the target backing (all measurements with a continuous beam) and for multiple scattering inside scintillators, and values which were inexplicably far from the 'coherent area' were neglected.

We are of the opinion that there exists a significant convergence between the absolute value of Condé et al. [4], the renormalized relative values of Schwartz et al. [5], the values of Ribe [6] and our own values [7, 8]. These values determine a 'convergence region'. At this point of our evaluation, we believe that we can determine theoretically (within a few per cent) the asymptotic values which the experimental results approach. Consequently, we choose the curve which goes through the mean point of the convergence region. Our recommended values are the theoretical values of the curve so defined. The associated accuracy is equal to the mean discrepancy between the converging experimental values and the theoretical ones. This accuracy approaches 4%.

Using Le Rigoleur's program [9], calculations in the R-matrix formalism were made in the one-level and two-open-channel approximation using the parameters given in Table I (the energy levels are those calculated by Barker [10]). Table II gives the recommended theoretical values which were obtained from the calculation.

Further experimental work is recommended as follows:

(1) More precise angular distributions for the (n, α) reaction are necessary.
(2) Elastic scattering angular distribution measurements between 100 keV and 500 keV are needed. They would help to discriminate between the values of Coates et al. [11] and ours.
(3) As suggested by Ribon [12], transmission measurements with a Van de Graaff and a linac on the same sample of an element having narrow and well-known resonances between 100 keV and 300 keV would help to determine exactly the energy of the $5/2^-$ resonance for which various investigators have reported values ranging from 247 keV to 268 keV.

New measurements of the ^{6}Li content in the glass scintillators used at Cadarache will be made in order to verify the necessity of renormalizing our results.

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DISCUSSION

E. FORT: The statement contained in the second recommendation at the end of my paper must be changed as a result of information presented at this panel. In the energy range 100 - 500 keV, the data presented by Mr. Coates and our own data are now basically in agreement within the quoted errors. We must now attempt to explain the disagreement with the data presented by Mr. Poenitz in this energy range.

W.P. POENITZ: I think there is no real discrepancy between your data and our data. The error bars still overlap. The absolute amplitude of my values has not yet been finally established; I may even consider normalizing to some average of experimental values around 100 keV.

Your curves marked C_1 and C_2 in Fig.1 seem to illustrate a problem which we experienced with theoretical fits around 100 keV. The calculated values were low compared with the experimental values in this energy range. One could try to adjust the calculation by changing the ratio of the contributions from the $1/2^+$ and $3/2^+$ states. To study this effect, we arbitrarily varied this ratio. In order to see what the maximum effect would be, we even allowed the contribution from one level to go all the way to zero while still requiring that the value of the thermal cross-section be reproduced correctly. The effect of this variation on the calculated cross-section around 100 keV was extremely small.

It seems to us that the calculated cross-section in the minimum around 100 keV can be effectively adjusted only by changing the widths of the contributing resonances. I noticed that Mr. Fort's theoretical curves also appear to lie on the low side of the experimental data around 100 keV.

E. FORT: I want to re-emphasize that the contributions from the various levels are not arbitrary, and this is what I have tried to illustrate with the curves marked C_1 and C_2 in Fig.1. This is due to the relatively well established relationship that the product of the partial width of the $3/2^+$ level and the corresponding partial width of the $5/2^-$ level is given by the coefficient B_1 from the Legendre polynomial fit of the elastic scattering cross-section in the energy range 100-500 keV where there can be no interference from other levels.

J.L. LEROY: I think that all the groups who have attempted to fit the ^{6}Li cross-sections over a wide energy range have experienced this difficulty. This is something to follow up in the coming months.

W.P. POENITZ: I think the problem is not limited to the fitting procedure, although it would be extremely helpful if the three laboratories (Harwell, Cadarache and Argonne) could agree on the formalism to be used

and on how results are to be quoted (widths in the laboratory versus centre-of-mass system, etc.). More importantly, I have the feeling that the total cross-section data may not be as well established as was previously thought. The peak value may be relatively well established, but in the energy range above 300 keV the fitting of the (n,α) cross-section depends strongly on the value of the total cross-section. I have found that in this energy range the newest total cross-section data of Uttley et al. and those of Meadows and myself differ by about 10%, which is a pretty large error for a total cross-section.

M.S. COATES: I would like to continue the comments of Mr. Poenitz:

(1) Uttley is willing to accept that his data at high energies are not very accurate because his sample was not sufficiently thick. We intend to make more total cross-section measurements both in the higher energy range and near the minimum around 100 keV in order to get more accurate data.

(2) I mentioned previously that Clements and Rickard have new data above 300 keV. Their measurements actually cover the range 160 keV - 3.9 MeV and were obtained using lithium sandwich detectors. The data have been normalized to the values of Uttley and Diment in the region 300-500 keV and are considerably lower than the data of Fort and Marquette.

It seems to me that at present the greatest experimental discrepancy in the $^{6}Li(n,\alpha)$ cross-section is in the region above 400 keV. It is almost a factor of 2.

W.P. POENITZ: If comparison is restricted to recent absolute measurements, the disagreement is not so great. Otherwise one must consider problems such as the ^{235}U fission cross-section. The old data of Gorlov et al. and of Gabbard et al. are very difficult to use.

M.S. COATES: The new data of Clements and Rickard are not absolute but are normalized. However, the flux measurement was with a flat-response detector over part of the energy range. At the highest energies the measurement was relative to the ^{238}U fission cross-section and is therefore perhaps not so accurate. Nevertheless there exists a big discrepancy with other values, and this must be explained.

III. LIGHT-ELEMENT STANDARDS

B. $^{10}B(n,\alpha)$ and $^{10}B(n,\alpha,\gamma)$ cross-sections for fast neutrons

PRELIMINARY MEASUREMENT OF THE RELATIVE $^{10}B(n, \alpha, \gamma)$ CROSS-SECTION

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Abstract

PRELIMINARY MEASUREMENT OF THE RELATIVE $^{10}B(n, \alpha, \gamma)$ CROSS-SECTION.

Recent measurements of the $^{10}B(n, \alpha, \gamma)$ cross-section on the 120-m flight path of the Harwell neutron-booster linac system have been re-analysed using a more accurate measurement of the neutron flux spectrum. The data are compared with a recent evaluation by Sowerby et al.

A measurement has been made of the relative $^{10}B(n, \alpha, \gamma)$ cross-section on the 120-m flight path of the neutron booster using the same neutron beam geometry as for the $^{6}Li(n, \alpha)$ cross-section measurements reported in paper IAEA-PL-246-2/17 in these Proceedings. The techniques of flux measurement and background determination are also those described there. The results have been published in a progress report [1]. Since then, the data have been re-analysed using a more accurate measurement of the neutron flux spectrum.

The sample was a $^{10}B_2O_3$ disc, 4.4 mm thick and 7.65 cm dia., containing 95% ^{10}B. Four NaI scintillation counters lying outside of the neutron beam detected the 478-keV gamma rays from the $^{10}B(n, \alpha, \gamma)^{7}Li$ reaction. Corrections for multiple scattering in the sample have been provided by Moxon [2]. Figure 1 shows the derived cross-section data, a numerical listing of which is given in Table I. The results are compared with the evaluation of the $^{10}B(n, \alpha)$ cross-section by Sowerby et al. [3], combined with the evaluated branching ratio of the $^{10}B(n, \alpha)$ reaction given by Gubernator and Moret [4]. The branching ratio is defined as the probability R for the reaction to go to the ground-state. The $^{10}B(n, \alpha, \gamma)$ cross-section is thus related to the $^{10}B(n, \alpha)$ cross-section by $\sigma(n, \alpha, \gamma) = \sigma(n, \alpha)\ [1 - R]$. The data are divided by (constant)/$\sqrt{E}$ so that a $1/v$ dependence of the cross-section is a horizontal straight line in the figure. The experimental points are fitted by eye to the curve derived from the evaluations between 1 and 2 keV. The results are slightly lower than the evaluated curve up to 100 keV, and above 100 keV there is no evidence for the resonance near to 150 keV as postulated by Sowerby et al. It should be noted that all data points which contribute to this feature depend on the scattering data of Mooring et al. [5]. It is intended to make another measurement of the $B(n, \alpha, \gamma)$ cross-section in this energy region using a ^{10}B metal sample.

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* On attachment from Imperial College, London University.

TABLE I. CROSS-SECTION $^{10}B(n,\alpha,\gamma)^{7}Li$ RELATIVE TO $19.30/\sqrt{E}$ b

Neutron energy (keV)	Ratio	Neutron energy (keV)	Ratio	Neutron energy (keV)	Ratio
1.03	0.911	9.00	0.879	58.6	0.880
1.08	0.921	9.51	0.889	62.9	0.873
1.13	0.932	10.1	0.891	67.8	0.883
1.18	0.928	10.7	0.899	73.2	0.891
1.25	0.918	11.3	0.899	79.2	0.883
1.31	0.910	12.1	0.892	86.1	0.913
1.38	0.915	12.9	0.871	93.9	0.904
1.46	0.920	13.8	0.898	100.0	0.911
1.54	0.940	14.8	0.891	105	0.906
1.64	0.935	15.8	0.885	110	0.908
1.74	0.924	17.1	0.879	116	0.895
1.84	0.910	18.4	0.883	121	0.908
1.96	0.909	19.9	0.880	128	0.903
2.09	0.916	21.2	0.874	135	0.881
2.24	0.925	22.1	0.879	142	0.912
2.04	0.913	23.1	0.882	150	0.909
2.58	0.910	24.1	0.876	159	0.904
2.78	0.927	25.3	0.890	168	0.893
3.01	0.932	26.5	0.875	179	0.861
3.26	0.916	27.8	0.875	190	0.873
3.54	0.920	29.1	0.877	203	0.852
3.86	0.915	30.6	0.898	217	0.843
4.24	0.907	32.2	0.893	232	0.815
4.67	0.906	33.9	0.919	243	0.801
5.16	0.891	35.8	0.905	250	0.790
5.74	0.883	37.9	0.902	258	0.771
6.43	0.883	40.1	0.894	266	0.734
6.97	0.877	42.5	0.893	274	0.734
7.31	0.889	45.2	0.894	282	0.744
7.68	0.897	48.1	0.881	291	0.731
8.09	0.901	51.2	0.883	300	0.693
8.52	0.870	54.7	0.886		

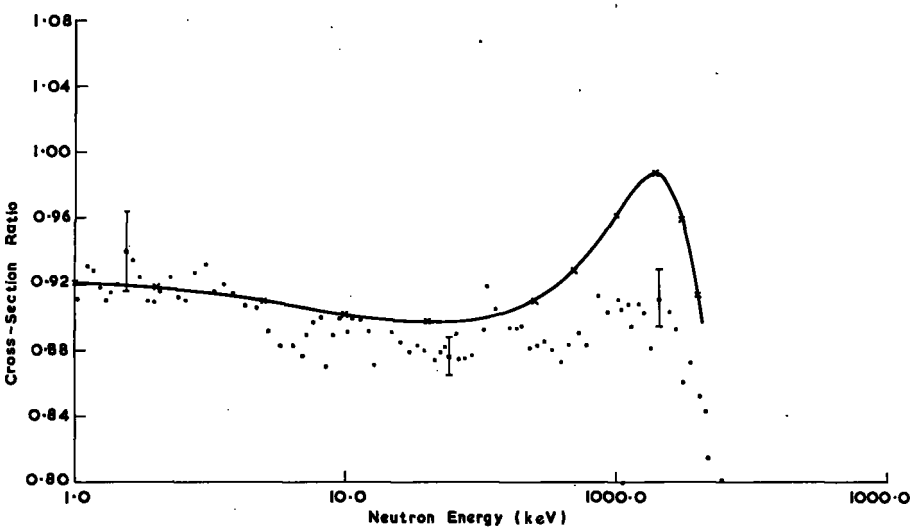

FIG. 1. $^{10}B(n,\alpha_1,\gamma)^7Li$ cross-section relative to $19.30/\sqrt{E}$ b.
• ┃ • • present data; *—* data by Sowerby et al. [3]; the evaluated branching ratio by Gubernator and Moret [4] is included.

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DISCUSSION

R.W. PEELLE: Are the experimental errors statistical or systematic?

M.S. COATES: They include a systematic error. The purely statistical error is better than 1% for the points shown. The systematic error which is included is due to determination of the background with the black-resonance technique. We have included a fairly generous error, I think.

C.D. BOWMAN: How large were the multiple scattering corrections in this experiment?

M.S. COATES: The combined multiple scattering and self-screening corrections were about 15%. We did not extend our measurements to higher energies because the sample was B_2O_3, and the presence of oxygen complicates the measurements. That is why we intend to use ^{10}B metal in our next experiments.

C.D. BOWMAN: Macklin and Gibbons used to claim that there was a resonance around 30 keV. How does this affect the 1/v behaviour of the cross-section?

M.S. COATES: My data may show a hint of a resonance around 35 keV, but the deviations from 1/v are barely outside the scatter of the points.

L. STEWART: My recollection is that Macklin and Gibbons thought that many people had failed to observe deviations from 1/v behaviour in the total $^{10}B(n,\alpha)$ cross-section because one channel drops below 1/v while the other channel rises above 1/v. The two partial cross-sections combine to give a total (n,α) cross-section which is approximately 1/v.

III. LIGHT-ELEMENT STANDARDS

C. The $^{3}He(n,p)T$ cross-section for fast neutrons

DIFFERENTIAL CROSS-SECTIONS FOR THE REACTIONS ^{3}He(n, p)T AND ^{3}He(n, d)D

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Abstract

DIFFERENTIAL CROSS-SECTIONS FOR THE REACTIONS ^{3}He(n, p)T AND ^{3}He(n, d)D.

Using the reciprocity theorem for nuclear reactions, differential cross-sections for the reactions ^{3}He(n, p)T and ^{3}He(n, d)D are deduced from the corresponding T(p, n)^{3}He and D(d, n)^{3}He data. At the same time, new best curves for the total ^{3}He(n, p)T and ^{3}He(n, d)D cross-sections are resulting from this conversion.

1. INTRODUCTION

Besides one measurement at 14.4 MeV [1, 2] no further experimental data about differential ^{3}He(n, p)T and ^{3}He(n, d)D cross-sections are available. Also, the CCDN compilation and evaluation of neutron cross-sections for ^{3}He [3] does not include differential data.

The differential data resulting from a re-evaluation [4] of the recent T(p, n)^{3}He and D(d, n)^{3}He cross-section evaluations [5, 6] were used in the present paper to carry out a conversion by the reciprocity theorem for nuclear reactions. At the same time, new best curves for the total ^{3}He(n, p)T and ^{3}He(n, d)D cross-sections are resulting from this conversion. They can claim the highest reliability because all available information about relative angular distributions, 0° differential and integrated cross-sections for both pairs of inverse reactions has been taken into account and had to be fitted simultaneously.

2. FORMALISM

For the nuclear reaction 2(1, 3)4, the cross-section will be indicated by $\sigma_{2(1,3)4}$. Correspondingly, the inverse reaction and cross-section are given by 4(3, 1)2 and $\sigma_{4(3,1)2}$. The theorem of reciprocity for nuclear reactions is based on the invariance against time inversion and leads to the simple relation between differential cross-sections [7]:

$$\frac{d\sigma_{2(1,3)4}}{d\Omega} \cdot k_{1,2}^2 \cdot g_{1,2} = \frac{d\sigma_{4(3,1)2}}{d\Omega} \cdot k_{3,4}^2 \cdot g_{3,4} \qquad (1)$$

where both sides of this equation have to be taken at centre-of-mass angles and energies which do correspond kinematically to each other.

Especially, the energies are related by

$$E_{1,2} = E_{3,4} + Q_{4(3,1)2} \qquad (2)$$

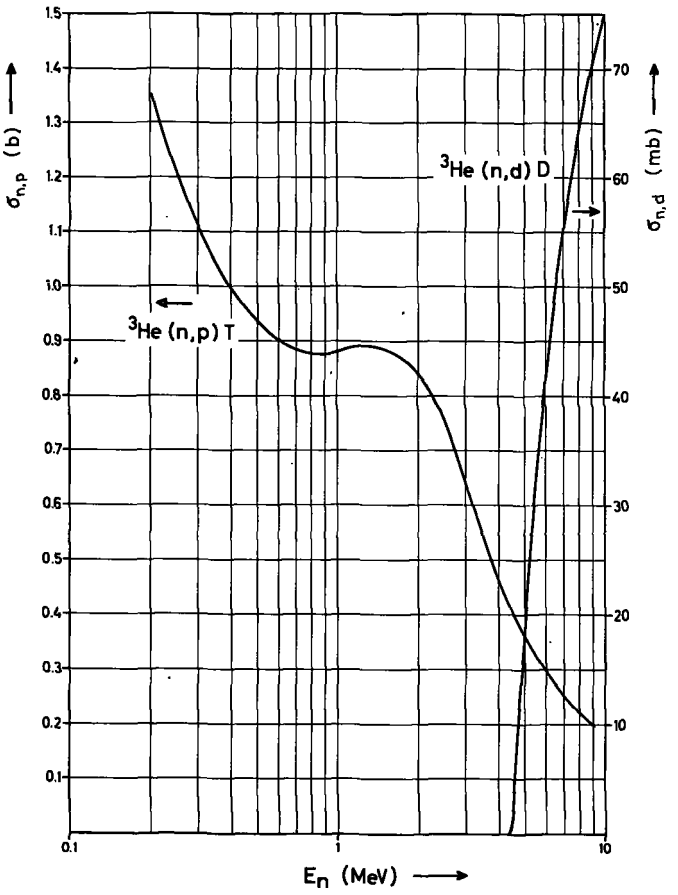

FIG. 1. The evaluated total cross-sections for the reactions ^{3}He(n, p)T and ^{3}He(n, d)D.

with $E_{i,j} = E_i M_j/(M_i + M_j)$ if the particle i with mass M_i and LAB energy E_i is hitting the particle j which has the mass M_j and is at rest.

$k_{i,j}$ and $g_{i,j}$ of Eq. (1) may be calculated according to

$$\hbar \cdot k_{i,j} = \frac{M_i \cdot M_j}{M_i + M_j} (2E_i/M_i)^{1/2} \tag{3}$$

$$g_{i,j} = (2I_i + 1)(2I_j + 1) \tag{4}$$

where I_i, I_j = spin of particles i, j.

For the ^{3}He(n, p)T reaction, one gets finally

$$\frac{d\sigma_{(n,p)}}{d\Omega}(E_n) = \frac{d\sigma_{(p,n)}}{d\Omega}(E_p)\frac{E_p}{E_n} \tag{5}$$

with $E_n = E_p - 1.019$ MeV.

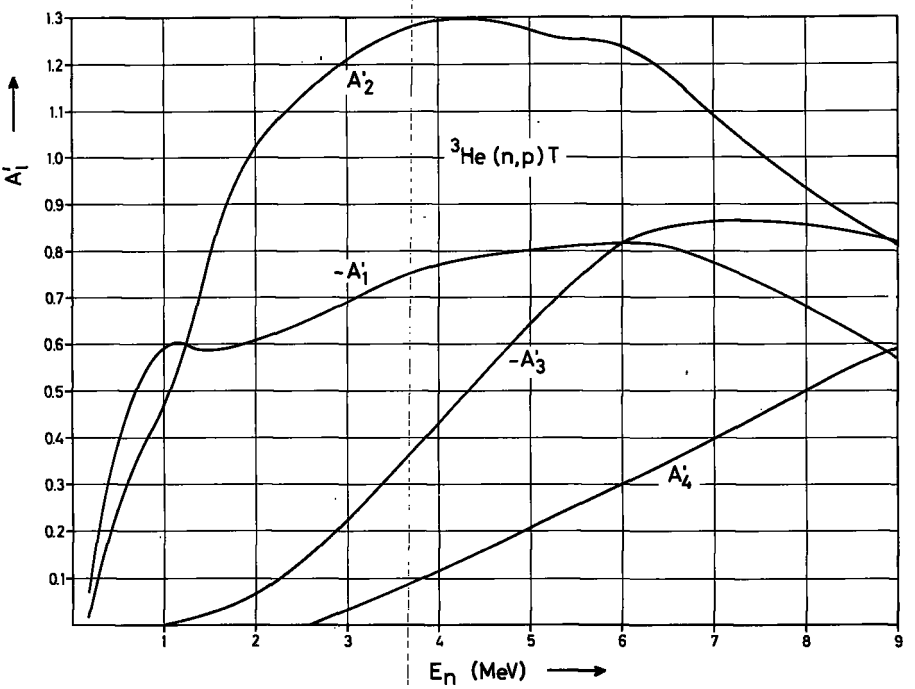

FIG.2. The normalized ($A'_0 = 1$) Legendre coefficients for the reaction $^3He(n,p)T$.

Integration over all solid angles leads to

$$\sigma_{(n,p)}\ (E_n) = \sigma_{(p,n)}\ (E_p)\frac{E_p}{E_n} \tag{6}$$

The corresponding results for the $^3He(n,d)D$ reaction are:

$$\frac{d\sigma_{(n,d)}}{d\Omega}\ (E_n) = \frac{d\sigma_{(d,n)}}{d\Omega}\ (E_d)\ \frac{2E_d}{E_n} \tag{7}$$

with $E_n = 0.668 \cdot E_d + 4.361$ MeV. Here, after integration, the right-hand side has to be divided by two because of the symmetry of the d-d reaction:

$$\sigma_{(n,d)}\ (E_n) = \sigma_{(d,n)}\ (E_d)\ \frac{E_d}{E_n} \tag{8}$$

The data to be transformed [5, 6] have been given as:

$$\frac{d\sigma}{d\Omega}(\theta) = \frac{d\sigma}{d\Omega}\ (0°) \sum_1 A_l P_l (\cos\theta), \quad \sum_1 A_l = 1 \tag{9}$$

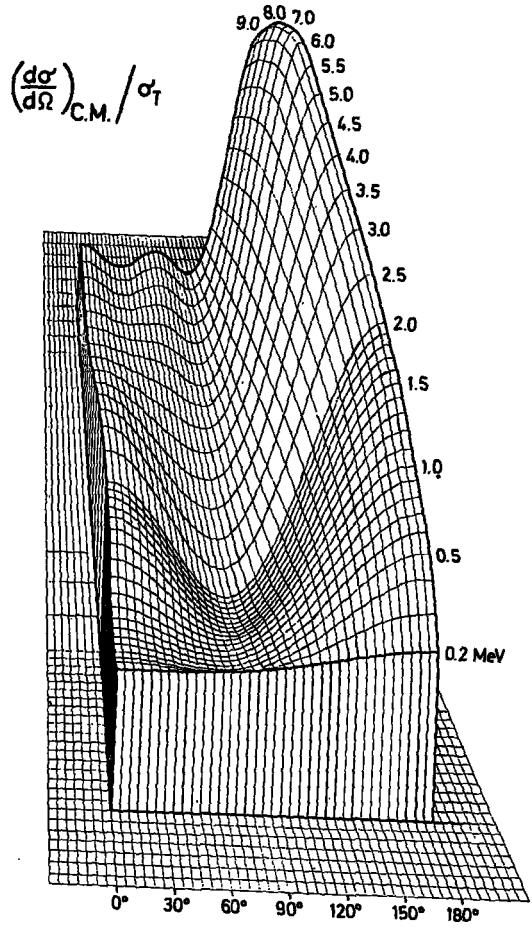

FIG. 3. The relative centre-of-mass anisotropy for the reaction $^3He(n,p)T$.

and

$$\sigma = 4\pi\, A_0 \frac{d\sigma}{d\Omega}(0°) \tag{10}$$

The conversion is performed by transforming σ and the energy using Eqs (5) to (8), while the A_1 coefficients remain unchanged. Differential cross-sections are given by

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} \sum_1 A'_1 P_1(\cos\theta), \qquad A'_0 = 1 \tag{11}$$

with $A'_1 = A_1/A_0$.

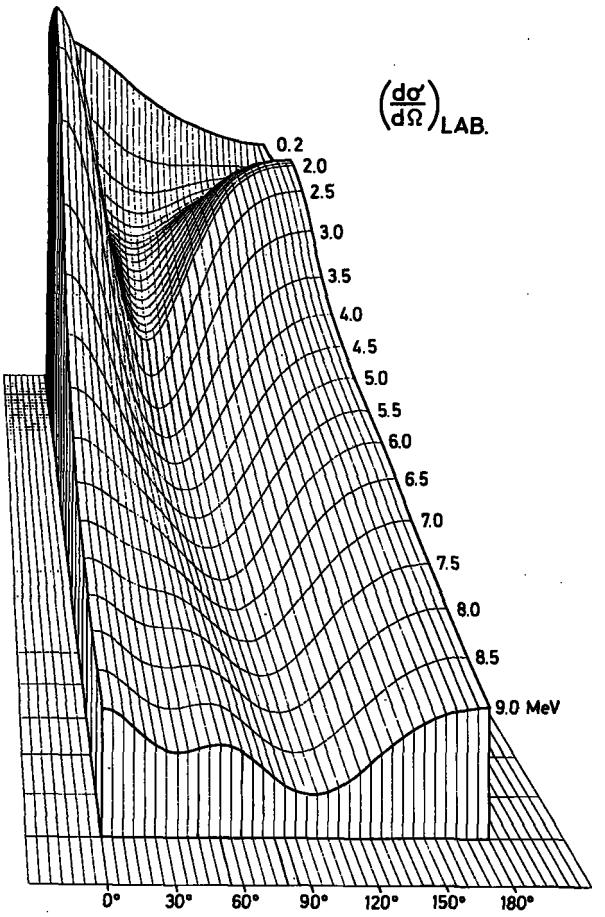

FIG. 4. The differential LAB cross-sections for the reaction $^3He(n, p)T$.

3. RESULTS

The tabulated pairs of energy and cross-section values for the inverse reactions were transformed following the equations of section 2. These transformed data were plotted, and from the smoothed curve the readings were taken and tabulated. A similar procedure was followed for the A_1' coefficients.

3.1. The reaction $^3He(n, p)T$

The plot of the total cross-section for the $^3He(n, p)T$ reaction as a function of the neutron energy is given in Fig. 1. The curve starts at 0.2 MeV because the evaluation of the $T(p, n)^3He$ cross-section [6] excludes the proton energy region between forward (1.019 MeV) and backward

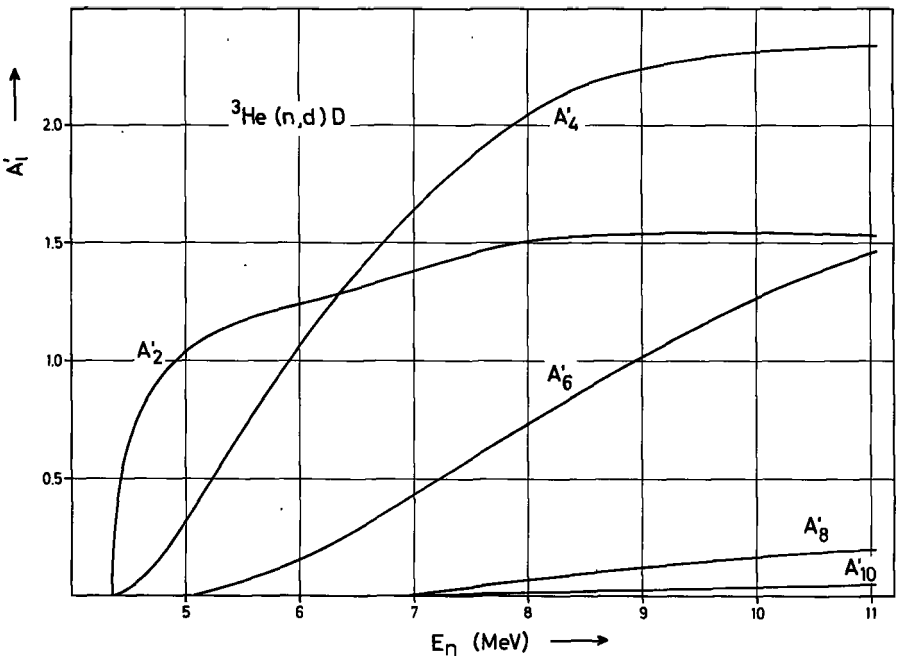

FIG. 5. The normalized ($A'_0 = 1$) Legendre coefficients for the reaction ^{3}He(n, d)D.

(1.148 MeV) threshold owing to lack of reliable differential data. But as far as this cross-section is concerned, data for the region from thermal energy to 0.2 MeV can be found in the CCDN evaluation of ^{3}He neutron cross-sections [3] which fit exactly to the curve presented here. Further on, the ^{3}He(n, p)T cross-section curve in Fig.1 stops at 9 MeV corresponding to the high-energy end of the T(p, n)^{3}He evaluation (10 MeV).

The plot of the A'_l coefficients in the same energy region is shown in Fig.2. Here, really no data are available between thermal energy and 0.2 MeV. But Fig.2 suggests that A'_1 and A'_2 continue to smoothly approach zero at thermal energy. How far centre-of-mass isotropy is already reached at 0.2 MeV neutron energy may also be seen from the three-dimensional plot in Fig.3.

The readings from Figs 1 and 2 are listed in Table I. A comparison of the total cross-section with the results of other evaluations is made in Table II. For practical use, Table III lists differential ^{3}He(n, p)T cross-sections in the LAB system calculated in steps of 5° for the same energy values as quoted in Table I. A survey about these data is presented in Fig.4. These three-dimensional plots are produced with the help of the computer program TRICE [8].

3.2. The reaction ^{3}He(n, d)D

Figure 1 also shows the plot of the cross-section curve for the ^{3}He(n, d)D reaction as a function of the neutron energy. The cross-section

TABLE I. EVALUATED $^3He(n, p)T$ CROSS-SECTIONS AND NORMALIZED LEGENDRE COEFFICIENTS A'_l

E_n (MeV)	$\sigma_{n,p}$ (b)	A'_1	A'_2	A'_3	A'_4
0.2	1.350	-0.095	0.032		
0.3	1.104	-0.215	0.112		
0.4	0.992	-0.308	0.183		
0.5	0.932	-0.386	0.246		
0.6	0.892	-0.450	0.302		
0.7	0.883	-0.501	0.352		
0.8	0.876	-0.540	0.393		
0.9	0.876	-0.572	0.428		
1.0	0.882	-0.590	0.467	-0.002	
1.1	0.887	-0.600	0.516	-0.005	
1.2	0.890	-0.602	0.575	-0.008	
1.3	0.891	-0.592	0.643	-0.013	
1.4	0.888	-0.587	0.718	-0.018	
1.5	0.884	-0.586	0.794	-0.024	
1.6	0.877	-0.587	0.810	-0.031	
1.7	0.870	-0.590	0.914	-0.038	
1.8	0.862	-0.595	0.959	-0.046	
1.9	0.850	-0.600	0.995	-0.056	
2.0	0.838	-0.606	1.024	-0.067	
2.5	0.756	-0.644	1.131	-0.137	
3.0	0.642	-0.690	1.210	-0.226	0.033
3.5	0.534	-0.737	1.265	-0.325	0.074
4.0	0.459	-0.769	1.294	-0.432	0.116
4.5	0.401	-0.789	1.293	-0.540	0.162
5.0	0.359	-0.801	1.271	-0.644	0.208
5.5	0.325	-0.811	1.253	-0.736	0.254
6.0	0.297	-0.817	1.235	-0.819	0.300
6.5	0.274	-0.808	1.178	-0.850	0.348
7.0	0.255	-0.777	1.088	-0.860	0.396
7.5	0.236	-0.729	1.008	-0.860	0.447
8.0	0.220	-0.680	0.934	-0.852	0.500
8.5	0.208	-0.626	0.867	-0.837	0.549
9.0	0.198	-0.568	0.811	-0.819	0.590

TABLE II. COMPARISON OF THE EVALUATED $^{3}He(n,p)T$ TOTAL CROSS-SECTION WITH THE RESULTS OF OTHER EVALUATIONS

E_n (MeV)	$\sigma_{n,p}$ (b)								
	This evaluation	ENDF/B-III MAT 1146	Dev.[a] (%)	UKNDL DFN 220	Dev.[a] (%)	CCDN-NW/6 Ref. A167	Dev.[a] (%)	KEDAK	Dev.[a] (%)
0.2	1.35	1.32	-2.2	1.32	-2.2	1.35	0.0	1.275	- 5.6
0.5	0.932	0.930	-0.2	0.930	-0.2	0.91	-2.4	0.830	-10.9
1.0	0.882	0.879	-0.3	0.879	-0.3	0.89	+0.9	0.796	- 9.9
2.0	0.838	0.825	-1.6	0.825	-1.6	0.84	+0.2	0.888	+ 5.6
5.0	0.359	0.372	+3.6	0.372	+3.6	0.37	+3.1	0.355	- 1.1
9.0	0.198	0.196	-1.0	0.202	+2.0	0.20	+1.0	0.172	-13.1

[a]Deviation from the results of this evaluation.

TABLE III. DIFFERENTIAL $^{3}He(n, p)T$ CROSS-SECTIONS IN THE LAB SYSTEM

Energy (MeV)	0.200	0.300	0.400	0.500	0.600	0.700	0.800	0.900
	Centre-of-mass input values for the reaction $^{3}He(n, p)T$							
σ_T (mb)	1350.000	1104.000	992.000	932.000	892.000	883.000	876.000	876.000
A' 0	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
A' 1	-0.095	-0.215	-0.308	-0.386	-0.450	-0.501	-0.540	-0.572
A' 2	0.032	0.112	0.183	0.246	0.302	0.352	0.393	0.428
Angle (degree)	Differential cross-sections (mb/sr) in the laboratory system							
0.0	0.129E 03	0.105E 03	0.957E 02	0.906E 02	0.876E 02	0.880E 02	0.887E 02	0.901E 02
5.0	0.129E 03	0.105E 03	0.955E 02	0.902E 02	0.872E 02	0.876E 02	0.882E 02	0.895E 02
10.0	0.129E 03	0.105E 03	0.947E 02	0.893E 02	0.861E 02	0.862E 02	0.867E 02	0.879E 02
15.0	0.128E 03	0.104E 03	0.935E 02	0.878E 02	0.843E 02	0.842E 02	0.844E 02	0.853E 02
20.0	0.127E 03	0.103E 03	0.919E 02	0.859E 02	0.820E 02	0.815E 02	0.813E 02	0.819E 02
25.0	0.126E 03	0.101E 03	0.901E 02	0.836E 02	0.794E 02	0.784E 02	0.778E 02	0.779E 02
30.0	0.125E 03	0.100E 03	0.880E 02	0.811E 02	0.765E 02	0.750E 02	0.740E 02	0.738E 02
35.0	0.124E 03	0.982E 02	0.859E 02	0.786E 02	0.736E 02	0.716E 02	0.703E 02	0.696E 02
40.0	0.122E 03	0.964E 02	0.837E 02	0.761E 02	0.708E 02	0.685E 02	0.668E 02	0.658E 02
45.0	0.121E 03	0.947E 02	0.817E 02	0.739E 02	0.683E 02	0.657E 02	0.637E 02	0.625E 02
50.0	0.119E 03	0.930E 02	0.799E 02	0.720E 02	0.663E 02	0.634E 02	0.613E 02	0.598E 02
55.0	0.117E 03	0.915E 02	0.784E 02	0.704E 02	0.647E 02	0.618E 02	0.595E 02	0.580E 02
60.0	0.116E 03	0.901E 02	0.771E 02	0.693E 02	0.636E 02	0.607E 02	0.585E 02	0.570E 02
65.0	0.114E 03	0.888E 02	0.762E 02	0.686E 02	0.631E 02	0.603E 02	0.582E 02	0.568E 02
70.0	0.112E 03	0.878E 02	0.755E 02	0.683E 02	0.631E 02	0.605E 02	0.585E 02	0.573E 02
75.0	0.111E 03	0.869E 02	0.752E 02	0.683E 02	0.635E 02	0.612E 02	0.595E 02	0.585E 02
80.0	0.109E 03	0.862E 02	0.751E 02	0.687E 02	0.643E 02	0.624E 02	0.609E 02	0.601E 02
85.0	0.107E 03	0.856E 02	0.752E 02	0.693E 02	0.653E 02	0.638E 02	0.627E 02	0.622E 02
90.0	0.106E 03	0.852E 02	0.754E 02	0.702E 02	0.666E 02	0.655E 02	0.647E 02	0.645E 02
95.0	0.104E 03	0.848E 02	0.758E 02	0.711E 02	0.680E 02	0.673E 02	0.668E 02	0.668E 02
100.0	0.103E 03	0.845E 02	0.763E 02	0.721E 02	0.694E 02	0.691E 02	0.689E 02	0.692E 02
105.0	0.101E 03	0.843E 02	0.768E 02	0.731E 02	0.709E 02	0.709E 02	0.709E 02	0.714E 02
110.0	0.100E 03	0.841E 02	0.772E 02	0.741E 02	0.722E 02	0.725E 02	0.728E 02	0.735E 02
115.0	0.992E 02	0.840E 02	0.777E 02	0.750E 02	0.734E 02	0.740E 02	0.745E 02	0.754E 02
120.0	0.980E 02	0.838E 02	0.781E 02	0.758E 02	0.745E 02	0.754E 02	0.760E 02	0.770E 02
125.0	0.968E 02	0.836E 02	0.785E 02	0.765E 02	0.755E 02	0.765E 02	0.773E 02	0.784E 02
130.0	0.958E 02	0.835E 02	0.788E 02	0.771E 02	0.763E 02	0.775E 02	0.783E 02	0.795E 02
135.0	0.948E 02	0.833E 02	0.790E 02	0.776E 02	0.769E 02	0.783E 02	0.792E 02	0.804E 02
140.0	0.939E 02	0.831E 02	0.792E 02	0.780E 02	0.775E 02	0.789E 02	0.799E 02	0.811E 02
145.0	0.931E 02	0.830E 02	0.793E 02	0.783E 02	0.779E 02	0.794E 02	0.804E 02	0.817E 02
150.0	0.924E 02	0.828E 02	0.794E 02	0.785E 02	0.782E 02	0.798E 02	0.808E 02	0.821E 02
155.0	0.918E 02	0.827E 02	0.795E 02	0.787E 02	0.784E 02	0.800E 02	0.811E 02	0.824E 02
160.0	0.913E 02	0.826E 02	0.795E 02	0.788E 02	0.786E 02	0.802E 02	0.813E 02	0.826E 02
165.0	0.910E 02	0.825E 02	0.796E 02	0.789E 02	0.787E 02	0.804E 02	0.814E 02	0.827E 02
170.0	0.907E 02	0.824E 02	0.796E 02	0.790E 02	0.788E 02	0.804E 02	0.815E 02	0.828E 02
175.0	0.905E 02	0.823E 02	0.796E 02	0.790E 02	0.788E 02	0.805E 02	0.815E 02	0.828E 02
180.0	0.904E 02	0.823E 02	0.796E 02	0.790E 02	0.788E 02	0.805E 02	0.816E 02	0.828E 02

TABLE III (cont.)

Energy (MeV)	1.000	1.100	1.200	1.300	1.400	1.500	1.600	1.700
	Centre-of-mass input values for the reaction $^3He(n,p)T$							
σ_T (mb)	882.000	887.000	890.000	891.000	888.000	884.000	877.000	870.000
A' 0	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
A' 1	-0.590	-0.600	-0.602	-0.592	-0.587	-0.586	-0.587	-0.590
A' 2	0.467	0.516	0.575	0.643	0.718	0.794	0.810	0.914
A' 3	-0.002	-0.005	-0.008	-0.013	-0.018	-0.024	-0.031	-0.038
Angle (degree)	Differential cross-sections (mb/sr) in the laboratory system							
0.0	0.937E 02	0.990E 02	0.106E 03	0.115E 03	0.123E 03	0.131E 03	0.132E 03	0.142E 03
5.0	0.931E 02	0.983E 02	0.105E 03	0.114E 03	0.122E 03	0.130E 03	0.131E 03	0.140E 03
10.0	0.912E 02	0.961E 02	0.102E 03	0.111E 03	0.119E 03	0.126E 03	0.127E 03	0.136E 03
15.0	0.882E 02	0.927E 02	0.986E 02	0.106E 03	0.113E 03	0.120E 03	0.120E 03	0.129E 03
20.0	0.844E 02	0.882E 02	0.934E 02	0.100E 03	0.106E 03	0.112E 03	0.112E 03	0.119E 03
25.0	0.799E 02	0.831E 02	0.874E 02	0.931E 02	0.983E 02	0.103E 03	0.103E 03	0.108E 03
30.0	0.752E 02	0.776E 02	0.809E 02	0.854E 02	0.894E 02	0.929E 02	0.931E 02	0.971E 02
35.0	0.705E 02	0.722E 02	0.745E 02	0.777E 02	0.805E 02	0.827E 02	0.828E 02	0.852E 02
40.0	0.662E 02	0.671E 02	0.684E 02	0.705E 02	0.720E 02	0.731E 02	0.730E 02	0.738E 02
45.0	0.624E 02	0.627E 02	0.631E 02	0.641E 02	0.644E 02	0.644E 02	0.641E 02	0.636E 02
50.0	0.594E 02	0.591E 02	0.588E 02	0.588E 02	0.581E 02	0.571E 02	0.567E 02	0.549E 02
55.0	0.573E 02	0.565E 02	0.556E 02	0.548E 02	0.532E 02	0.515E 02	0.510E 02	0.482E 02
60.0	0.561E 02	0.550E 02	0.537E 02	0.522E 02	0.500E 02	0.477E 02	0.471E 02	0.435E 02
65.0	0.558E 02	0.546E 02	0.529E 02	0.510E 02	0.484E 02	0.457E 02	0.450E 02	0.409E 02
70.0	0.564E 02	0.551E 02	0.533E 02	0.511E 02	0.483E 02	0.453E 02	0.446E 02	0.404E 02
75.0	0.577E 02	0.564E 02	0.546E 02	0.523E 02	0.495E 02	0.465E 02	0.457E 02	0.415E 02
80.0	0.595E 02	0.584E 02	0.567E 02	0.545E 02	0.517E 02	0.488E 02	0.480E 02	0.441E 02
85.0	0.618E 02	0.609E 02	0.594E 02	0.573E 02	0.547E 02	0.521E 02	0.513E 02	0.477E 02
90.0	0.643E 02	0.637E 02	0.624E 02	0.606E 02	0.583E 02	0.560E 02	0.552E 02	0.521E 02
95.0	0.669E 02	0.665E 02	0.656E 02	0.641E 02	0.622E 02	0.603E 02	0.595E 02	0.570E 02
100.0	0.695E 02	0.694E 02	0.688E 02	0.676E 02	0.661E 02	0.647E 02	0.639E 02	0.620E 02
105.0	0.720E 02	0.722E 02	0.719E 02	0.710E 02	0.700E 02	0.689E 02	0.682E 02	0.669E 02
110.0	0.743E 02	0.747E 02	0.746E 02	0.742E 02	0.736E 02	0.729E 02	0.722E 02	0.715E 02
115.0	0.763E 02	0.770E 02	0.773E 02	0.771E 02	0.768E 02	0.766E 02	0.759E 02	0.758E 02
120.0	0.781E 02	0.790E 02	0.796E 02	0.797E 02	0.797E 02	0.799E 02	0.792E 02	0.796E 02
125.0	0.796E 02	0.807E 02	0.815E 02	0.819E 02	0.822E 02	0.827E 02	0.821E 02	0.829E 02
130.0	0.809E 02	0.821E 02	0.831E 02	0.837E 02	0.843E 02	0.851E 02	0.844E 02	0.856E 02
135.0	0.818E 02	0.832E 02	0.844E 02	0.852E 02	0.861E 02	0.871E 02	0.864E 02	0.880E 02
140.0	0.826E 02	0.841E 02	0.854E 02	0.864E 02	0.875E 02	0.887E 02	0.880E 02	0.899E 02
145.0	0.832E 02	0.848E 02	0.862E 02	0.873E 02	0.886E 02	0.899E 02	0.893E 02	0.914E 02
150.0	0.836E 02	0.853E 02	0.868E 02	0.880E 02	0.894E 02	0.909E 02	0.903E 02	0.926E 02
155.0	0.839E 02	0.856E 02	0.872E 02	0.886E 02	0.900E 02	0.916E 02	0.910E 02	0.934E 02
160.0	0.842E 02	0.859E 02	0.875E 02	0.889E 02	0.905E 02	0.922E 02	0.915E 02	0.941E 02
165.0	0.843E 02	0.860E 02	0.877E 02	0.892E 02	0.908E 02	0.926E 02	0.919E 02	0.946E 02
170.0	0.844E 02	0.861E 02	0.878E 02	0.894E 02	0.910E 02	0.928E 02	0.922E 02	0.949E 02
175.0	0.844E 02	0.862E 02	0.879E 02	0.895E 02	0.911E 02	0.930E 02	0.923E 02	0.951E 02
180.0	0.844E 02	0.862E 02	0.879E 02	0.895E 02	0.912E 02	0.930E 02	0.924E 02	0.951E 02

TABLE III (cont.)

Energy (MeV)	1.800	1.900	2.000	2.500	3.000	3.500	4.000	4.500
	Centre-of-mass input values for the reaction $^3He(n,p)T$							
σ_T (mb)	862.000	850.000	838.000	756.000	642.000	534.000	459.000	401.000
A' 0	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
A' 1	-0.595	-0.600	-0.606	-0.644	-0.690	-0.737	-0.769	-0.789
A' 2	0.959	0.995	1.024	1.131	1.210	1.265	1.294	1.293
A' 3	-0.046	-0.056	-0.067	-0.137	-0.226	-0.325	-0.432	-0.540
A' 4	0.000	0.000	0.000	0.000	0.033	0.074	0.116	0.162
Angle (degree)	Differential cross-sections (mb/sr) in the laboratory system							
0.0	0.145E 03	0.146E 03	0.145E 03	0.133E 03	0.112E 03	0.910E 02	0.745E 02	0.610E 02
5.0	0.143E 03	0.144E 03	0.144E 03	0.132E 03	0.111E 03	0.900E 02	0.737E 02	0.603E 02
10.0	0.138E 03	0.139E 03	0.139E 03	0.127E 03	0.107E 03	0.870E 02	0.714E 02	0.585E 02
15.0	0.131E 03	0.132E 03	0.131E 03	0.121E 03	0.102E 03	0.823E 02	0.677E 02	0.557E 02
20.0	0.121E 03	0.122E 03	0.121E 03	0.112E 03	0.945E 02	0.762E 02	0.629E 02	0.520E 02
25.0	0.110E 03	0.110E 03	0.110E 03	0.101E 03	0.855E 02	0.691E 02	0.574E 02	0.479E 02
30.0	0.981E 02	0.981E 02	0.976E 02	0.900E 02	0.758E 02	0.615E 02	0.515E 02	0.435E 02
35.0	0.856E 02	0.853E 02	0.848E 02	0.781E 02	0.657E 02	0.536E 02	0.454E 02	0.391E 02
40.0	0.737E 02	0.731E 02	0.725E 02	0.664E 02	0.560E 02	0.460E 02	0.395E 02	0.348E 02
45.0	0.629E 02	0.621E 02	0.613E 02	0.556E 02	0.469E 02	0.389E 02	0.340E 02	0.306E 02
50.0	0.538E 02	0.527E 02	0.517E 02	0.462E 02	0.390E 02	0.326E 02	0.290E 02	0.267E 02
55.0	0.467E 02	0.454E 02	0.443E 02	0.387E 02	0.325E 02	0.274E 02	0.246E 02	0.231E 02
60.0	0.418E 02	0.403E 02	0.390E 02	0.332E 02	0.277E 02	0.233E 02	0.211E 02	0.200E 02
65.0	0.391E 02	0.374E 02	0.360E 02	0.299E 02	0.246E 02	0.206E 02	0.185E 02	0.175E 02
70.0	0.384E 02	0.367E 02	0.352E 02	0.287E 02	0.233E 02	0.192E 02	0.169E 02	0.157E 02
75.0	0.395E 02	0.378E 02	0.363E 02	0.295E 02	0.236E 02	0.191E 02	0.164E 02	0.148E 02
80.0	0.422E 02	0.404E 02	0.389E 02	0.319E 02	0.255E 02	0.202E 02	0.169E 02	0.147E 02
85.0	0.460E 02	0.443E 02	0.429E 02	0.357E 02	0.285E 02	0.225E 02	0.184E 02	0.156E 02
90.0	0.506E 02	0.490E 02	0.476E 02	0.404E 02	0.325E 02	0.256E 02	0.209E 02	0.174E 02
95.0	0.556E 02	0.542E 02	0.529E 02	0.457E 02	0.371E 02	0.295E 02	0.240E 02	0.199E 02
100.0	0.609E 02	0.596E 02	0.584E 02	0.513E 02	0.421E 02	0.337E 02	0.277E 02	0.230E 02
105.0	0.660E 02	0.648E 02	0.637E 02	0.568E 02	0.472E 02	0.382E 02	0.317E 02	0.265E 02
110.0	0.709E 02	0.698E 02	0.688E 02	0.621E 02	0.522E 02	0.428E 02	0.359E 02	0.303E 02
115.0	0.753E 02	0.744E 02	0.735E 02	0.670E 02	0.569E 02	0.471E 02	0.400E 02	0.341E 02
120.0	0.793E 02	0.785E 02	0.777E 02	0.714E 02	0.613E 02	0.513E 02	0.439E 02	0.379E 02
125.0	0.827E 02	0.821E 02	0.814E 02	0.753E 02	0.652E 02	0.550E 02	0.476E 02	0.414E 02
130.0	0.857E 02	0.851E 02	0.845E 02	0.786E 02	0.686E 02	0.584E 02	0.509E 02	0.446E 02
135.0	0.881E 02	0.877E 02	0.871E 02	0.814E 02	0.715E 02	0.613E 02	0.538E 02	0.475E 02
140.0	0.901E 02	0.897E 02	0.892E 02	0.837E 02	0.740E 02	0.638E 02	0.564E 02	0.501E 02
145.0	0.917E 02	0.914E 02	0.909E 02	0.855E 02	0.760E 02	0.659E 02	0.585E 02	0.523E 02
150.0	0.929E 02	0.927E 02	0.922E 02	0.870E 02	0.776E 02	0.676E 02	0.603E 02	0.541E 02
155.0	0.939E 02	0.937E 02	0.932E 02	0.881E 02	0.789E 02	0.690E 02	0.618E 02	0.556E 02
160.0	0.946E 02	0.944E 02	0.940E 02	0.889E 02	0.799E 02	0.701E 02	0.629E 02	0.567E 02
165.0	0.951E 02	0.949E 02	0.945E 02	0.895E 02	0.806E 02	0.709E 02	0.637E 02	0.576E 02
170.0	0.954E 02	0.953E 02	0.949E 02	0.899E 02	0.811E 02	0.714E 02	0.643E 02	0.582E 02
175.0	0.956E 02	0.955E 02	0.951E 02	0.902E 02	0.814E 02	0.717E 02	0.647E 02	0.586E 02
180.0	0.957E 02	0.955E 02	0.952E 02	0.902E 02	0.815E 02	0.718E 02	0.648E 02	0.587E 02

TABLE III (cont.)

Energy (MeV)	5.000	5.500	6.000	6.500	7.000	7.500	8.000	8.500	9.000
	Centre-of-mass input values for the reaction ^{3}He(n, p)T								
σ_T (mb)	359.000	325.000	297.000	274.000	255.000	236.000	220.000	208.000	198.000
A' 0	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
A' 1	-0.801	-0.811	-0.817	-0.808	-0.777	-0.729	-0.680	-0.626	-0.568
A' 2	1.271	1.253	1.235	1.178	1.088	1.008	0.934	0.867	0.811
A' 3	-0.644	-0.736	-0.819	-0.850	-0.860	-0.860	-0.852	-0.837	-0.819
A' 4	0.208	0.254	0.300	0.348	0.396	0.447	0.500	0.549	0.590
Angle (degree)	Differential cross-sections (mb/sr) in the laboratory system								
0.0	0.503E 02	0.425E 02	0.365E 02	0.326E 02	0.296E 02	0.281E 02	0.273E 02	0.274E 02	0.278E 02
5.0	0.499E 02	0.421E 02	0.361E 02	0.322E 02	0.293E 02	0.277E 02	0.269E 02	0.268E 02	0.272E 02
10.0	0.485E 02	0.410E 02	0.352E 02	0.313E 02	0.283E 02	0.266E 02	0.257E 02	0.254E 02	0.256E 02
15.0	0.463E 02	0.393E 02	0.339E 02	0.300E 02	0.270E 02	0.251E 02	0.239E 02	0.235E 02	0.235E 02
20.0	0.437E 02	0.373E 02	0.323E 02	0.285E 02	0.255E 02	0.235E 02	0.221E 02	0.214E 02	0.212E 02
25.0	0.407E 02	0.351E 02	0.307E 02	0.271E 02	0.242E 02	0.222E 02	0.206E 02	0.197E 02	0.192E 02
30.0	0.376E 02	0.329E 02	0.291E 02	0.259E 02	0.232E 02	0.212E 02	0.196E 02	0.186E 02	0.180E 02
35.0	0.345E 02	0.307E 02	0.277E 02	0.249E 02	0.226E 02	0.208E 02	0.192E 02	0.182E 02	0.175E 02
40.0	0.314E 02	0.286E 02	0.263E 02	0.240E 02	0.222E 02	0.207E 02	0.193E 02	0.183E 02	0.177E 02
45.0	0.284E 02	0.264E 02	0.248E 02	0.231E 02	0.218E 02	0.206E 02	0.195E 02	0.188E 02	0.182E 02
50.0	0.254E 02	0.241E 02	0.230E 02	0.219E 02	0.212E 02	0.204E 02	0.197E 02	0.191E 02	0.187E 02
55.0	0.224E 02	0.217E 02	0.211E 02	0.204E 02	0.202E 02	0.198E 02	0.194E 02	0.191E 02	0.189E 02
60.0	0.197E 02	0.193E 02	0.188E 02	0.186E 02	0.188E 02	0.187E 02	0.186E 02	0.185E 02	0.184E 02
65.0	0.172E 02	0.169E 02	0.166E 02	0.166E 02	0.170E 02	0.171E 02	0.172E 02	0.173E 02	0.173E 02
70.0	0.153E 02	0.148E 02	0.144E 02	0.145E 02	0.151E 02	0.153E 02	0.154E 02	0.156E 02	0.157E 02
75.0	0.140E 02	0.132E 02	0.126E 02	0.127E 02	0.132E 02	0.134E 02	0.135E 02	0.137E 02	0.137E 02
80.0	0.134E 02	0.123E 02	0.115E 02	0.114E 02	0.116E 02	0.117E 02	0.117E 02	0.118E 02	0.118E 02
85.0	0.138E 02	0.122E 02	0.110E 02	0.106E 02	0.107E 02	0.105E 02	0.103E 02	0.102E 02	0.101E 02
90.0	0.149E 02	0.130E 02	0.114E 02	0.107E 02	0.104E 02	0.992E 01	0.951E 01	0.921E 01	0.891E 01
95.0	0.169E 02	0.145E 02	0.126E 02	0.116E 02	0.108E 02	0.100E 02	0.932E 01	0.878E 01	0.828E 01
100.0	0.195E 02	0.168E 02	0.146E 02	0.131E 02	0.120E 02	0.108E 02	0.981E 01	0.901E 01	0.831E 01
105.0	0.227E 02	0.196E 02	0.171E 02	0.153E 02	0.138E 02	0.122E 02	0.109E 02	0.985E 01	0.895E 01
110.0	0.261E 02	0.228E 02	0.201E 02	0.179E 02	0.160E 02	0.141E 02	0.125E 02	0.112E 02	0.101E 02
115.0	0.297E 02	0.262E 02	0.233E 02	0.208E 02	0.186E 02	0.163E 02	0.144E 02	0.129E 02	0.116E 02
120.0	0.333E 02	0.296E 02	0.265E 02	0.238E 02	0.213E 02	0.187E 02	0.166E 02	0.149E 02	0.134E 02
125.0	0.367E 02	0.329E 02	0.297E 02	0.268E 02	0.240E 02	0.212E 02	0.189E 02	0.170E 02	0.154E 02
130.0	0.399E 02	0.360E 02	0.328E 02	0.297E 02	0.266E 02	0.236E 02	0.211E 02	0.191E 02	0.173E 02
135.0	0.427E 02	0.388E 02	0.356E 02	0.323E 02	0.291E 02	0.259E 02	0.232E 02	0.211E 02	0.192E 02
140.0	0.453E 02	0.414E 02	0.381E 02	0.347E 02	0.313E 02	0.280E 02	0.252E 02	0.229E 02	0.210E 02
145.0	0.475E 02	0.435E 02	0.402E 02	0.368E 02	0.333E 02	0.298E 02	0.269E 02	0.246E 02	0.226E 02
150.0	0.493E 02	0.454E 02	0.421E 02	0.385E 02	0.349E 02	0.314E 02	0.284E 02	0.260E 02	0.240E 02
155.0	0.508E 02	0.469E 02	0.436E 02	0.400E 02	0.363E 02	0.327E 02	0.297E 02	0.272E 02	0.251E 02
160.0	0.520E 02	0.481E 02	0.448E 02	0.412E 02	0.375E 02	0.338E 02	0.307E 02	0.282E 02	0.261E 02
165.0	0.529E 02	0.490E 02	0.457E 02	0.421E 02	0.383E 02	0.346E 02	0.315E 02	0.289E 02	0.268E 02
170.0	0.535E 02	0.497E 02	0.464E 02	0.427E 02	0.389E 02	0.352E 02	0.320E 02	0.295E 02	0.273E 02
175.0	0.539E 02	0.501E 02	0.468E 02	0.430E 02	0.393E 02	0.355E 02	0.323E 02	0.298E 02	0.276E 02
180.0	0.540E 02	0.502E 02	0.469E 02	0.432E 02	0.394E 02	0.356E 02	0.324E 02	0.299E 02	0.277E 02

TABLE IV. EVALUATED $^{3}He(n, d)D$ CROSS-SECTIONS AND NORMALIZED LEGENDRE COEFFICIENTS A'_l

E_n (MeV)	$\sigma_{n,d}$ (mb)	A'_2	A'_4	A'_6	A'_8	A'_{10}
4.5	1.7	0.633	0.028			
5.0	18.8	1.036	0.313			
5.5	32.7	1.170	0.703	0.060		
6.0	42.6	1.239	1.060	0.155		
6.5	50.3	1.304	1.377	0.285		
7.0	56.1	1.378	1.642	0.430	0.007	0.005
7.5	61.0	1.452	1.863	0.583	0.038	0.010
8.0	64.9	1.503	2.043	0.734	0.068	0.013
8.5	68.0	1.528	2.169	0.879	0.096	0.020
9.0	70.7	1.538	2.239	1.019	0.121	0.027
9.5	72.9	1.540	2.282	1.148	0.146	0.031
10.0	74.8	1.540	2.310	1.267	0.168	0.038
10.5	76.3	1.538	2.326	1.369	0.183	0.042
11.0	77.7	1.529	2.332	1.455	0.197	0.049

rises from zero at the forward threshold of 4.36 MeV to about 70 mb at 10 MeV. The readings listed in Table IV extend up to 11 MeV, corresponding to the high-energy end (10 MeV) of the D(d, n)^{3}He cross-section evaluation [5].

In Fig. 5 the $A_1^!$ coefficients are plotted versus the neutron energy scale. The $A_2^!$ coefficient decreases steeply to zero at the reaction threshold. Numerical values of the coefficients are also given in Table IV.

For the ^{3}He(n, d)D reaction, the backward threshold is at 6.57 MeV neutron energy. As this reaction is of minor importance in neutron metrology, no differential cross-sections in the LAB system are given here. To obtain them, the differential centre-of-mass cross-sections calculated from Eq. (11) and the data of Table IV have to be divided by $J = d\Omega_{LAB}/d\Omega_{CM}$, the ratio of the differential LAB and centre-of-mass solid angles. Values of J can be calculated, for example, following the equations and the computer program of Horstmann and Liskien [9].

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THE $^{3}He(n,p)T$, $^{6}Li(n,\alpha)T$ AND $^{10}B(n,\alpha)$ STANDARD CROSS-SECTIONS*

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Abstract

THE $^{3}He(n,p)T$, $^{6}Li(n,\alpha)T$ AND $^{10}B(n,\alpha)$ STANDARD CROSS-SECTIONS. -

The evaluated cross-sections in the ENDF/B-III library are reviewed and compared with recent experimental measurements over the energy range established for each standard cross-section. While the $^{3}He(n,p)$, $^{6}Li(n,\alpha)$ and $^{10}B(n,\alpha)$ data seem to be adequately established up to approximately 10 keV, the ^{10}B data should be separated into the α_0 and α_1 channels, which is now allowed by a recent ENDF format revision. All of the cross-sections require further study above 10 keV to gain the precision necessary for standards. Recent experiments on ^{10}B carried out at Gulf Radiation Technology, San Diego, Calif. (GRT), are briefly discussed and the results presented.

1. INTRODUCTION

This paper is confined to a discussion of the standard neutron cross-sections, in particular to the light-element absorption cross-sections, since the hydrogen scattering cross-section is well established as a primary standard from a few hundred keV to 20 MeV. Below 1 keV, the hydrogen scattering cross-section is constant, while the $^{3}He(n,p)T$, $^{6}Li(n,\alpha)T$ and $^{10}B(n,abs)$ cross-sections are generally given by a 1/v energy dependence plus a small constant term, $\Delta\sigma$. A more precise representation of the absorption cross-section is obtained from the low-energy S-wave expansion derived by Bergman and Shapiro [1]:

$$\sigma_{abs} = A/\sqrt{E_n} + \Delta\sigma + B\sqrt{E_n} + CE_n + \ldots \qquad (1)$$

where the constants are quoted with E in eV and σ in barns.

Many of the present data for σ_{abs} have been obtained through fitting measurements of the total cross-section and generally assuming that the constants B and C are zero. Some recent measurements of the scattering cross-sections have been combined with data on σ_{tot} to obtain a value for the constant $\Delta\sigma$, which was found to be small, again neglecting terms containing B and C. Several years ago, Gubernator and Moret [2] fitted all available data on σ_{tot} and σ_{abs} for ^{10}B to obtain values of all of the parameters listed in Eq.(1).

* Work performed under the auspices of the US Atomic Energy Commission.

The standard cross-sections on the ENDF/B-III files have been evaluated by various CSEWG[1] organizations and reviewed by the CSEWG Standards Subcommittee. A status report on the standards data was recently written by Drake [3], and the summary documents for each evaluated file have been included in Drake's report.

Since other standard cross-sections are often measured with respect to the hydrogen scattering cross-section, the theoretical analysis of Hopkins and Breit [4] was used to generate the hydrogen evaluation for ENDF/B-III at the Los Alamos Scientific Laboratory (LASL) [5]. These data are available as file MAT 1148 and it should be noted that the angular distributions of the elastic neutrons are neither isotropic nor symmetric about 90° above a few MeV, as assumed in all of the previous evaluated files. Even though this cross-section is assumed to be known with high accuracy, it should be pointed out that angular distribution measurements of the neutrons and recoil protons are still required in order to determine the accuracy with which hydrogen can be used as a standard.

At low energies, exoergic reactions which have a large cross-section and a smooth energy dependence make the best standards since such reactions produce easily detectable signatures in neutron flux monitors. These criteria combine to make the $^{3}He(n,p)T$, $^{6}Li(n,\alpha)T$ and $^{10}B(n,abs)$ reactions excellent standard cross-sections up to the region of approximately 100 keV. However, since ^{3}He is a gas and is easily contaminated with T, lithium and boron have enjoyed wider application. Each of these reactions is described in more detail in the following sections.

2. $^{3}He(n,p)T$ $\quad Q = +0.7645$ MeV

MAT 1146 on ENDF/B-III is an evaluation performed by Stewart and LaBauve [6] several years ago. The thermal value of 5327 b was derived from measurements made by Als-Nielsen and Dietrich [7], and the cross-section is assumed to go as 1/v up to 1.7 keV. Above this energy, the slope is changed to reflect higher-energy measurements of Gibbons and Macklin [8] and Macklin and Gibbons [9] as shown in Fig.1. These comparisons are extended to 100 keV in Fig.2. Figure 3 shows an extension of the data to 1 MeV including measurements at other laboratories[2]. Although the thermal cross-section is known to better than 1%, the energy at which this cross-section deviates from 1/v is not well established. In addition, it should be pointed out that precise experimental measurements have not been carried out above a few eV, thereby placing severe restrictions upon the accuracy accompanying the use of the $^{3}He(n,p)T$ cross-section standard. The 10% error estimates on the Oak Ridge National Laboratory (ORNL) data are directly related to uncertainties in the analysis of the samples employed. Certainly, further work is needed on this cross-section standard, especially above 100 eV.

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[1] The Cross-Section Evaluation Working Group which is chaired by the Head of the National Neutron Cross-Section Center at Brookhaven National Laboratory.

[2] The experimental data of Costello et al. (Nucl. Sci. Eng. 39 (1970) 409), which show a constant cross-section of about 900 mb from 300 keV to 1 MeV, have been omitted for the sake of clarity.

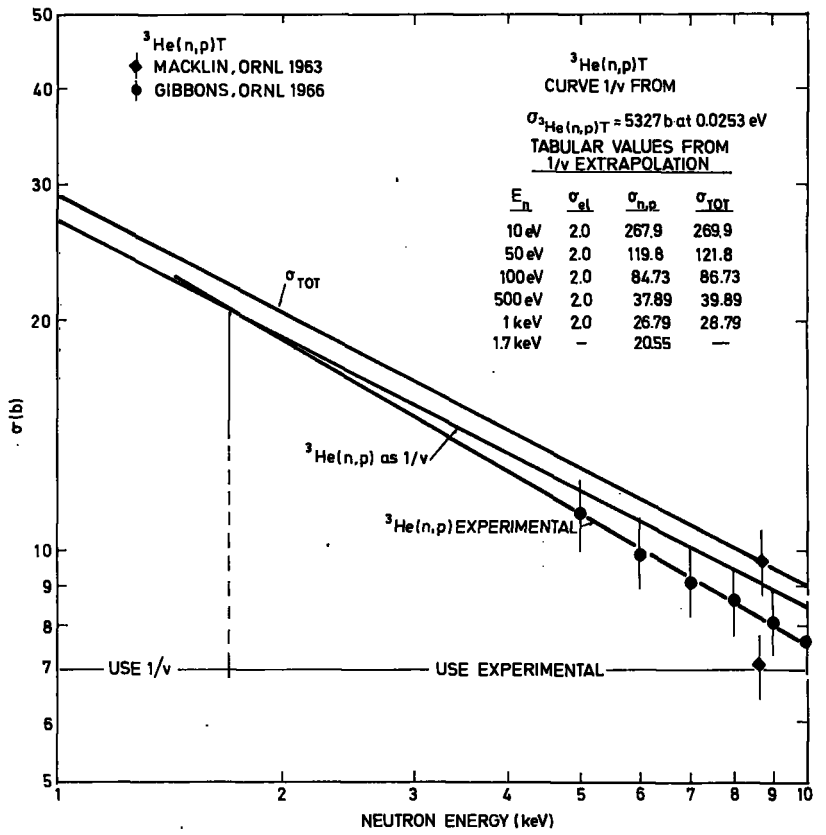

FIG. 1. The (n, p) and total cross-sections for ^{3}He from 1 to 10 keV. The curve drawn through the experimental points deviates from 1/v at 1.7 keV.

3. ^{6}Li(n,α)T Q = +4.786 MeV

The ENDF/B-III data, MAT 1115, were furnished by LASL [10] and were based on the evaluation by Uttley et al. [11] below 500 keV. It is well known that gross inconsistencies have been observed in the (n,α) and total cross-sections for ^{6}Li, both in the magnitude of the cross-section and in the energy scale, especially near the 250-keV resonance. These inconsistencies made it virtually impossible to perform an evaluation using all of the partial cross-sections. The evaluation of Uttley et al., based on a theoretical analysis of the total cross-section, gave results that are consistent with preliminary measurements of Coates et al. [12] on the (n,α) cross-section. As this evaluation showed consistency between the partial and total cross-section, it was chosen until the time when the experimental problems can be studied in more detail.

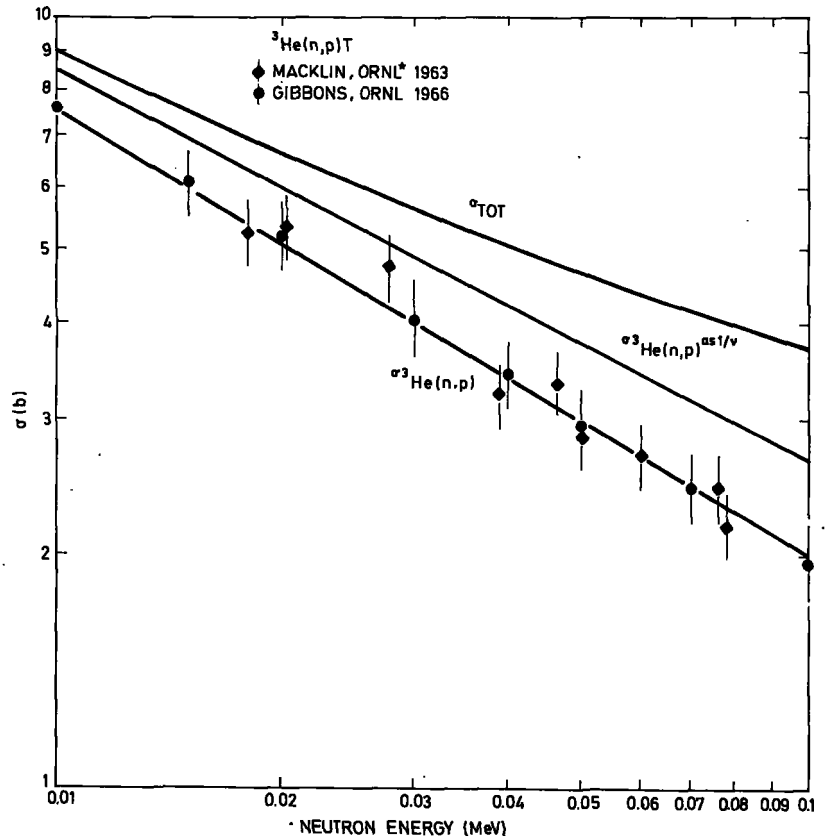

FIG. 2. The (n, p) and total cross-sections for ^{3}He from 10 to 100 keV.

During the past 2-3 years, much progress has been made. For example, 'thin' and 'thick' lithium glass detector responses have been studied, especially at Harwell, and consistent results obtained. Further work is under way at Cadarache and at Harwell to determine the cross-section and to establish the energy scale near the resonance peak. Preliminary (n,α) results at Argonne [13] show better agreement with the Uttley evaluation than the older data of Meadows.

The (n,α) data in MAT 1115 are calculated from the formula:

$$\sigma_{n,\alpha} = (149.56/\sqrt{E_n}) - 0.024 \qquad (2)$$

with the energy in eV and the cross-section in barns. The cross-sections in the file deviate from a strict 1/v dependence by a maximum of 0.4% up to 10 keV, giving a thermal cross-section of 940.25 b. Figure 4 shows the evaluation of Diment and Uttley (from which the ENDF/B-III data were derived), along with some of the recent experimental measurements.

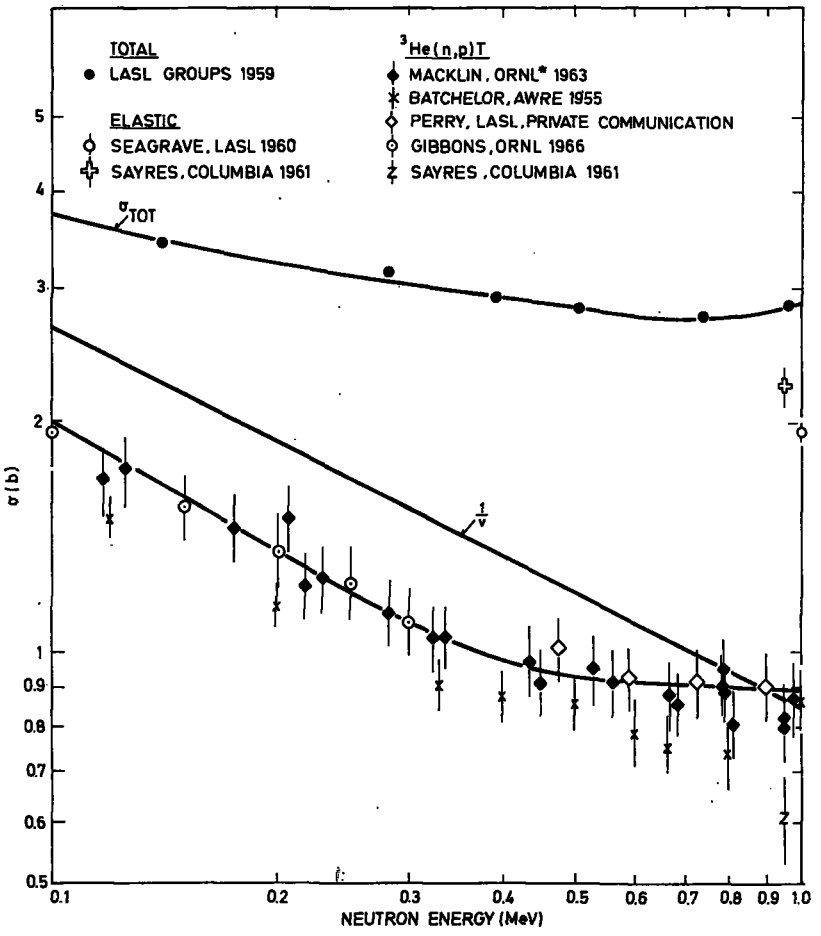

FIG. 3. The (n,p), elastic, and total cross-sections for ^{3}He from 100 keV to 1 MeV. The Costello data, which indicate a cross-section of about 900 mb from 300 keV to 1 MeV, have been omitted for the sake of clarity.

These data indicate that the (n,α) cross-section up to 10 keV is accurately determined unless discrepancies evolve in establishing the energy scale in this energy region. It is important, however, that the cross-section be extended to 100 keV with high precision for use as a standard. Further extension to 1 MeV would be useful.

4. $^{10}B(n,\alpha_0)^7Li$; $(n,\alpha_1)^7Li^*$ $Q_0 = +2.792$ MeV; $Q_1 = +2.314$ MeV

MAT 1155 on ENDF/B-III contains a ^{10}B evaluation by Irving [14] performed several years ago. To conform to the recommendations made by the CSEWG Standards Subcommittee for ENDF/B-III, however,

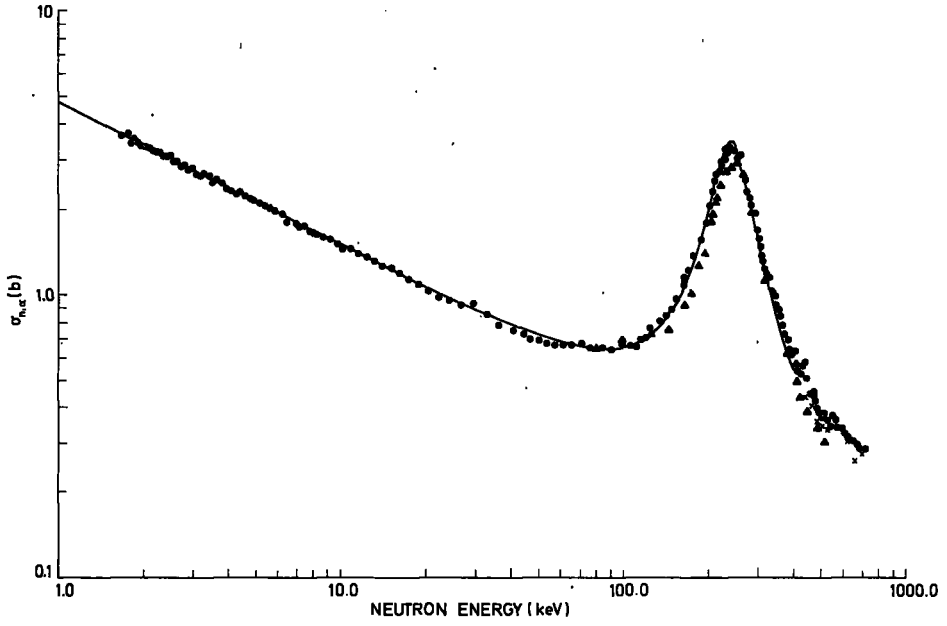

FIG. 4. The ^{6}Li(n, α)T cross-section from 1 keV to 1 MeV.
..... experimental data of Coates et al.; ——— Diment and Uttley calculation, representing the ENDF/B-III evaluation; x x x x Diment and Uttley experimental values; ▲▲▲▲▲▲ Fort values.

LaBauve [15] at LASL made changes in the (n,α) cross-section over the energy region where it is employed as a standard. Therefore, the (n,α) data in MAT 1155 were taken from the evaluation by Sowerby et al. [16] up to 100 keV. This procedure gave consistency between the ^{6}Li/^{10}B ratio measurements and the elastic scattering data of Asami and Moxon [17] on ^{10}B.

The (n,α) cross-sections up to 100 keV were calculated from the formula of Sowerby et al. [16], which is given below:

$$\sigma_{n,\alpha} = \frac{13.837}{\sqrt{E_n}} - 0.312 - 1.014 \times 10^{-2} \sqrt{E_n} + \frac{2.809 \times 10^{5}}{\sqrt{E_n}[(170.3-E_n)^2 + 2.243 \times 10^{4}]} \qquad (3)$$

with σ in barns and E in keV. The thermal cross-section in MAT 1155 obtained with the above formula is 3836.45 b.

The total (n,α) cross-section given in ENDF/B-III is not an adequate representation of the standard cross-section since the 478-keV γ-ray is often the observed quantity in a flux monitor. It is only recently that the ENDF/B format[3] has been extended to allow the (n,α_0) and (n,α_1) channels to be input as separate reactions.

[3] MT = 780 is now assigned for (n, α_0) and MT = 781 for (n, α_1). The sum of these two reactions will still be input into MT = 107 which is the total (n, α) cross-section.

Measurements have been made on the $(n,\alpha_1\gamma)$ and total (n,α) cross-sections at GRT by Friesenhahn et al. [18] which will be considered in the next evaluation for the ENDF/B files, along with other recent experiments. Since the GRT data have not yet been published, these experiments are discussed briefly below and the results presented.

Both the total (n,α) and $(n,\alpha_1\gamma)$ cross-sections were measured relative to hydrogen scattering in the region from 4 to 750 keV with an extension to 1 MeV for the latter. Quoting from Friesenhahn et al. [18]: "The data were obtained with neutrons from an electron linac target by employing time-of-flight techniques. Hydrogen and methane gas proportional counters were used to determine the neutron flux from ≈ 1 keV to 1 MeV. In order to allow consistency checks to be made, the $^{10}B(n,\alpha)$ cross-section was measured with both BF_3 gas proportional counters and with a parallel plate ionization chamber containing ^{10}B-loaded self-supporting films. The $^{10}B(n,\alpha_1\gamma)$ cross-section was measured with a lithium-drifted germanium spectrometer."

The total (n,α) cross-sections are plotted in Fig.5 along with measurements from other laboratories and the ENDF/B-III evaluation. Note that the GRT experimental results are higher above about 60 keV than earlier experiments and, therefore, the ENDF/B curve; they also show more

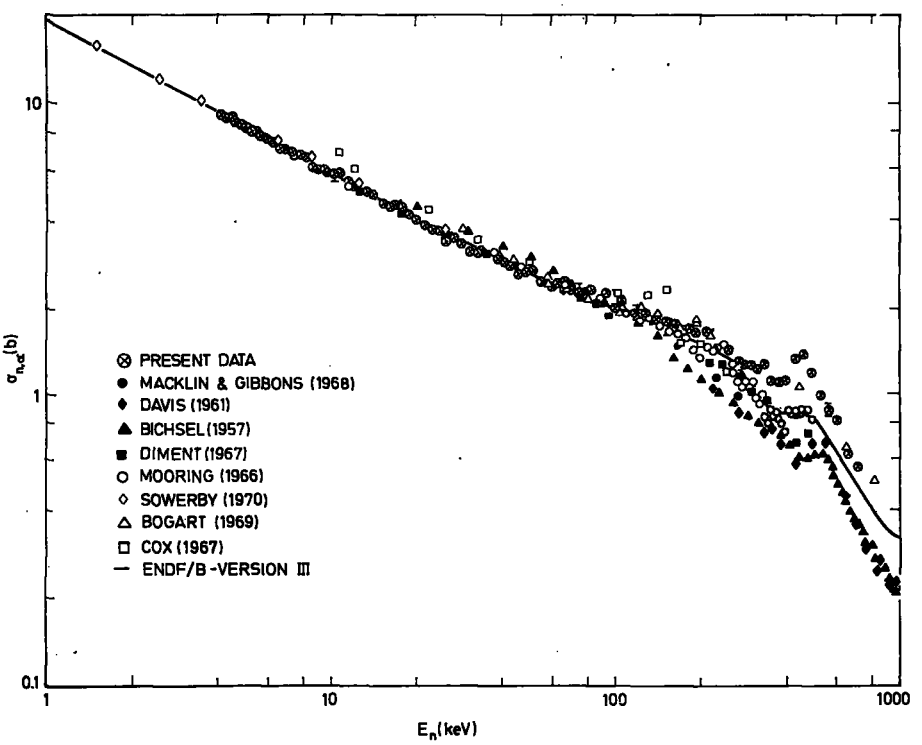

FIG. 5. The $^{10}B(n,\alpha)$ cross-sections with the GRT results labelled 'present data'. (This graph is Fig. 45 of Ref. [18].)

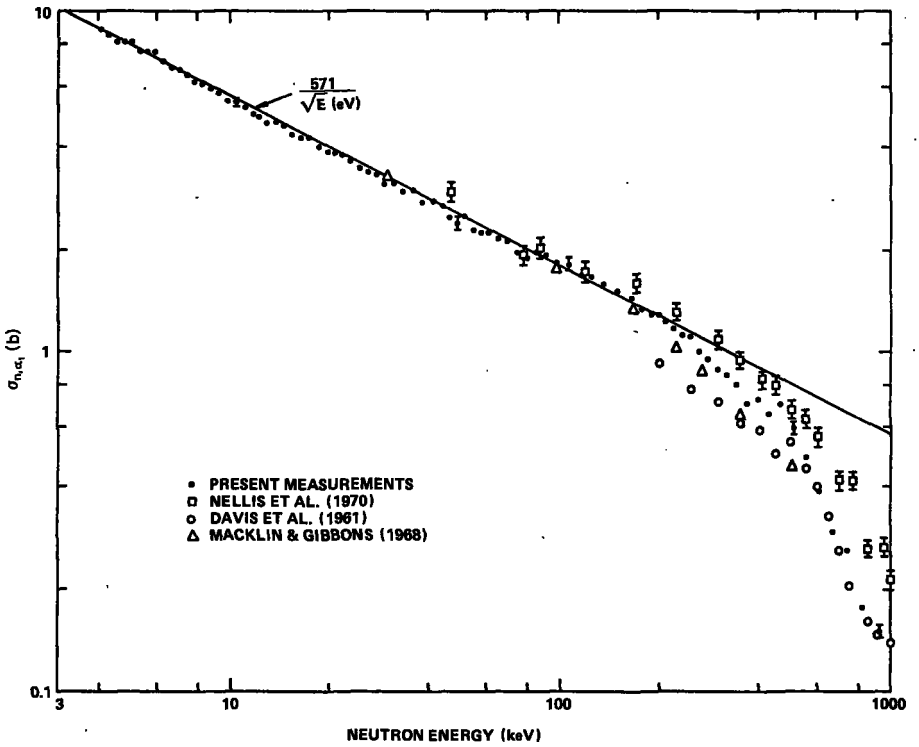

FIG. 6. The $^{10}B(n, \alpha_1\gamma)$ cross-section up to 1 MeV with the GRT measurements labelled 'present measurements'. (This graph is Fig. 42 of Ref. [18].)

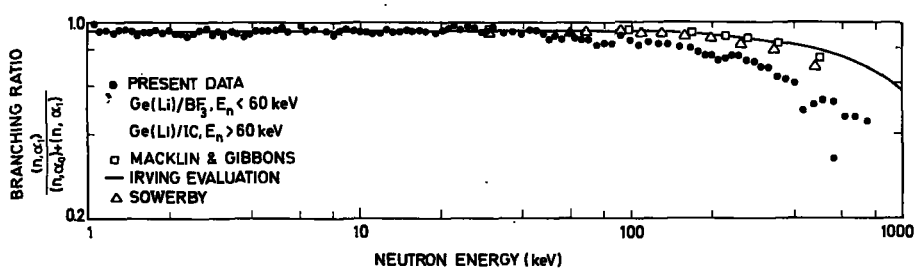

FIG. 7. The branching ratio, i.e. the ratio of the $(n, \alpha_1\gamma)$ to the total (n, α) cross-section with the GRT results labelled 'present data'. (This graph is Fig. 46 of Ref. [18].)

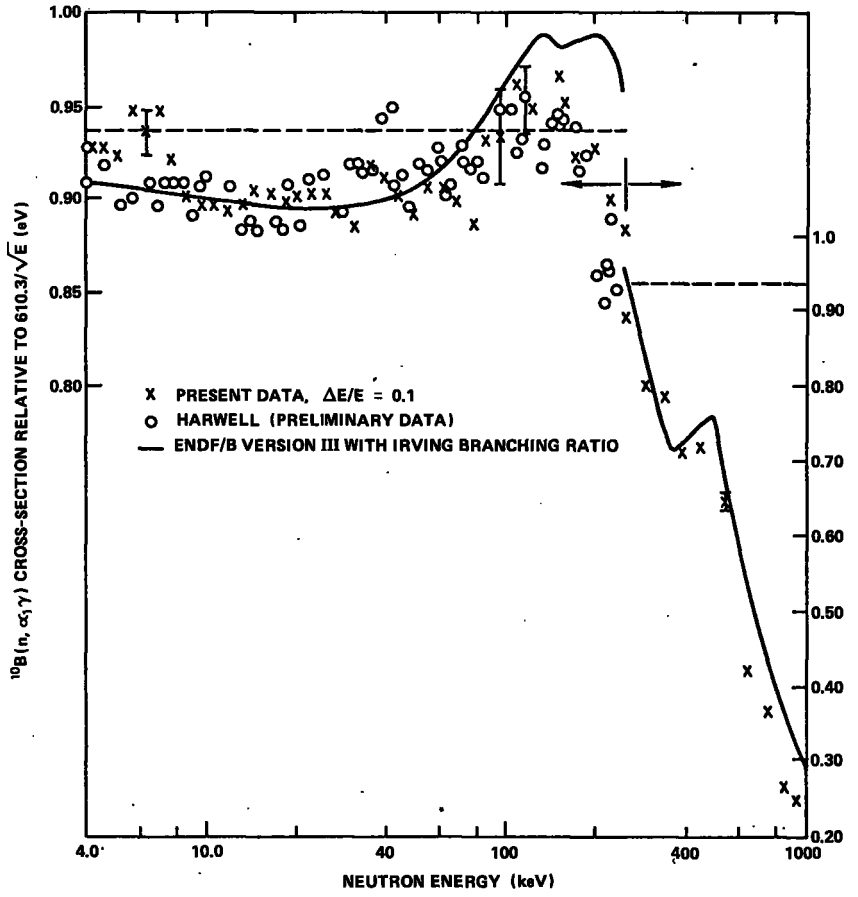

FIG. 8. The $^{10}B(n, \alpha_1\gamma)$ cross-section relative to $610.3/\sqrt{E}$ (eV) with the GRT measurements labelled 'present data'. (This graph is Fig. 43 of Ref. [18].)

structure than previously observed near 450 keV. On the other hand, the GRT $(n,\alpha_1\gamma)$ data fall in between the measurements of Nellis et al. [19] and Macklin and Gibbons [20] above 200 keV, as shown in Fig.6.

These results combine to give a branching ratio[4] which drops significantly below the measurements of Macklin and Gibbons [20] and of Sowerby et al. [21], as indicated in Fig.7. The Irving evaluation shown by the smooth curve is discussed in Ref.[14]. Certainly, these data show disagreement with earlier measurements at the higher energies which has not been resolved. On the other hand, the GRT $(n,\alpha_1\gamma)$ data show good agreement with preliminary measurements at Harwell up to 200 keV. The $(n,\alpha_1\gamma)$ cross-sections are plotted relative to $610.3/\sqrt{E}$(eV) in Fig.8, using the ENDF/B-III total (n,α) cross-section and Irving's branching

[4] Defined here as the ratio of the $(n, \alpha_1\gamma)$ to the total (n, α) cross-section.

ratio. Currently under way at GRT is a programme to extend these measurements to energies below 4 keV and to confirm the present (n,α) results above 100 keV.

In a re-evaluation of the $^{10}B(n,\alpha)$ cross-sections, it should be borne in mind that the (n,γ), (n,p) and $(n,t2\alpha)$ cross-sections have positive Q-values and all of these reactions are assumed to make a zero contribution to the absorption cross-section below 1.2 MeV in the ENDF/B file. This assumption has not been verified experimentally, as pointed out by Stewart [22] several years ago. The problem is twofold, since the (n,α) cross-section is often derived from measurements of the total cross-section, assuming $\sigma_{n,\alpha} \equiv \sigma_{tot}$. Such an assumption could even place the normalization point itself in question. Following previous evaluations, however, this assumption is carried over to the ENDF/B file up to 1.2 MeV. Although some experimental work is currently under way in the USA to determine the importance of the $(n,t2\alpha)$ reaction at low energies, the (n,γ) and (n,p) contributions have not received attention.

5. CONCLUSIONS

Although hydrogen scattering has been accepted as a well-known standard cross-section, further angular distribution measurements are required to determine the accuracy with which it can be applied experimentally. Suffice it to conclude that such accuracy will be intimately related to the accuracy with which the angular distributions themselves can be determined.

The $^{3}He(n,p)T$ cross-section is assumed to follow a 1/v dependence up to 1.7 keV, but the energy at which the deviation occurs is not well established. Below 100 eV, this assumption is perhaps valid to 2%. The $^{6}Li(n,\alpha)T$ and $^{10}B(n,abs)$ cross-sections closely follow a 1/v dependence to approximately 10 keV and are therefore well accepted standards. It would be extremely useful to establish the $^{6}Li(n,\alpha)$, $^{10}B(n,\alpha)$ and $^{10}B(n,\alpha_1\gamma)$ reactions to higher energies with good precision in order to overlap the energy range where hydrogen is the accepted standard. Work is under way at GRT which should extend these cross-sections to 1 MeV, although the observed structure may limit their usefulness over part of the energy range.

ACKNOWLEDGEMENTS

I sincerely appreciate receiving the GRT (n,α) data on ^{10}B from M.P. Fricke and permission to reproduce Figs 5-8 from their work prior to publication. I also wish to thank M.S. Coates for permission to use the preliminary data on ^{6}Li in Fig.4. Finally, without the contributions of the following persons to the ENDF/B-III files this review could not have been prepared: M.E. Battat, M.K. Drake, D.C. Irving, R.J. LaBauve and P.G. Young.

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DISCUSSION

M.S. COATES: I would like to support your remarks about the $^{3}He(n, p)T$ reaction. One must always be able to use a standard. Unless it is possible to build a simple counter in which it is possible to have confidence, the standard will not be used even though it may be very well known. The hydrogen cross-section at lower energies is such an example.

The attractiveness of using ^{3}He in a gas scintillation counter adds weight to your recommendation that the $^{3}He(n,p)T$ cross-section should be better known. For exactly this reason, higher accuracy has been requested for this cross-section in the UK nuclear data request list, but no work has been done. Perhaps this Panel should make a similar recommendation.

L. STEWART: When I visited Mol about one year ago, they were interested in using ^{3}He counters but could not get pure enough ^{3}He.

GENERAL DISCUSSION

A.J. DERUYTTER: To what accuracy do you think the $^{6}Li(n,\alpha)$ cross-section is known up to 100 keV based on your evaluation?

L. STEWART: I would like to divide up the energy range. Up to 10 keV the cross-section is known to 2%; up to 100 keV, to 3-3.5%.

A.J. DERUYTTER: I agree with that. Unfortunately, one of the conclusions of the Argonne Symposium from the working group on light-element standards was that, in the energy range up to 100 keV, the $^{6}Li(n,\alpha)$ cross-section was given by an expression proportional to $1/v$ to $\pm 1\%$.

C.D. BOWMAN: We now have an accuracy of perhaps 3% on the $^{6}Li(n,\alpha)$ cross-section at 100 keV, and if we want accuracies of 1-2%, perhaps we must give up the idea of using the total cross-section to provide guidance about the value of the (n,α) cross-section. The (n,α) cross-section is about 30% of the total cross-section. Therefore, if one measures the total cross-section to 1%, this implies a limit of about 3% on the accuracy to which one can get information about the (n,α) cross-section. In addition, a shape analysis with R-matrix theory requires knowledge of the bound levels and also of the excited-state levels to well above the resonance. Considering all these uncertainties, I think it is not possible to get information from the total cross-section which will help to bring the accuracy of the $^{6}Li(n,\alpha)$ cross-section below 4%. These types of two-channel R-matrix analyses of the 240-keV resonance probably represent the greatest effort that has gone into fitting any resonance anywhere. I am doubtful that one can go much further.

H. LISKIEN: In the case of standards I have always thought that more information than just the integral cross-sections was required. Are there plans to include angular differential cross-sections in ENDF/B?

L. STEWART: At present, the only angular distributions in ENDF/B are for reactions which have neutrons in the exit channel. Inclusion of angular distributions for reactions such as $^{3}He(n,p)T$ and $^{6}Li(n,\alpha)T$ is being discussed in our national Cross-Section Evaluation Working Group (CSEWG).

A.J. DERUYTTER: For the $^{10}B(n,\alpha)$ cross-section, if the total (n,α) cross-section and the branching ratio are included in the files, is it necessary to include the $^{10}B(n,\alpha,\gamma)$ cross-section as well?

L. STEWART: At present, the ENDF/B files allow only cross-sections and not branching ratios, so we shall probably put in the (n,α_0) and (n,α_1) cross-sections from which the branching ratio can be calculated.

M.S. COATES: I think it is important to include the (n,α,γ) cross-section as well as the total (n,α). In many applications it is easier to measure neutron flux by counting the 478-keV gamma rays from the $^{10}B(n,\alpha,\gamma)$ reaction. If it is necessary to go through several extra steps to obtain a standard $^{10}B(n,\alpha,\gamma)$ cross-section, it is much more difficult to identify inherent errors in the experimental technique.

L. STEWART: I agree that the $^{10}B(n,\alpha_0)$ and $^{10}B(n,\alpha_1)$ reactions should be evaluated separately. We shall still include in our files the total (n,α) cross-section so the system will be over-determined. I hope it will be consistent.

IV. FISSION AND CAPTURE STANDARDS

A. The $^{235}U(n,f)$ cross-section for fast neutrons

ORELA MEASUREMENTS OF THE $^{235}U(n, f)$ CROSS-SECTION TO 100 keV*

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Abstract

ORELA MEASUREMENTS OF THE $^{235}U(n,f)$ CROSS-SECTION TO 100 keV.

Two sets of measurements of the $^{235}U(n,f)$ cross-section made with ORELA in the energy range below 100 keV are compared. In both measurements, fission was detected by coincidence between pulses from a multi-plate fission chamber and a two-segment boron-loaded scintillator tank. Neutron flux was measured using a beryllium-walled parallel-plate pulse ion chamber containing $^{10}BF_3$. The response of the ion chamber was assumed to be proportional to the shape of the $^{10}B(n,\alpha)$ cross-section, for which the shape given by Sowerby et al. was assumed. In the experiment of Gwin et al., ORELA was operated at sufficiently low pulse frequency so that measurements could extend through the thermal energy range. They were normalized to the value 83.74 b·eV over the energy interval 0.02 to 0.4 eV. The data of Perez et al. were obtained at higher accelerator pulse frequency and are normalized over the energy range 100-200 eV where several recent measurements are in agreement. Preliminary data from both ORELA experiments are compared with other measurements.

The purpose of this paper is to elucidate briefly two measurements of the $^{235}U(n, f)$ cross-section in the 0.1- to 100-keV range. Both measurements were made at the Oak Ridge Electron Linear Accelerator (ORELA) using apparatus chosen primarily to allow simultaneous measurements of fission and capture cross-sections, but these measurements have weight and need to be considered. Most of these results are believed to be essentially final, and for each experiment the respective authors are now preparing more complete documentation [1, 2].

In each case, fission was detected by coincidence between pulses from a multi-plate fission chamber utilizing 99.7% pure ^{235}U and a large two-segment boron-loaded scintillator tank [3], each segment biased at a fast pulse height corresponding to an electron energy release of about 0.2 MeV. A total deposited energy of $\gtrsim 3$ MeV was required in the scintillator for an event to be recorded. The fission chamber has 20 plates, 7.5 cm in diameter, spaced by ~ 1.5 mm, with electroplated ^{235}U deposits of about 1.7 mg/cm^2 on both positive and negative plates. As operated, the fission chamber efficiency for detecting a fission event was about 0.6, and the tank efficiency for a fission was about 0.85 or 0.6, depending on whether or not a fast coincidence was required between the two segments of the tank.

Background, including that from off-energy neutrons, was determined by the 'notch filter' technique to be $< 1\%$. By studying tagged events from a ^{252}Cf-loaded fission chamber present during the measurements, it was possible to exclude any effects of efficiency variations as a function of neutron energy.

* Research sponsored by the US Atomic Energy Commission under contract with the Union Carbide Corporation.

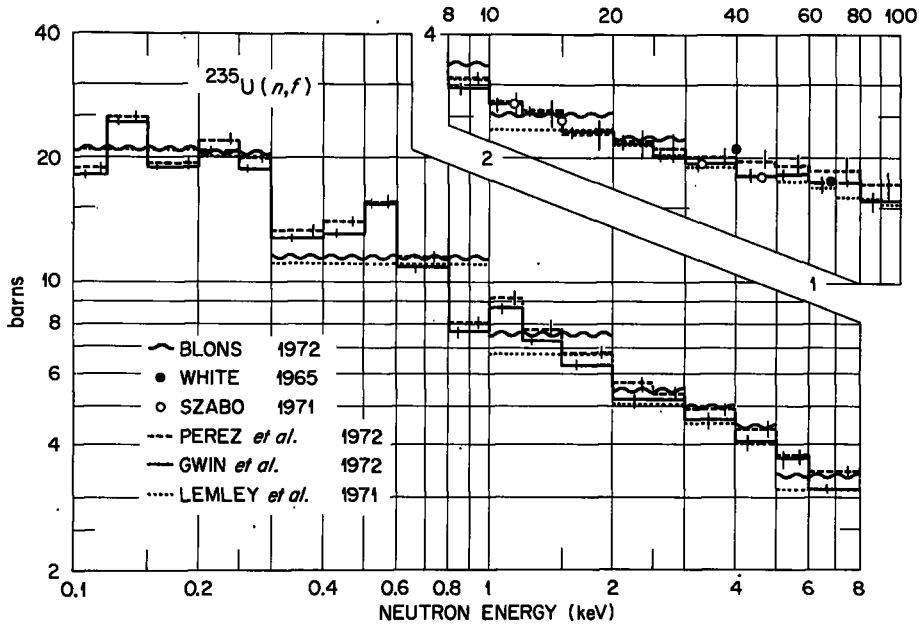

FIG. 1. Averaged $^{235}U(n,f)$ cross-sections of Perez et al. [1] and Gwin et al. [2] compared with some other representative values from the literature.

Neutron flux was measured using a beryllium-walled parallel-plate pulse ion chamber containing ~ 9 mg/cm^2 of $^{10}BF_3$. This chamber was used in the transmission mode and recorded the spectrum during the fission measurements. A stable fraction of the spectrum was selected for use, and it was then assumed that the efficiency of the chamber was proportional to the $^{10}B(n,\alpha)$ cross-section.

Beyond the similarities of equipment listed above, the experiments and analysis of Perez et al. [1] and of Gwin et al. [2] were quite independent. Both, however, assumed the shape of the $^{10}B(n,\alpha)$ cross-section given by Sowerby [4].

Gwin, Silver, Ingle, and Weaver performed their main experiments at a flight distance of 40 m and in some cases used a sufficiently low burst repetition rate so that the measurements included the thermal neutron energy range. These measurements were normalized to the value of 83.74 b·eV over the energy interval 0.02 to 0.4 eV, implying a normalization of about 2.5% below the values given by Deruytter and Wagemans in 1971 for the 7.4- to 10-eV interval [5]. (Different thermal cross-sections were assumed.)

Perez, de Saussure, Silver, Ingle, and Weaver utilized measurements at both 40 m and 150 m; the results at the shorter distance are now prepared for publication, while those at the longer flight path are considered more preliminary. The present results are to be used in preference to the preliminary results released in 1971 based on part of the same experimental

TABLE I. AVERAGED CROSS-SECTIONS AND ESTIMATED STANDARD ERRORS FOR NEUTRON FISSION of $^{235}U(n, f)$

$E_1 - E_2$ (keV)	Perez at al. [1] σ_f (b)	Perez at al. [1] $\Delta\sigma_f$ (b)	Gwin et al. [2] σ_f (b)	Gwin et al. [2] $\Delta\sigma_f$ (b)	Ratio Perez/Gwin
0.10 – 0.12	18.9	0.5	18.2	0.4	1.04
0.12 – 0.15	25.0	0.8	24.4	0.5	1.02
0.15 – 0.20	19.4	0.6	19.0	0.4	1.02
0.20 – 0.25	21.9	0.7	20.7	0.4	1.06
0.25 – 0.30	19.9	0.6	18.8	0.4	1.06
0.30 – 0.40	13.3	0.4	12.8	0.3	1.04
0.40 – 0.50	14.0	0.4	13.1	0.3	1.07
0.50 – 0.60	15.6	0.5	15.5	0.4	1.01
0.60 – 0.80	11.5	0.3	10.9	0.3	1.06
0.80 – 1.00	7.98	0.24	7.6	0.3	1.05
1.0 – 1.2	9.14	0.36	8.64	0.3	1.06
1.2 – 1.5	7.72	0.31	7.22	0.2	1.07
1.5 – 2.0	6.76	0.27	6.28	0.2	1.08
2.0 – 2.5	5.65	0.23	5.14[a]	0.2	1.07
2.5 – 3.0	5.30	0.21			
3.0 – 4.0	4.86	0.19	4.58	0.2	1.06
4.0 – 5.0	4.36	0.17	4.08	0.2	1.07
5.0 – 6.0	3.77	0.15	3.72	0.2	1.01
6.0 – 8.0	3.44	0.14	3.11	0.1	1.11
8.0 – 10.0	3.14	0.13	2.95	0.1	1.06
10 – 12	2.74	0.22	2.70	0.10	1.01
12 – 15	2.56	0.20	2.59	0.10	0.99
15 – 20	2.29	0.18	2.29	0.10	1.0
20 – 25	2.19	0.17	2.18	0.09	1.0
25 – 30	2.11	0.17	2.04	0.09	1.03
30 – 40	2.02	0.16	1.94	0.08	1.04
40 – 50	1.96	0.15	1.81	0.07	1.08
50 – 60	1.92	0.15	1.82	0.07	1.05
60 – 80	1.85	0.14	1.74	0.09	1.06
80 – 100	1.73	0.13	1.57	0.09	1.10

[a] For 2.0- to 3.0-keV region.

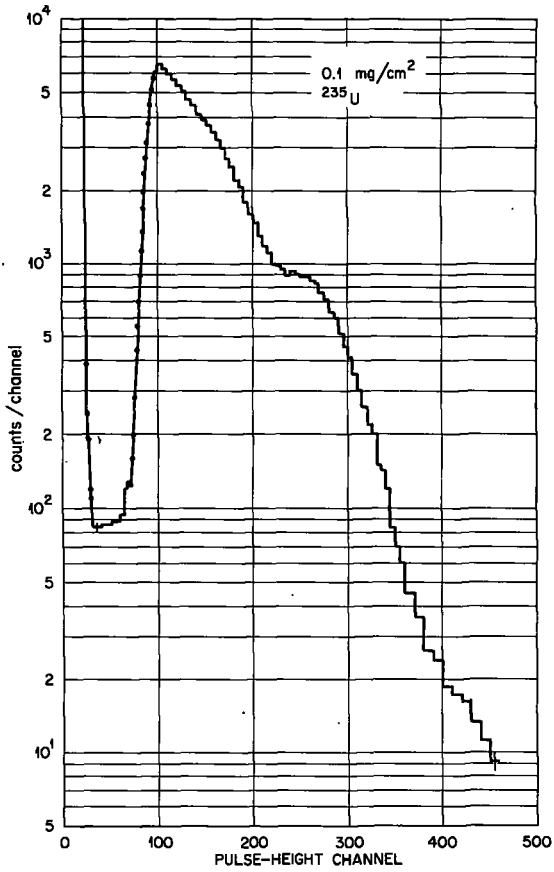

FIG. 2. Spectrum of integrated charge pulses for ^{235}U fission. A 0.1-mg/cm^2 deposit was placed on the negative electrode of the parallel plate ion chamber with ~ 6-mm spacing.

data [6]. For the 40-m measurements a boron filter was used to cut off the neutrons of lowest energy to enable a higher repetition rate to be used. The results were normalized in the 100- to 200-eV range to a value consistent with the ORNL-RPI experiment [7], the Saclay work of Blons [8] and the LASL work of Lemley [9]. (In turn, the ORNL-RPI values are normalized to 127.9 b for the resonance integral from 0.45 to 10 eV. For the fission integral from 5.0 to 10.0 eV, the ORNL-RPI normalization is ~1% below that proposed by Deruytter and Wagemans [5] and by inference somewhat above that now adopted by Gwin et al.) The results of Perez et al. at the 150-m flight-path length were in turn normalized to the integral of the 40-m data over the 2- to 10-keV range. The measurements at 150 m have the advantage of a smooth incident flux because ORELA replaced aluminium with beryllium in the container for the water moderator surrounding the tantalum electron target.

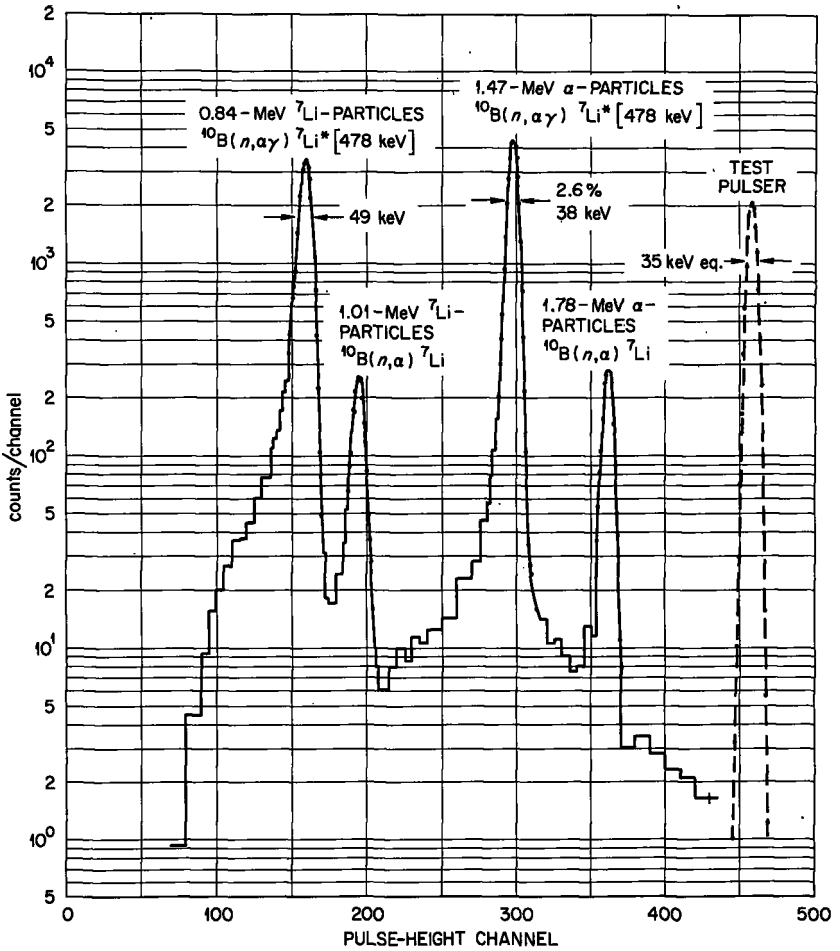

FIG.3. Pulse-height spectrum of the ^{10}B(n,α)^{7}Li reaction observed with the gridded ion chamber.

Figure 1 illustrates the unit-weight average cross-section results of both experiments on a segmented scale, along with some results of other well-known experiments. Table I contains the corresponding experimental results. It is seen that the two sets of ORELA results are almost parallel in the energy region shown, so that the main difference between them is a matter of normalization which is to some extent subject to review by data evaluators. Since the results of Gwin et al. [2] nearly agree with the ORNL-RPI data below 10 eV, one correctly concludes that the shape of the Gwin data between 20 and 100 eV does not quite agree with that given in the ORNL-RPI work. No explanation for this difference has been located after considerable effort, and indeed the corresponding data sets for ^{239}Pu(n, f) are in better agreement.

To check the question of low-energy shape for $^{235}U(n, f)$, Gwin and Weston prepared an independent test with a 20-m flight path using the fission chamber without the presence of the scintillator tank. The shape of the recent Gwin data appeared to be confirmed.

The uncertainties assigned to the average cross-sections do not include any component from uncertainty in the shape of the $^{10}B(n, \alpha)$ cross-section. Uncertainty in the efficiency of the boron chamber for recording a reaction is somewhat difficult to estimate, but pulse-height spectral measurements by Gwin imply an uncertainty for his data of about 4% at the highest energies. The plotted uncertainties do include observed fluctuations in results from one experimental run to the next, and uncertainties in effecting the stated normalizations. The data of Perez et al. above 10 keV have for the present an additional uncertainty assigned because these data will be subject to further analysis. If the two ORELA data sets were normalized together in the 7- to 11-eV region (by joining ORNL-RPI with the Perez et al. data), the two data sets would disagree by about 5% in the 1- to 10-keV region, not inconsistent with the estimated uncertainties.

To compare these measurements of $^{235}U(n, f)$ cross-sections with others using continuous sources, one may inspect the results of Blons [8] and of Lemley [9] which both agree with the results of Perez et al. in the 100- to 200-eV interval. Blons' measurements generally agree with those of Perez et al., but the data of Lemley (measured against $^{6}Li(n, \alpha)$) drop to lower values above a few hundred eV. For orientation, all the results fall below the 40-keV value of White [10] and the 24-keV value of Perkin et al. [11] and are nearer to the results by Szabo et al. given in 1971 [12].

Work on the $^{235}U(n, f)$ cross-section at ORNL is expected to continue in two forms. The measurements of Perez et al. will be refined and extended in support of further efforts to measure the ratio of capture in ^{238}U to fission in ^{235}U. Peelle and Weston are undertaking measurements pointed directly toward the $^{235}U(n, f)$ cross-section, but no data from this investigation have yet been analysed. The emphasis so far is on the use of 'thin' ionization chambers with which high efficiencies can be achieved in recording the few nuclear events taking place. At first we will be satisfied with normalizations at low energy in the style of the measurements described above. Figures 2 and 3 show pulse-height distributions observed for low-energy neutrons with the counters now ready for use. The Frisch-gridded boron chamber would probably be biased just above the group of pulses corresponding to the ^{7}Li particles from the ground-state reaction.

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DISCUSSION

E. MIGNECO: You have described measurements of the same quantity at the same laboratory which have been normalized in different ways. Presumably, there must be technical reasons for the different choices of normalization. I think it is very important that the experimentalists publish the reasons for their selection of a specific normalization so that their reasons are known to evaluators and users in other laboratories. Can you describe in greater detail the reasons for the various choices of normalization?

R.W. PEELLE: I also hope that the final reports will contain the reasons for selection of a particular normalization. The reason that different choices are made is fairly simple. Experimentalists do not all agree on the best way of doing things.

Gwin felt it was essential to normalize at thermal energies, and in order to do so he was willing to operate the accelerator at 10 pulses per second, which resulted in a very long, slow experiment.

De Saussure and Perez believed that there were already sufficiently many experiments in the energy region below a few hundred eV and that therefore it was not necessary for new experiments to extend all the way to thermal energy or even down to 10 eV. They therefore chose to operate the accelerator with a repetition rate which produced a higher counting rate and required less running time but which did not permit normalization at thermal energy.

This mode of operation required that the data be normalized at some energy. Perez and de Saussure selected the 100- to 200-eV range and normalized to the previous ORNL-RPI values of de Saussure et al. [7] because several different experiments were in accord in this energy range. Were it not for the agreement with other experiments, some other energy region would have been selected.

Below 100 keV the present data of Gwin differ in shape from other experiments. For instance, if the seemingly random part of the error is removed, these data differ by about 5% from the ORNL-RPI data of de Saussure et al. in the region between about 30 eV and about 100 eV. (The present measurement by de Saussure et al., which is described in this paper, does not cover this energy region.)

J.L. LEROY: In the parallel plate ionization chamber used to monitor the flux, how do you determine the transmission correction for boron self-shielding and how large is it?

R.W. PEELLE: Gwin gives an uncertainty owing to the transmission correction of about 1%, which represents a significant fraction of his normalization uncertainty. The transmission correction is one argument not to extend measurements to thermal energies because there the correction is several per cent while it is negligible above a few eV.

J.L. LEROY: One reason for the efficiency of a fission chamber with thick layers of fissile material to vary with neutron energy is that the angular distribution of fission fragments varies with energy. The angular distribution is known to vary in a stepwise manner whenever a new fission channel opens. Many of the angular distribution data are old. Does anyone know of new, complete results on the angular distribution of fission fragments from ^{235}U?

R.W. PEELLE: The thick layers of fissile material were used to permit simultaneous measurement of the capture and fission cross-sections and would not have been used in a simple measurement of the fission cross-section alone.

The thick layers are a technical weakness of the experiment. As I remember from the old data, it is fortuitous that the fission-fragment angular distribution changes much less rapidly with neutron energy for ^{235}U than for, say, ^{239}Pu.

In considering the effects of changes in the angular distributions of the fragments, the design of the fission chamber should also be considered. The spacing of the plates and the thickness of the foils bias against fragments emitted both perpendicular and parallel to the plates. Fragments emitted at intermediate angles have higher probability of detection. The coincidence requirement with the scintillator tank probably does not introduce much additional bias in favour of fragments emitted at intermediate angles. As far as I know, none of the experimenters at ORNL has made a detailed calculation of the counter's efficiency as a function of fragment angle. I do not think the calculation would be a simple one since multiple scattering and other effects would have to be considered.

W.P. POENITZ: Meadows made an approximate calculation in connection with measurements of the ^{238}U-capture-to-^{235}U-fission ratio. Around 2 MeV in either ^{238}U or ^{235}U, there is a big change in fragment angular distribution of the order of 1%. For 2π or 4π counting in a 200 $\mu g \cdot cm^{-2}$ foil, the resulting correction for absorption in the foil was of the order of 1%. In the energy range of the ORNL experiments the effect should be much smaller unless there are changes in angular distribution from resonance to resonance; I think there are no data about such effects.

B.D. KUZMINOV: Another effect which could influence the efficiency of the chamber is changes in energy of the fission fragments. However, Obninsk studies show that changes in fragment energy are not more than about 0.5% from thermal energy up to 5 MeV incident neutron energy. Such a change would not require a significant correction to the efficiency.

A.J. DERUYTTER: Perhaps it is easier to make corrections for a thick foil since a thin foil can be of very non-uniform thickness unless it is carefully made.

B.C. DIVEN: I should think that below 100 keV these effects which we have been discussing would not be very important compared to other uncertainties. However, it would be nice to have some quantitative information.

A.J. DERUYTTER: The new value of the 2200 m/s fission cross-section for ^{235}U will probably be somewhat higher than the value in ENDF/B-III. If the Gwin data were renormalized to the higher value, I think they might agree better with other data.

R.W. PEELLE: I agree that the Gwin data ought to be renormalized to take into account recent experiments and evaluations. The evaluator ought to select the best low-energy value for the renormalization, but whether the best value is the thermal value or the 10-eV value is open to discussion. I have already mentioned that normalization at 10 eV avoids the uncertainty in the correction for self-shielding in boron.

DISCREPANCIES OBSERVED IN FISSION CROSS-SECTION MEASUREMENTS USING FISSION FOILS FROM DIFFERENT ORIGINS

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Abstract

DISCREPANCIES OBSERVED IN FISSION CROSS-SECTION MEASUREMENTS USING FISSION FOILS FROM DIFFERENT ORIGINS.

During the past few years, the fission cross-sections of ^{235}U and ^{239}Pu have been measured below 1 MeV using a Van de Graaff accelerator and time-of-flight techniques. Measurements obtained with three fission chambers have been intercompared. One fission chamber containing a ^{235}U foil was constructed, calibrated and used previously by P.H. White. Two other thin-walled chambers, identical to each other, were equipped with ^{235}U and ^{239}Pu foils, respectively, prepared and assayed by the Central Bureau for Nuclear Measurements (CBNM). The count rates of the three chambers in a thermal neutron beam were also intercompared at CBNM. Since the cross-section data obtained using the thin-walled ^{235}U chamber were systematically low while ratio data were systematically high, a detailed examination of sources of the systematic error was begun. Loss of material from the ^{235}U foil prepared by CBNM has been confirmed by both 2π and high-geometry alpha counting but the times at which losses have occurred have not yet been established. The data for the ^{235}U fission cross-section based on the White chamber and reported at the Argonne Symposium in 1970 remain unchanged. The data for the same cross-section reported at the Knoxville Conference in 1971 must be corrected because of the loss of material from the CBNM foil and because of a new value for the half-life of ^{234}U.

1. INTRODUCTION

During the last few years, we measured the fission cross-sections of ^{235}U, ^{239}Pu and ^{241}Pu at neutron energies lower than 1 MeV. For ^{235}U, two sets of measurements were made, the first one (Ref.[1]) using a fission chamber, constructed and calibrated by P.H. White, and the second one (Ref.[2]) using our thin-walled chamber containing a ^{235}U foil from the Central Bureau for Nuclear Measurements (CBNM), Geel, Belgium. As the two fission chambers were used separately in measurements at different neutron energies, no significant difference could be established between the two sets of results obtained.

For the extended measurements in the 1- to 2-MeV region, we have used both the White chamber and our own one, symmetrically placed with regard to the incident proton beam, and the results so far indicate a difference between the two chambers varying from a few tenth per cent to 4%. The discrepancy did not exceed the sum of the errors. However, the fact that in most measurements our fission chamber gave lower fission cross-sections indicated the possibility of systematic errors. Thus a careful investigation of all possible sources of systematic errors was needed.

This paper discusses the various measurements made in order to find out the origin of the discrepancy. In section 2, the fission measurements are described in which a difference between the two chambers was observed.

Section 3 is devoted to an attempt to resolve the disagreement. Section 4 analyses the various sources of error and discusses some problems connected with the fission foils used.

2. DISCREPANCIES IN FISSION CROSS-SECTION MEASUREMENTS

2.1. Experimental procedure (Fig.1)

Neutrons were produced with the pulsed Van de Graaff accelerator via the $^{7}Li(p,n)^{7}Be$ and $T(p,n)^{3}He$ reactions. The two fission chambers were placed along two generators of a cone whose apex was at the target. The neutron flux was measured by a calibrated counter placed along a third generator of the same cone. The time-of-flight technique was used to determine the neutron background. The pulse-height and time-of-flight spectra of each fission chamber were simultaneously recorded and compared to the flux detector count rate. Thus, from a single experiment, absolute values of the fission cross-section and their ratio could be simultaneously deduced. Such an arrangement could also be used to compare two foils of the same isotope.

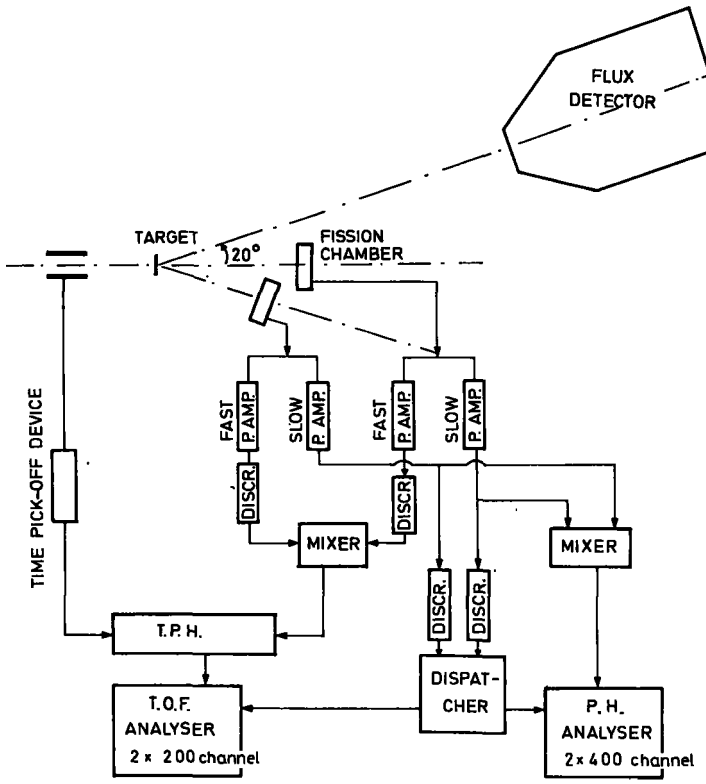

FIG. 1. Experimental arrangement.

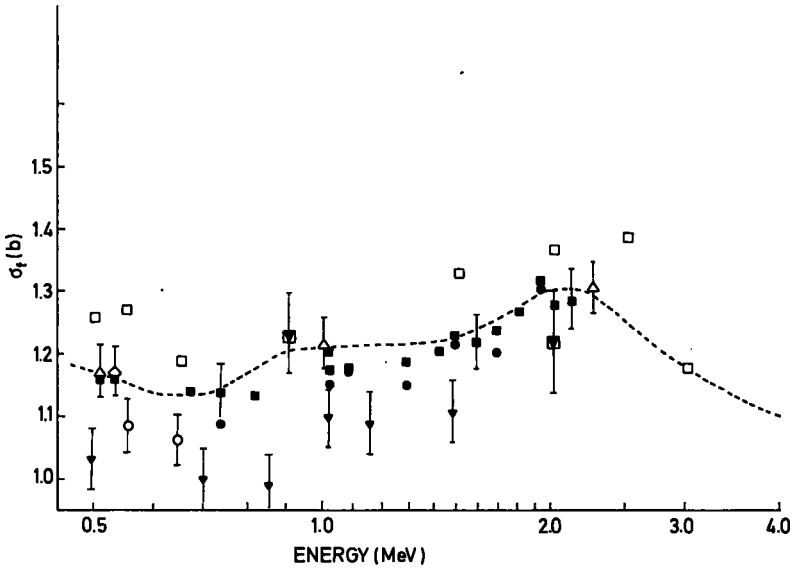

FIG. 2. Results of ^{235}U fission cross-section measurements.
----- Davey, △ White, ▼○ Poenitz, □ Allen, ▣ Dorofeev, ◇ Käppeler.
Present work: ■ White's foil, ● CBNM foil.

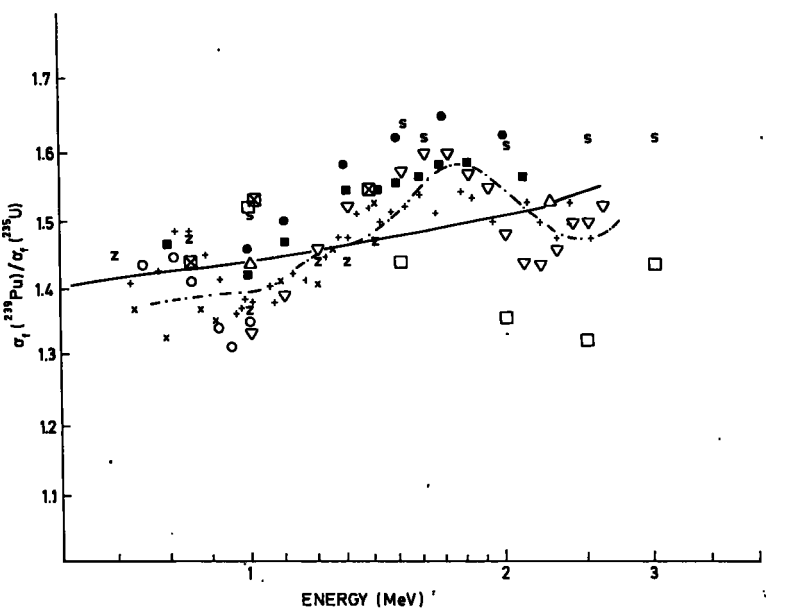

FIG. 3. ^{239}Pu and ^{235}U fission cross-section ratio.
S = Smith, ⊠ Henkel, ▽ Netter, + Smirenkin, × Dubrovina-Shigin, □ Allen-Ferguson, △ White, Z = Poenitz, ○ Pfletschinger et al. Present work: ■ White's ^{235}U foil, ● CBNM ^{235}U foil.

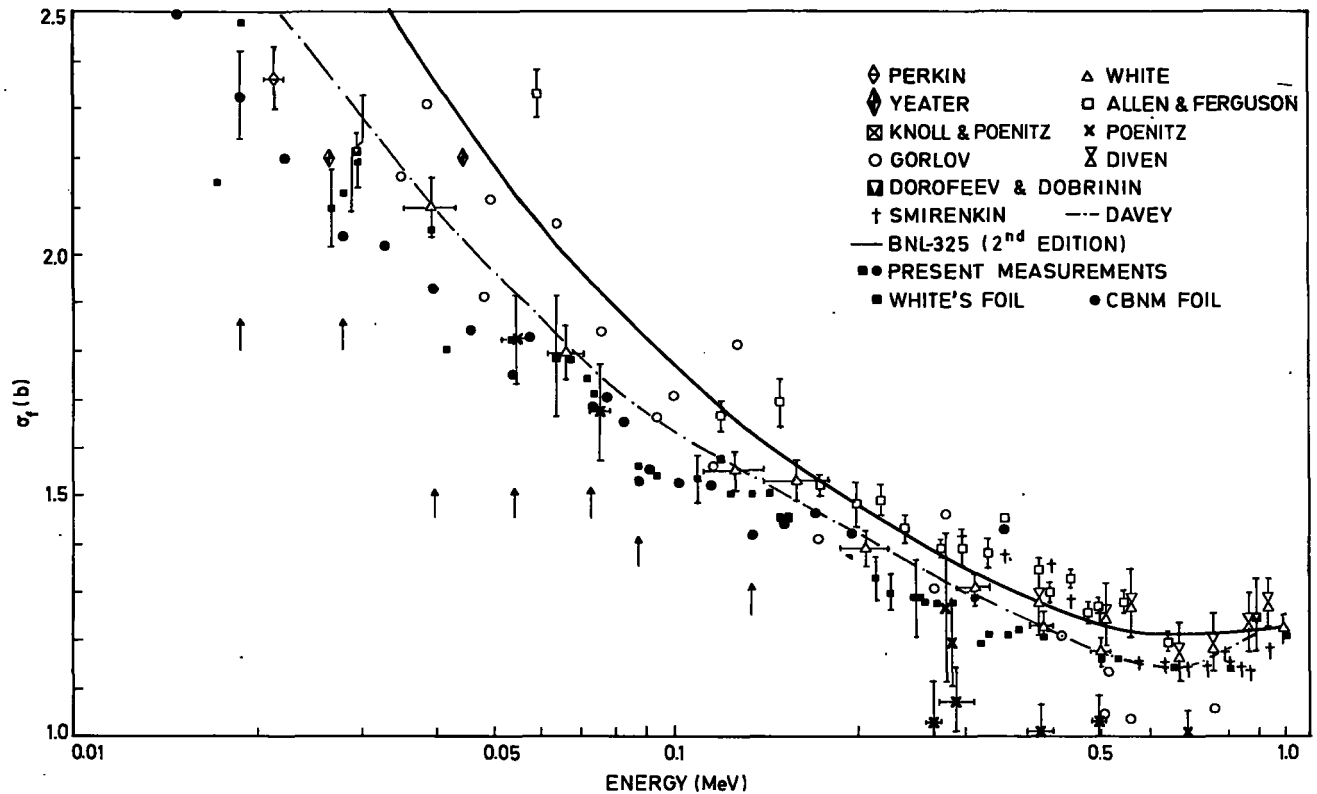

FIG. 4. Fission cross-sections of ^{235}U showing present measurements with the White and CBNM foils. Arrows indicate simultaneous measurements.

2.2. Measurements

During 1971, three sets of measurements were made, each of them involving the comparison of two of the following chambers:

(a) Chamber U I (containing a ^{235}U foil), described in Ref.[3], constructed and calibrated by P.H. White
(b) Chamber U II, the ^{235}U foil of which was prepared and assayed by CBNM (Ref.[2])
(c) Chamber Pu which is identical to U II but contains a ^{239}Pu foil from CBNM.

In February 1971, chambers U II and Pu were used to measure both the fission cross-section of ^{235}U and ^{239}Pu and their ratio at energies lower than 200 keV. Results of this first set of measurements are given in Ref.[2]. As mentioned in the Introduction, no noticeable difference was seen between the new σ_f values obtained with chamber U II and the older ones measured by means of chamber U I [1].

From May to August 1971, an intercomparison of the three chambers was made in the 1- to 2-MeV region. Discrepancies of a few per cent were observed for both $\sigma_f(^{235}U)$ (Fig.2) and the ratio $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ (Fig.3). In the whole energy range, chamber U II gave lower ^{235}U fission cross-sections and higher ratios. Systematic errors were suspected.

In December 1971, continuing the comparison of chambers U I and U II in the keV region where previously no difference could be detected since the measurements were not simultaneous, we observed here also discrepancies from 2 to 6% (Fig.4).

3. COMPARISON AT THERMAL ENERGY

Because of the discrepancies observed in the above measurements, the correctness of the ^{235}U fission cross-section measured seemed questionable. To check the reliability of the two chambers, we used them to measure the well-known ^{235}U fission cross-section and the $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ ratio at thermal energy. As these quantities are known with an accuracy better than 1%, the 'right' chamber would be the one which gives results closest to the recommended values of $\sigma_f(^{235}U)$ and $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ at thermal energy.

The measurements were done in May 1972 in collaboration with A.J. Deruytter from CBNM and included several comparisons:

(a) Chambers U I and U II
(b) Chambers U I and Pu
(c) Chambers U II and Pu
(d) Chamber U II and a boron chamber which should measure the absolute neutron flux and hence permit to deduce the absolute value of $\sigma_f(^{235}U)$ at thermal energy.

Unfortunately, comparison (d) had to be withdrawn because of the high sensitivity of our boron ionization chamber to γ-rays. The pulse-height spectrum was distorted in such a way that no accurate measurement of the absolute neutron flux could be made. Nevertheless, from comparisons (a), (b) and (c) it was possible to deduce the ratio of the count rates of chambers U I and U II and $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ ratios at thermal energy.

3.1. Experimental procedure

The thermal neutron beam was extracted from a tangential beam hole of the Belgian BR-2 reactor. The geometry of the beam tube and the slow chopper are described in detail in Ref.[4].

The two fission chambers to be compared were alternatively placed in the beam for a pre-determined number of counts of the beam monitor (^{235}U layer + solid-state detector outside the beam). The chambers were moved perpendicularly with regard to the beam axis by means of a chamber changer. The length of the flight path changes from 1607.1 ± 0.8 mm for thin-walled chambers to 1608 ± 0.8 mm for the White chamber.

The time-of-flight and pulse-height spectra were recorded simultaneously in two subgroups of a 1024-channel analyser. The pulses were counted simultaneously when the chamber was in the counting position. Measurements were made alternatively with and without a cadmium filter in the beam.

TABLE I. ^{235}U FISSION CROSS-SECTION – PRELIMINARY RESULTS

E_n (MeV)	$\sigma_f(^{235}U)$ White's chamber	$\sigma_f(^{235}U)$ CBNM foil	$\frac{\sigma_f(\text{CBNM})}{\sigma_f(\text{White})} - 1$ (%)
0.730	1.14	1.09	- 4.5
1.020	1.18	1.153	- 2.3
1.08	1.18	1.18	- 0.1
1.282	1.19	1.15	- 3.3
1.405	1.205		
1.484	1.24	1.241	+ 0.2
1.484	1.22	1.196	- 2
1.578	1.22		
1.680	1.24	1.20	- 3.3
1.797	1.27		
1.915	1.32	1.308	- 0.9
1.997	1.28	1.28	+ 0.1
2.1	1.284		
0.019	2.48	2.323	- 6.3
0.040	2.05	1.923	- 6.2
0.055	1.822	1.745	- 4.2
0.075	1.707	1.680	- 1.6
0.088	1.556	1.522	- 2.2
0.135	1.491	1.459	- 2.8

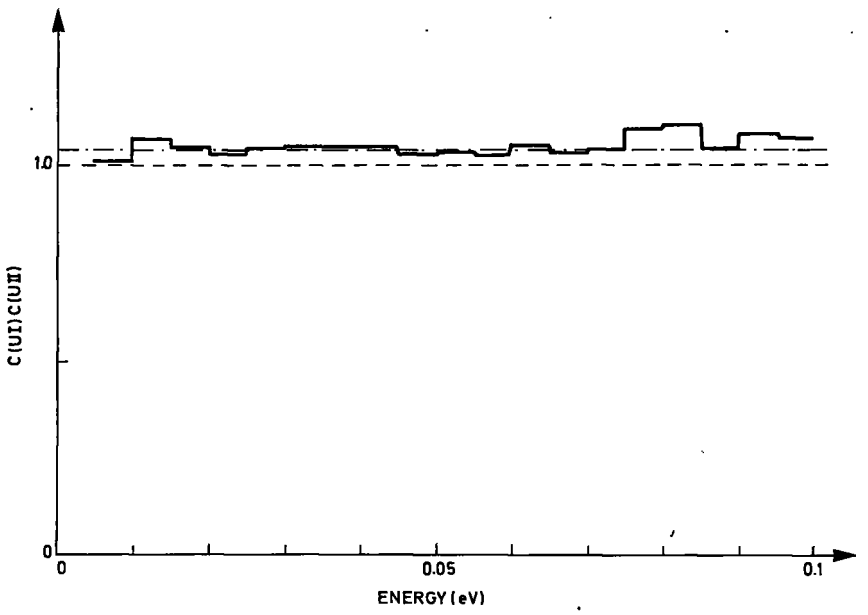

FIG. 5. Count-rate ratio of the ^{235}U fission chambers.

3.2. Data

Preliminary results of the ^{235}U fission cross-section measurements are given in Table I.

The count-rate ratio $C(U\ I)/C(U\ II)$ of chambers U I and U II from 0.01 to 0.1 eV is plotted in Fig.5. This ratio is normalized with regard to the contents of ^{235}U atoms of the two foils and must be in principle equal to unity. Table II summarizes the mean values of the count-rate ratio for different energy intervals. The deviation from unity is about 4.7%. As shown in Fig.6, the $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ ratios successively measured with chambers U I and U II differ by the same amount. The dashed line is obtained by dividing the upper curve (U II as ^{235}U standard) by the count-rate ratio C(U I)/C(U II) plotted in Fig.5. Thus the dashed line represents an indirect determination of the $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ ratio as referred to the White chamber.

TABLE II. COMPARISONS AT THERMAL ENERGY

Energy range (eV)	C(U I)/C(U II)	$\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ (U I)	$\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ (U II) [a]	$\frac{\sigma_f(^{239}Pu)/\sigma_f(^{235}U)(U\ II)}{\sigma_f(^{239}Pu)/\sigma_f(^{235}U)(U\ I)}$ [a]
0.02 - 0.03	1.041 ± 0.022	1.280 ± 0.029	1.349 ± 0.024	1.053 ± 0.030
0.01 - 0.06	1.048 ± 0.021	1.307 ± 0.024	1.386 ± 0.022	1.060 ± 0.026
0.005 - 0.1	1.047 ± 0.019	1.369 ± 0.023	1.442 ± 0.022	1.053 ± 0.025

[a] Calculated with original ^{235}U content.

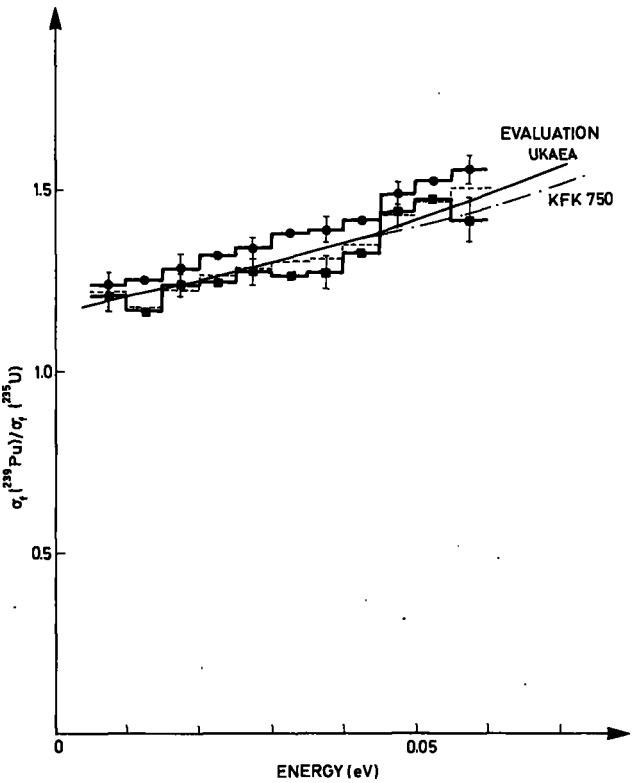

FIG. 6. Fission cross-section ratio $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$.
White's ^{235}U foil, CBNM ^{235}U foil.

The averaged discrepancy is somewhat greater at thermal energy than in the keV and MeV regions. There is good agreement between the fission cross-section ratio obtained by means of the White chamber and that measured by other authors, but no definitive conclusion can be made about the CBNM foil. The problem was becoming more and more complicated. At the end of the measurements at thermal energy it was decided, as a check, to re-measure the CBNM foil activity. The results deduced from low-geometry alpha counting differed by minus 5% from the original activity which was measured in January 1971 before the foil was sent to Cadarache. If the new value of the ^{235}U content is used for calculation, the count-rate ratio C(U I)/C(U II) becomes nearly equal to unity and the $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ ratios measured at thermal energy by means of both ^{235}U foils are in agreement. Thus the discrepancy seems to be resolved.

Although the appearance of the CBNM foil surface is good and does not show any obvious smear, the decrease in activity indicates an important loss of material. A 2π-geometry alpha counting was made with the foil removed from the ionization chamber. The residual alpha activity was about 0.3% of the total activity and it was proved that some loss of material had occurred.

TABLE III. ALPHA COUNTING

CBNM foil				White's foil			
Date	Method	Activity	Deviation from mean value (%)	Date	Method	Activity	Deviation from mean value (%)
10.1.1971	low geometry	1504.7	0	1969	2π	9470	+ 0.14
21.1.1971	2π	1509.5	- 0.43	1.9.1971	2π	9425	- 0.34
1.9.1971	2π	1516.9	+ 0.06	17.11.1971	2π	9460	+ 0.03
17.11.1971	2π	1519.5	+ 0.23	12.4.1972	2π	9476	+ 0.2
12.4.1972	2π	1517.9	+ 0.12	14.10.1972	2π	9455	- 0.02
14.5.1972	low geometry	1428.5	- 5.1 [a]	Mean	2π	9457	
14.10.1972	2π	1467.0	- 3.23				
Mean (2π) up to 4.1972		1516.0					

[a] Deviation from original low-geometry assay.

It is important to find out when and how the fissile material has been lost. Table III gives the various 2π alpha countings that were made in the course of the experiments, at the beginning of a set of fission cross-section measurements. The foil activity remained constant from January 1971 to April 1972. The alpha measurement in April 1972 was made just before the chambers were sent to CBNM for thermal energy measurements. The low-geometry alpha assay of May 1972 revealed a decrease of activity of 5%. It seemed that material was lost during package manipulations and transport from Cadarache to CBNM. Under these circumstances, we can only rest assured that all the fission cross-section measurements described in section 2 were made with the same amount of ^{235}U atoms in the CBNM foils. However, some loss might have occurred also during the first transport when the foil was delivered to us. The absolute activity of the foil, as measured in various 2π-geometry alpha assays, indicates that up to April 1972 the original ^{235}U content was still on the foil. But some doubt about this remains since 2π-geometry assay is less accurate than low-geometry assay. We plan to calibrate our 2π-assay method with regard to the low-geometry alpha counting used at CBNM.

Finally, the measurements at thermal energy indicate a good agreement between the two ^{235}U foils if we take into account a loss of 5% of ^{235}U atoms for the CBNM foil, as measured by low-geometry counting. For the fission cross-section ratio $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$, both chambers give values that agree with the evaluated data. Unfortunately, the loss of material complicates the problem and makes the renormalization of measurements at high energy more difficult.

4. DISCUSSION

Besides the neutron flux measurement, problems encountered in most fission cross-section measurements are mainly related to the absolute determination of the number of fissile atoms in the layers, the efficiency of the detector used and corrections which must take into account some parasitic effects such as neutron scattering. A general and complete survey of these problems is outside the scope of this paper. For a recent survey see Ref.[5]. Our examination is more restricted and concerns mainly the ^{235}U foils used in our measurements. With respect to the ^{235}U content of the foils, great care was taken in order to minimize systematic errors. Both ^{235}U foils were assayed by at least two independent methods [1-3]. However, despite the good agreement between the various methods used at each laboratory, there is still the possibility of systematic differences between two or more laboratories. A few years ago, it was recommended to implement a foil exchange programme and an intercomparison of the assaying methods used at different laboratories. This was first done by CBNM in collaboration with Chalk River Laboratory and Idaho Nuclear Corporation, as reported by Lauer [6].

It seems advisable that laboratories, which are not mainly engaged in the assay of fission foils but use them for cross-section measurements, also participate in the exchange and intercomparison programme. A second step, including foil users, has been initiated by CBNM.

Fission cross-section experimenters have to check the fissile quantity before, after and even during the measurements. For this purpose we used 2π-geometry alpha counting. The accuracy of this method is sufficient to control the reproducibility of the foil activity and hence to detect a significant loss of material. The method seems to us very convenient since alpha counting can be made without removing the foil from the ionization chamber where it is placed for fission cross-section measurements. However, this method is not suitable for high-alpha-activity foils. Also, a more accurate method is needed for determining the absolute amount of fissile atoms at delivery of the foil. Low-geometry alpha counting is the most suitable method for this purpose. When we received the CBNM foil, an attempt was made with our low-geometry alpha counting set-up. As the geometry factor (10^{-4}) was too small and the foil activity was low, this attempt suffered from very poor statistics. As there was no reason for suspecting at that moment a loss of material, this attempt was not continued. With regard to problems associated with handling, it must be noticed that the White foil was never removed from its chamber while the CBNM foil was taken out of its chamber several times before it was packed and sent to CBNM. The two ionization chambers differ also in that there is no gas circulation in the White chamber, which only had to be refilled from time to time, while the gas was continuously circulated inside the thin-walled chamber. So it was not surprising that particles were lost from the chamber, and this could explain the fact that the residual contamination of the chamber was only 0.3% instead of the 5% which were actually lost.

There are several problems in connection with the efficiency of the ionization chambers. The absorption of a small part of the fission fragments in the foils is closely connected with the inhomogeneity of the layer and hence with the preparation technique. The White ^{235}U foil was painted whereas the CBNM foil was electrosprayed. Although the foils are of the same thick-

ness (0.5 mg/cm^2), the foil absorption corrections differ by nearly 2%. The correction factor for the White chamber given by the author himself is 1.059. For the CBNM foil, the correction factor of 1.042 was deduced from measurements of White on electrosprayed foils [7]. The absorption correction for the specific White foil used in our experiments is somewhat greater than the value of 5% which can be deduced from the slope of the curve plotted in Fig.2 of Ref.[7].

In the literature, absorption correction data are not very numerous, and further work on this subject is needed. For the absolute counting of fission events, an extrapolation of fission pulse-height spectra to zero bias is also necessary. The extrapolation procedure is not unique but varies with the experimenter. For the two ^{235}U foils which we used, the extrapolation was made in the same way as described in Ref.[3]. The extrapolation problem is severe and is one of the main criticisms of 2π-geometry fission counting. At the present state of knowledge of foil absorption and extrapolation to zero bias, a 1% error on the detection efficiency is not so pessimistic for foils with a thickness of a few hundred μg/cm^2.

The neutron scattering correction was calculated with the Monte-Carlo method. The results for the White chamber were verified. In Ref.[3], neutron scattering in the chamber structure was analytically calculated and checked by complementary experiments. The values given there were compared with the results of our Monte-Carlo calculation at the same energy. In most cases, the difference is less than 1%.

5. CONCLUSION

As mentioned in section 3, the fission cross-section ratio of ^{239}Pu and ^{235}U measured at thermal energy with the White chamber is in good agreement with the recommended value. Thus, the results obtained for $\sigma_f(^{235}U)$ with the White chamber, which were published in Ref.[1], need not be changed. As regards the CBNM foil, the detected loss of fissile material makes the results published in Ref.[2] questionable. However, it may be possible to correct these measurements, and complementary experiments are planned for this purpose. As shown in Table III, the loss of fissile material as measured by low-geometry alpha counting is 5% while the relative variation of the alpha activity as indicated by 2π-geometry alpha counting is 3%. It can be assumed that the fissile material was lost in two parts, i.e. 2% during manipulation and transport from CBNM to Cadarache and 3% during the return to CBNM. This can be checked by calibrating our 2π-geometry alpha counting with respect to low-geometry alpha counting of CBNM. In case the above assumption is confirmed, the data of Ref.[2] have to be raised by 4% (2% because of the new half-life value of ^{234}U (CBNM) and 2% for material loss before the measurements).

ACKNOWLEDGEMENTS

We are grateful to M.M. Neve de Mevergnies and A.H.W. Aten for welcoming us at BR-2 and CBNM. The measurements at BR-2 were done in close collaboration with A.J. Deruytter. We are indebted to W. Becker

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DISCUSSION

L. STEWART: Do the data in Table I replace values in the previous literature?

J.L. LEROY: No. The preliminary values in Table I serve only to illustrate the difference between the two chambers. We prefer not to publish more values until the matter of loss of material has been finally resolved.

L. STEWART: Are the values which you published previously now in question?

J.L. LEROY: The measurements presented at the 1970 Argonne Symposium were made with White's chamber and foil and are not in question regarding the loss-of-material problem. The measurements presented at the 1971 Knoxville Conference will very probably have to be raised by 4%, i.e. 2% because of loss of material, but this figure must still be confirmed, and 2% because the ^{234}U half-life value used was not the best one available.

F. KÄPPELER: I was told by experimenters from Geel that destructive analysis by the isotopic dilution technique can determine the content of fissile samples to an accuracy of 0.5%. Why was this method not used to resolve the problems with the Cadarache CBNM foils?

J.L. LEROY: We have only one foil, and we do not want to destroy it before all other possible explanations of the discrepancy have been checked. I believe the last thing which remains to be checked is the 2π geometry, in which an error of 2% would explain all the experimental observations.

T.A. BYER: In the measurement of the ratio $^{239}Pu/^{235}U$, the correction for loss of material from the uranium foil will effectively increase the ^{235}U cross-section. This will bring the ratio of fission cross-sections for $^{239}Pu/^{235}U$ into much better agreement with the data of Pfletschinger and Käppeler (Nucl. Sci. Eng. 40 (1970) 375). Have you also checked for similar losses in the plutonium foil?

J.L. LEROY: Our low-geometry alpha-counting results, which are much easier obtained than the results for ^{235}U because of the higher activity, have always been in excellent agreement with the original count rates determined at CBNM, so we had no reason to suspect a change in the plutonium foil.

A.J. DERUYTTER: A change of this magnitude in a uranium foil is very unusual. We have exchanged foils with Idaho Falls, Chalk River and Aldermaston and have observed some very small effects but never anything of this size.

J.L. LEROY: I believe the high loss of fission material was due to an accident. Even though the probability of such accidents is very low, we shall check for them routinely in the future.

A.J. DERUYTTER: I have two recommendations for experimentalists who order precision foils from standards laboratories such as CBNM, Geel.

(1) Order many foils rather than just one. Several foils should be retained by the standards laboratory and checked periodically. At the experimentalist's laboratory, for example in a fission experiment, one foil should be used in the fission chamber and two others should be kept for regular comparison with the foil used in the chamber. Both the standards laboratory and the experimental laboratory then have a complete history of intercomparisons among the various foils, and these records can be extremely useful in tracing possible errors during the course of an experiment.

(2) In the case of ^{235}U-foils, high enrichments may not always be desirable. In a sample enriched to 99.9% in ^{235}U, the ^{234}U content is less than 0.1%. The accuracy of alpha counting in 2π geometry will be greatly reduced because of the low specific activity, which is due to ^{234}U. Many measurements at neutron energies below the fission threshold of ^{234}U can tolerate 1% of ^{234}U in the sample. Such a ^{234}U content will greatly improve the accuracy of both 2π and low-geometry alpha counting, which are so useful in assaying, intercomparing and checking experimental samples.

A.J. DERUYTTER: Without opening White's fission chamber the foil can only be examined in 2π geometry, no matter what technique is used. Would it be worth while to open the chamber and to examine the foil by, for example, low-geometry alpha counting?

J.L. LEROY: We have examined the White chamber in a thermal neutron flux, which I think may be a better technique than alpha counting. As Szabo has explained in a previous paper, a correction is required for loss of fission fragments in the foil. This correction depends on how the foil was constructed, for example whether it was painted or electrosprayed. The comparison of foils in a thermal flux takes all these corrections into account, and therefore I think it may be the better method.

MEASUREMENTS OF THE ^{235}U FISSION CROSS-SECTION IN THE FAST NEUTRON ENERGY RANGE *

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Abstract

MEASUREMENTS OF THE ^{235}U FISSION CROSS-SECTION IN THE FAST NEUTRON ENERGY RANGE.

The fission cross-section of ^{235}U was measured in the energy range from 35 to 3500 keV. The shape of the cross-section was measured using the Grey Neutron Detector with different moderators. The shape curve was normalized with three absolutely measured values, one using the associated-activity method, the second using the neutron flux integration technique, and the third using the Black Neutron Detector for absolute neutron flux counting. The present results are subject to final fail assay and will be supplemented by additional measurements.

1. INTRODUCTION

The fission cross-section of ^{235}U has a unique importance for reactor evaluations and cross-section measurements alike. Uranium-235 is a major fuel material for fast reactors. Most other fission cross-sections as well as some capture cross-sections are measured relative to that of ^{235}U in the fast energy range.

However, existing data for ^{235}U fission differ by more than a factor of two, and recent absolute measurements show differences in the order of 20%. A general downward trend exists in the values of measurements reported between 1940 and the present [1]. A comparison of absolutely measured capture cross-sections of ^{238}U and gold with those measured relative to ^{235}U leads to the conclusion of possible lower fission cross-section values of ^{235}U [2]. This was supported by preliminary measurements using roughly the same experimental method as applied to the absolute capture cross-section measurements [3].

Because of the importance of the ^{235}U fission cross-section, a programme was initiated to measure this cross-section by several independent techniques. The results were planned to be independent not only of the flux determination but of experimental techniques and fissile-mass determinations as well. The results presented here represent a portion of this effort. The fissile masses were determined, with the exception of spherical counters, only by alpha counting.

* Work performed under the auspices of the US Atomic Energy Commission.

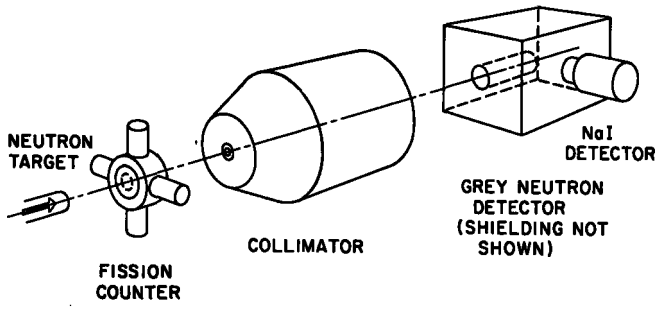

FIG. 1. Schematic set-up for shape measurements.

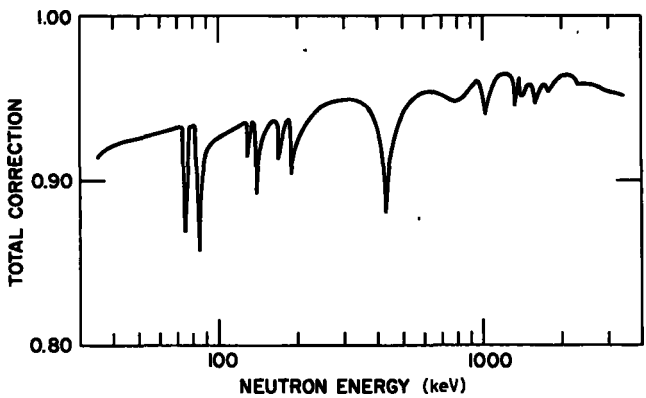

FIG. 2. Correction for shape measurements.

2. SHAPE MEASUREMENTS

A cross-section is characterized by its shape and absolute amplitude. In most experiments some systematic uncertainties apply to all values (for example, mass assignment). Thus, separating the shape measurements from the absolute measurement is well justified. In addition, this separation may result in more favourable experiment designs for a shape measurement if the absolute value is not required and vice versa. This approach, previously applied to capture cross-section measurements [2, 4], was followed here. However, an open geometry was used for the fission counter in order to overcome the tremendous count-rate problem in a closed geometry. The set-up is shown in Fig. 1.

The ^{7}Li(p, n) reaction was used as a neutron source. Measurements were carried out at angles of 0° and 60° to the incoming neutron beam. Target thicknesses ranged from 3 keV in the low-energy range to 70 keV in the high-energy range. The primary proton beam was pulsed and bunched to about 1-2 ns width and with a repetition rate of 2 MHz.

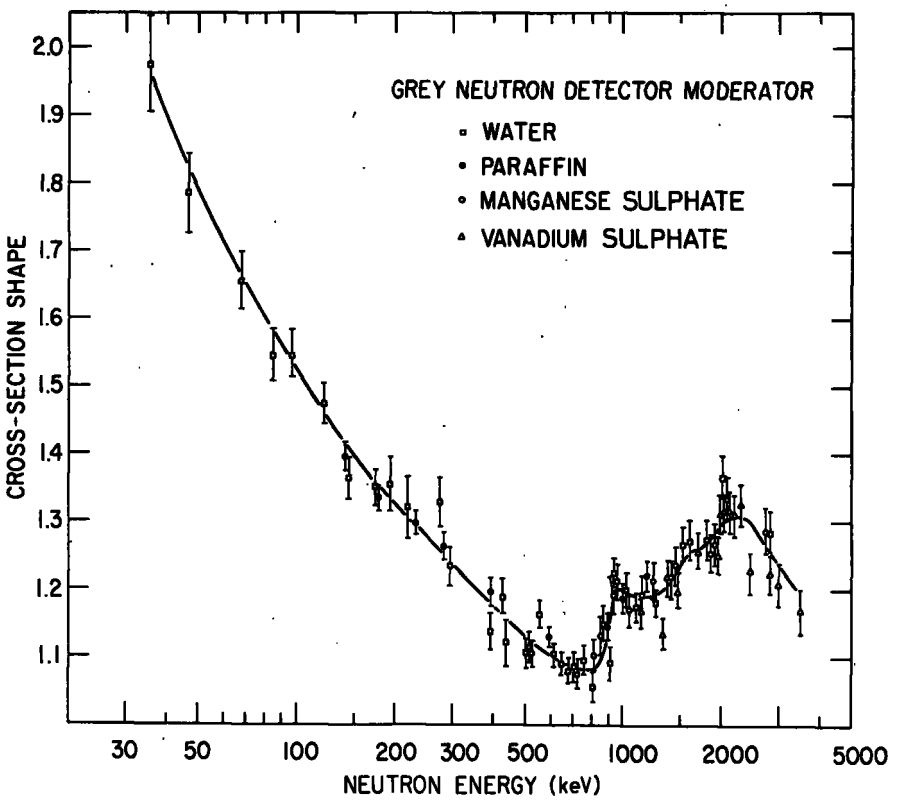

FIG. 3. Results of ^{235}U shape measurements.

The fission counter was designed for obtaining a low scattering background. The detector was a gas scintillation chamber of cylindrical shape with a diameter of 22 cm and a height of 16 cm. The scintillation light was viewed by four photomultipliers. The signals were added in pairs and a twofold coincidence was required. The fissile deposit had a diameter of 5 cm and was electroplated on a 0.013-cm-thick molybdenum backing. The uranium consisted of 99.85% ^{235}U, 0.054% ^{238}U, 0.027% ^{234}U and 0.062% ^{236}U. A thickness of 400 $\mu g/cm^2$ was used. The fissile deposit was located on one side of the detector, between 10-25 cm away from the target, and perpendicular to the incident neutron beam.

The Grey Neutron Detector described elsewhere (paper IAEA-PL-246-2/12 in these Proceedings) was used as a neutron detector. $MnSO_4 \cdot H_2O$, $VOSO_4 \cdot H_2O$ and water moderators were used in the detector with an effective radius of 46 cm. Utilizing the different capture γ-ray spectra allows a check on the evaluated efficiencies. In addition, a 28-cm effective radius detector of paraffine was used in part of the energy range.

Corrections were applied for elastic and inelastic scattering in the molybdenum backing (<1%), transmission through air (usually < 5%, but

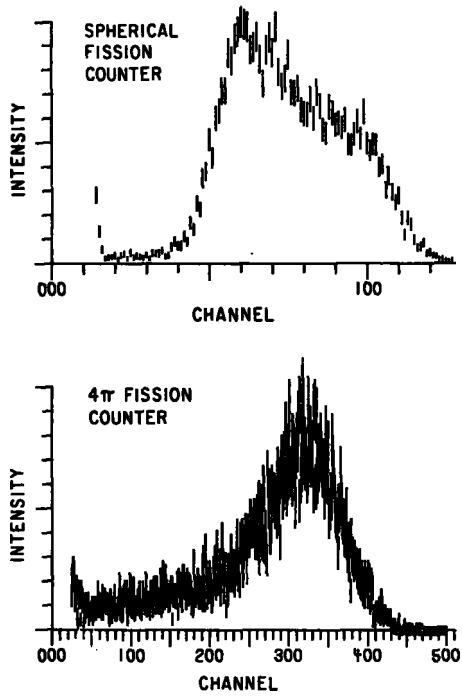

FIG. 4. Fission fragment energy spectra for spherical ionization chamber and 4π gas scintillation counter.

about 9% in the oxygen and nitrogen resonances around 440 keV), the second neutron group of the $^{7}Li(p,n)$ reaction (< 1%), transmission through the fission counter wall and backscatter from the wall and multipliers (usually about 2%, but larger in some strong iron resonances), and change of the fission counting efficiency with energy (< 1%). Figure 2 shows the total correction applied as a function of energy.

The background for the Grey Neutron Detector was determined by runs with a closed collimator channel. Most background from the fission detector was eliminated by the time-of-flight technique. The time-of-flight spectrum from the fission counter was stored with an on-line computer. The energy spectra from selected time-of-flight intervals were also stored. The γ-spectra from the Grey Neutron Detector were stored in the on-line computer via the same data terminal in order to avoid dead-time effects.

The results from the shape measurements are shown in Fig. 3. The contributions from statistical uncertainties to the total uncertainties of the values shown in the figure are dominating in the energy range below 1.5 MeV (1.5 - 2.5%). At higher energies, the uncertainty from the detector efficiency is an increasing factor and dominates at 3.5 MeV (1.5-2.8%). Systematic differences between different sets of measurements and reproducibility are within the statistical uncertainties, as can be seen in Fig. 3. An eye-guide curve was drawn through the experimental points.

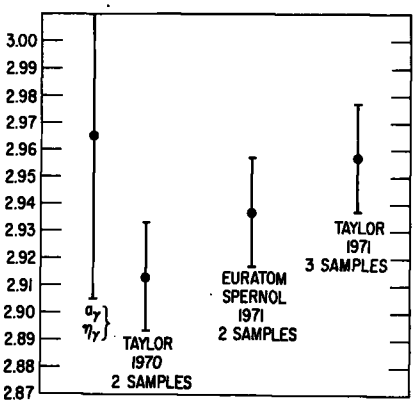

FIG. 5. Calibration factors for NaI detector.

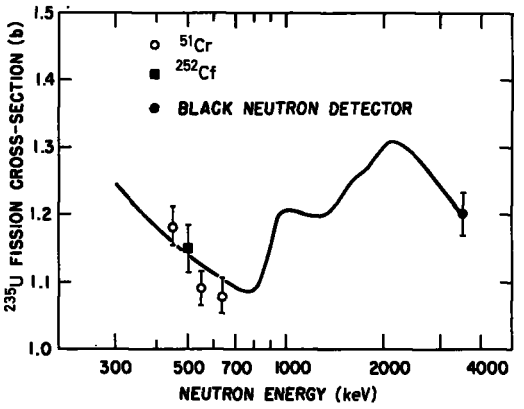

FIG. 6. Absolute values obtained for the ^{235}U fission cross-section.

3. ABSOLUTE MEASUREMENTS

3.1. Associated activity

This type of measurement has been described previously [5]. A spherical fission counter surrounds a vanadium target. The $^{51}V(p,n)^{51}Cr$ reaction is used as a neutron source. The associated ^{51}Cr activity is utilized to determine the absolute neutron flux. Additional measurements were carried out to improve the uncertainty from the mass assignment which was previously based on one sphere only. Two new spheres were used and the fissile material determined by destructive analysis. Fission spectra obtained with these fission counters are shown in Fig. 4. In addition, new absolutely calibrated ^{51}Cr samples were obtained from Euratom and

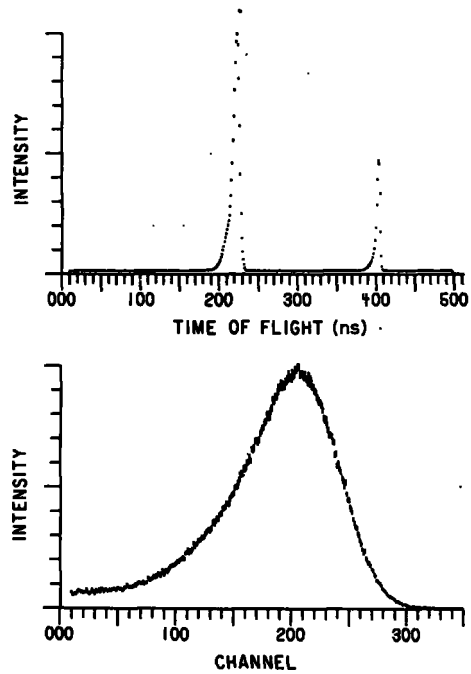

FIG. 7. Time-of-flight and energy spectra of the Black Neutron Detector at 3.5 MeV.

from Chalk River Nuclear Laboratories. The calibration values are shown in Fig. 5. The new average value for this efficiency is approximately that obtained with the Euratom samples and caused the major change of the previous values. A previous analytical estimate of the major correction, i. e. the scattering of neutrons in the inner shell, was replaced by a Monte-Carlo evaluation. The change was about 12% of the original correction and affected the result only by 0.3%. The results for the fission cross-section of ^{235}U are shown in Fig. 6. All values are averages of two or three measurements.

3. 2. Black Neutron Detector

Absolute cross-section measurements using a Black Neutron Detector (BND) are most favourable in the 2.5- to 5.0-MeV energy range. Below 1.0 MeV the cut-off energy influences the efficiency and may cause some systematic uncertainty at higher energies. Above 5.0 MeV the efficiency drops below 95% and thus the uncertainty of the evaluated efficiency increases proportionately.

The major problem for an absolute fission cross-section measurement using the Black Neutron Detector is the incompatibility of the count rates obtained from a fission counter with that of the BND. The BND has an efficiency close to one, and a count rate of 1000 counts/s is reasonable for the electronic and computer equipment. The 1000 counts/s correspond to 1000 n/s in a collimated beam. A 500 $\mu g/cm^2$ uranium foil would yield

about six fissions per hour in this beam. To reconcile both efficiencies, a double collimator system was used. The measurement was carried out at 3.5 MeV.

A neutron beam was first collimated such that the entire deposit of a 4π ^{235}U sample positioned in a gas scintillation counter at a flight path of 173.4 cm was radiated, but not the structural material of the scintillator chamber. A second collimator provided a lower-intensity beam for the Black Neutron Detector. The fission counter was a 4π gas-scintillation chamber. A 400-$\mu g/cm^2$, 5-cm-dia. fissile deposit on Vyns-backing was supplied by the Central Bureau for Nuclear Measurements, Euratom. The amount of fissile material was determined by α-counting in a low-geometry α-counter. A destructive analysis will be obtained at a later date. The fission spectrum obtained in this measurement is shown in Fig. 4.

The neutron time-of-flight spectrum and the energy spectrum of the BND are shown in Fig. 7. The energy spectrum shown is that obtained in the neutron time-of-flight peak after subtracting the energy spectra from an equally wide time-of-flight range adjacent to the neutron peak.

Corrections were applied for the transmission through the collimator and scattering from the collimator (0.5%), the efficiency of the fission counter (4.2%), the attenuation in air (3.2%), the less-than-one efficiency of the neutron counter (3.3%), and the second neutron group (0.4%). The result for the absolute fission cross-section of ^{235}U at 3.5 MeV is shown in Fig. 6.

3.3. Calibrated vanadium bath

A third absolute measurement was carried out utilizing a well calibrated ^{252}Cf source [6]. A collimated neutron beam of 500 keV passes through a gas scintillation counter and is captured in the Grey Neutron Detector with a vanadium-sulphate solution. The 2.25-MeV γ-ray from the neutron capture in hydrogen is used as a neutron monitor. The same fission counter and 4π foil as described in section 3.2 were used.

The counting efficiency was determined by radiating the set-up for 10 min with a 500-keV neutron beam and measuring for two 10-min periods the 1.4-MeV γ-ray from the decay of ^{52}V, then repeating the same procedure with the ^{252}Cf source.

Corrections for leakage from the vanadium bath (0.7% and 1.4%) and the absorption in the channel material (0.2%) were made in addition to the corrections mentioned in section 3.2. The value obtained for the ^{235}U fission cross-section is also shown in Fig. 6.

4. RESULTS AND DISCUSSIONS

The eye-guide curve shown in Fig. 3 was adjusted with the three absolute measurements shown in Fig. 6 by minimizing the average deviations. The resulting curve is also shown in Fig. 6. This curve has an uncertainty of about 3.5%, combined from about 1-2% statistical uncertainty and reproducibility and 2.5% from the absolute measurements. Above 950 keV the agreement of the present results with measurements by White [7], Szabo et al. [8], and Gilliam and Knoll [9] is good. At lower energies the agreement is best with the measurements by Szabo et al. [8] after normalizing these data with

values which they obtained with a fission chamber independent from that of White. The present results are essentially lower than previous evaluation results (BNL-325, ENDF/B-II, KFK-120), and partially lower than the more recent evaluation of ENDF/B-III. Final normalized values will be published after obtaining destructive analyses of the uranium samples and completing the analyses of additional data measured relative to $^{6}Li(n,\alpha)$ and $Au(n,\gamma)$.

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DISCUSSION

Supersession of previous results

R.W. PEELLE: Do the results which you have presented here replace the preliminary values obtained with the Grey Neutron Detector four years ago?

W.P. POENITZ: Yes. The present data replace those of 1968[1], which were marked preliminary.

The data from 1970[2] have also been modified but not directly replaced. We obtained new calibrated γ-ray sources which resulted in a 1% correction to the 1970 data, but additional new measurements have been combined with the older measurements. All other changes were minor compared to 1%.

Correction for fission fragments absorbed in the foil

A.J. DERUYTTER: In the 4π geometry fission pulse-height spectrum which you showed, how large was your correction for lost fission events?

W.P. POENITZ: Basically we have used the experimental data of White. There also exist several evaluations of this effect for both 4π and 2π foils. After studying the differences in the evaluations for 4π and 2π foils,

[1] POENITZ, W.P., Neutron Cross Sections and Technology (Proc. Conf. Washington, D.C., 1968), USAEC (1968) 503.
[2] POENITZ, W.P., Neutron Standards and Flux Normalization (Proc. Symp. Argonne, 1970), CONF-701002, AEC Symp. Ser. 23, USAEC, Oak Ridge (1971) 281.

we decided to reduce White's correction by something like 20-30% for the 400 $\mu g \cdot cm^{-2}$ foil. The main difficulty was to adjust the correction for the effect of à Vyns versus an aluminium backing.

The 4π counter gives a much nicer spectrum than the 2π counter because the total energy deposited by the fragments can be added together; however, the total absorption cannot be determined so well.

F. KÄPPELER: We attempted to determine directly the fractions of fission fragments which were absorbed in 2π- and 4π-counting arrangements. A small amount of ^{252}Cf was mixed with the fissile sample. The absolute activity of the ^{252}Cf could be determined very accurately by low-geometry counting so that we could compare the count rate obtained in 2π geometry with that obtained in low geometry and thus determine the absorption losses.

A.J. DERUYTTER: The method requires that the ^{252}Cf be distributed homogeneously within the target, and I do not know whether this problem has been solved as yet.

J.L. LEROY: The idea is interesting, but it has the disadvantage that a fission counting background is introduced where the real fission count rate is already very low.

F. KÄPPELER: The background would not be disturbing. The ^{252}Cf concentration can be kept very low, and in a measurement with a pulsed neutron source the background will be time-independent so that it can be conveniently subtracted.

Double collimation and foil uniformity

C.D. BOWMAN: In the absolute measurement with the Black Neutron Detector, not all of the neutrons which pass through the fissile deposit reach the flux monitor because this is the purpose of the collimator between the fission detector and the flux monitor. Have you verified that there are no problems owing to variation in the uniformity of the deposit since the flux monitor does not view the entire foil?

W.P. POENITZ: The first collimator located between the neutron source and the fissile sample produces a beam which is slightly larger than the 5-cm-diameter fissile deposit and at the same time prevents neutrons from being scattered from the photomultipliers and the heavier structural parts of the fission chamber. Since transmission is nearly 100%, normal variations in the sample thickness are unimportant. At a flight path of 1.8 m the neutron beam from the $^{7}Li(p,n)$ source was assumed to be uniform. Of course the first collimator between the neutron source and the fission chamber must not be so close to the source that the beam becomes non-uniform. If the beam is indeed uniform after it passes through the fission chamber, the second collimator between the fission chamber and the flux monitor should cause no problems.

The 4π fission chamber

C.D. BOWMAN: Would you explain how you obtained from the 4π fission chamber the pulse-height distribution shown in Fig. 4? I am particularly interested in how you eliminated noise from α-particles, etc., without eliminating real fission events at the same time.

W. P. POENITZ: The signals from the four photomultipliers which viewed the gas scintillator fission chamber were added together using various coincidence requirements, which did not have very great effect.

We always recorded two-dimensional (time-of-flight and pulse-height) spectra. Figure 4 shows an energy spectrum at the time-of-flight peak, which is quite sharp because the time resolution of the gas scintillator is about 3 ns. The α-peak is not covered by the energy scale of the figure.

A background spectrum determined from an adjacent time range was subtracted from the spectrum in the time-of-flight peak. This eliminated noise as well as the time-independent part of the α-background. Another background spectrum determined from measurements made with the fissile foil removed from the fission chamber was subtracted to eliminate background from other sources.

There is a small α-background which appears in the time-of-flight peak and which is due to (n,α) and (n,p) reactions in the argon and nitrogen gas used to fill the fission chamber. The size of this background is sensitive to the bias settings on the fission chamber. We have observed this peak in measurements without the fissile foil, and I have attempted to ensure that it was properly subtracted from the fission cross-section measurements.

Shape measurements with the large fission chamber and the Grey Neutron Detector

E. FORT: In many of your experiments you measured the neutron flux after transmission through the fissile sample. You must therefore know accurately the transmission through the fission detector. At Cadarache, we used symmetric angles from the beam so that we did not have to worry about transmission through our fission detector.

W. P. POENITZ: In the fission measurements the transmission corrections were not large because the foils were thin. Only the walls of the chamber are of concern. In the fission detector used with the Grey Neutron Detector, one of the walls was unfortunately not so thin as would have been possible, and therefore the correction for transmission was larger than necessary. Even so, this correction was only of the order of 3% over most of the energy range.

The fission chamber used in the shape measurement was made as large as possible in order to obtain a low scattering background. We thought it would be simpler to evaluate the transmission correction than to correct for scattering from the irregular geometry of the detector.

The transmission correction includes corrections for transmission through air and through the back wall of the detector and for scattering in the sample backing. In the shape measurements, molybdenum foil was used as backing because it has a smooth cross-section without resonance structure. Most of the structure in the correction curve shown in Fig. 2, such as the resonance at 430 keV, is due to transmission through air. Only a few of our measurements fall within the energy range of one of these resonances. The calculated corrections are based on reasonably well-known cross-sections.

The neutron source reaction: $^{7}Li(p,n)$ versus $T(p,n)$

M. S. COATES: You have used the $^{7}Li(p,n)$ reaction as a neutron source throughout the energy range of your measurements. Did you have problems with multiple neutron groups?

W. P. POENITZ: We corrected for the second neutron group. The ratio of the second group to the primary group is rather well known. I have also measured it using the Black Neutron Detector and time-of-flight up to an energy where I could no longer resolve the time-of-flight peaks.

The second neutron group contributes a maximum of about 10% of the total neutrons. Between the energies of the two groups, the ^{235}U cross-section varies by not much more than 10%. These numbers imply a maximum effect of 1%. If the correction is known within only 20%, the uncertainty in the measured cross-section is about 0.2%.

The ratio of the zero-degree angular differential cross-sections is really needed to produce each neutron group, and there are some data available. However, our sample was sufficiently far away so as to subtend a small enough solid angle so that the effects of the angular distribution were negligible. If it were possible to study effects as small as 0.2%, then perhaps the angular distribution would be a more important consideration.

At higher energies where we are planning to make measurements, we shall have to use a different neutron source such as $T(p,n)^{3}He$. Above about 3.5 MeV, neutrons are produced in inconveniently large numbers in the backing of the lithium target by reactions such as Ta(p,n). At sufficiently high energies, ^{7}Li itself disintegrates in neutron-producing reactions. At energies where it is possible to make corrections for the second neutron group, we have continued to use the $^{7}Li(p,n)$ source because we find it more convenient than a $T(p,n)^{3}He$ source, which would require special arrangements for handling the tritium.

MEASUREMENT OF THE ^{235}U FISSION CROSS-SECTION IN THE ENERGY RANGE 1 keV TO 1 MeV

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Abstract

MEASUREMENT OF THE ^{235}U FISSION CROSS-SECTION IN THE ENERGY RANGE 1 keV TO 1 MeV.

Measurements of the fission cross-section of ^{235}U using a 45-MeV linac and time-of-flight techniques have been extended into the energy region where most measurements have been made with Van de Graaff accelerators. The reported relative measurements cover the range 1 keV to 1 MeV. Fission events were detected by observing the fission neutrons with proton recoil detectors using pulse-shape discrimination against gamma rays. Most measurements were made with two detectors at 45° and two at 90°. The incident neutron spectrum was measured with a calibrated boron-vaseline plug detector. Background was determined by the 'black' resonance filter technique. Corrections were applied for self-shielding, multiple scattering and changes of fission neutron detection efficiency resulting from changes with incident energy in the temperature and angular distribution of the fission neutron spectrum. The measured relative cross-section has been normalized to an average value of 2.349 b in the 10- to 30-keV energy interval in agreement with the evaluation of Sowerby et al. The normalized data agree well with the Sowerby evaluation except below 3 keV and in particular confirm the shape of the cross-section in the 30- to 100-keV range as reported by Sowerby, in contrast with other recent evaluations. The estimated error of the relative cross-section is less than 4%.

1. INTRODUCTION

The discrepancies in the existing $^{235}U(n, f)$ data above the resonance region are disquieting because not only is the cross-section for this reaction used as a standard, but in the 10-keV to 1-MeV neutron energy region its value is of considerable importance to the design of fast reactors. Further accurate measurements are required and we have extended the time-of-flight technique on the 45-MeV linac to energies where most measurements have been made on Van de Graaffs. Fission events were recorded by observing the prompt fission neutrons, and the spectrum of the incident neutron beam was determined with a calibrated boron-vaseline plug detector [1].

2. EXPERIMENTAL METHOD

Both the fission yield and spectrum measurements were made on a 100-m flight path using the booster [2] target of the linac as the primary neutron source. All measurements were taken with an electron pulse length of 150 ns and a repetition frequency of 192 Hz. The events were recorded on a tape recorder with channels of 125 ns width. The overall resolution for both detector arrangements was 1.7 ns/m. The neutron beam was collimated

to 8 cm in diameter at the 100-m station and passed only through thin Melinex (Mylar) windows, a 5.6-m path in air and a graphite-clad overlap filter containing 0.43 g/cm^2 of ^{10}B. The only significant structure introduced into the spectrum of the incident neutron beam by the flight path was a dip of ~ 15% at 440 keV due to the path length in air.

2.1. The fission detector

The prompt fission neutrons were detected in one or more of the four NE 213 proton recoil detectors placed around the ^{235}U sample but out of the incident neutron beam. The samples, 7.9 cm in diameter, were mounted in a boron sleeve which served to reduce the background from backscattered neutrons. Gamma rays from neutron capture or inelastic scattering were rejected with a pulse-shape discrimination system using the charge-comparison method. This system was designed for high instantaneous count rates (up to 10^5 Hz) and gave good gamma discrimination at low energies [3]. The detection of scattered neutrons with incident energies below 1 MeV was prevented by setting the neutron discriminator bias of each detector above this energy. With the bias nominally at 1.5 MeV, time-of-flight tests with gold and iron samples showed that the detection of gamma rays in the ^{235}U fission yield measurements would be negligible at all energies. Similar tests with scattering samples showed that the detection of scattered neutrons would also be insignificant up to at least an energy of 1 MeV.

Above 100 keV incident neutron energy, the initially isotropic fission fragment emission is known to become forward peaked. The fission neutrons can be expected to exhibit a similar but less pronounced anisotropy and, in order to allow for this effect, the fission yields were measured with the detectors placed at various angles to the incident beam. Most of the measurements were made with two detectors at 45° and two detectors at 90°, the yields being recorded simultaneously. Further measurements were also made with detectors placed at 22.5°, 67.5°, 135° and 157.5° to the beam. The most accurate data were taken at 90° and these were used to derive the measured cross-section after correction for the angular dependence of neutron emission.

With the particular geometrical arrangement and bias setting used, the efficiency of each NE 213 detector for detecting a prompt fission neutron was $\lesssim 0.5\%$. In this case and for a thin sample, the observed fission yield per incident neutron of energy E is accurately proportional to $\epsilon(E)\overline{\nu}_p(E)\sigma_f(E)$, where $\epsilon(E)$ is the detector efficiency for a single prompt neutron, $\overline{\nu}_p(E)$ is the number of prompt neutrons per fission and $\sigma_f(E)$ is the fission cross-section. In practice, samples of thickness 1.08×10^{-3} and 3.8×10^{-3} ^{235}U atoms/b were used and corrections for self-shielding and multiple scattering in the samples had to be applied.

2.2. The spectrum measurement

The most accurate method of measuring the neutron booster spectrum above the energy region where '1/v' detectors are available is to use the 'black' detector of Coates et al. [4]. However, this is a device whose relative neutron detection efficiency cannot be predicted above 700 keV and, because of its relatively slow time resolution, its use above ~ 100 keV requires a 300-m flight path. For these reasons, the spectrum at the

100-m station was measured with a 'secondary standard' detector. This consisted of a boron-vaseline cylinder, 7 cm in diameter and 10 cm long, surrounded by four NaI crystals which detect the 478-keV gamma rays from the $^{10}B(n,\alpha,\gamma)^{7}Li$ reaction. The secondary detector was calibrated from 1 to 700 keV against the black detector using the linac 300-m flight path, and from 68 keV to 2 MeV against the Harwell long counter [5] using the pulsed Van de Graaff IBIS.

The fission and spectrum measurements were made with the same instrumentation and used the same flight-path collimation. Both measurements were also made with the detectors in the same position, and the time resolution in each case was therefore almost identical. This is of importance above 100 keV where the booster spectrum shows structure (dips of up to ~ 8%) caused by the stainless-steel booster fuel element cladding.

2.3. Background determination

The backgrounds in the time-of-flight measurements were determined with the 'black' resonance filter technique using samples of manganese, aluminium and SiO_2. All the background points between the 2.38-keV manganese resonance and the 440-keV oxygen resonance could be well fitted by a simple power law in flight time and this was used to extrapolate the background to 1 MeV. With the measurements extending to high energies it was not feasible to leave permanent resonance filters in the beam since these would introduce troublesome structure into the spectrum. Instead, measurements were made for each background resonance with several filters of different thickness. The background removal factor for the filter could then be determined and the open-beam background derived (i.e. the background for zero filter thickness).

In the fission measurements the ratio of the time-dependent background component to the true counts in the open beam was found to vary from ~ 0.002 at 2.38 keV to 0.028 at 440 keV, and the extrapolated ratio at 1 MeV was 0.02. In the spectrum measurements the time-dependent background fraction was found to be about 50% greater than the above values.

In both measurements the time-constant background component was only significant at the lowest energies and was accurately determined with the usual background gate.

3. CORRECTIONS

After the time-of-flight measurements have been corrected for background and any count loss effect, three further factors have to be applied before the cross-section is obtained. These are the corrections for self-shielding and multiple scattering in the sample, for the energy dependence of the neutron detector efficiency, $\epsilon(E)$, and for $\bar{\nu}_p(E)$. To some extent, the first two corrections can be dealt with experimentally.

3.1. Sample thickness

For the thicker of the two fissile samples, the self-shielding factor

$$\{1 - T(E)\}/\ln\{1/T(E)\}$$

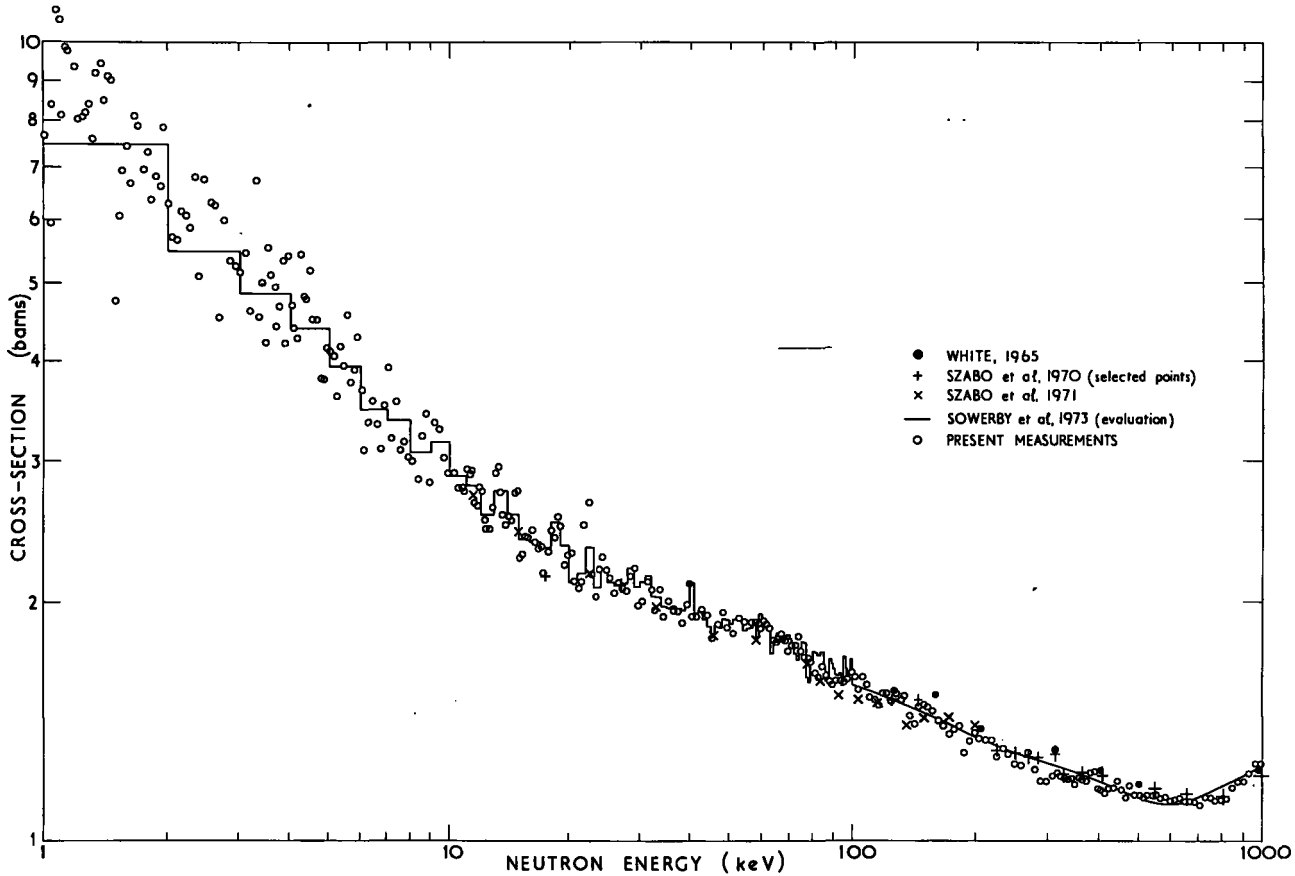

FIG. 1. ^{235}U fission cross-section.

was determined from a time-of-flight measurement of the sample transmission, T(E). The measurements were made at the 100-m position with a thin boron-plug detector. For both samples the self-shielding and multiple-scattering corrections were calculated with the Monte-Carlo code of Lynn and Moxon [6] using average cross-sections from the UKNDL-DFN 271D. For the present neutron energy range and because of the small ^{235}U level spacing, it was considered unnecessary to include resonance self-shielding in the calculations. For the thicker sample, the measured and calculated self-shielding factors were in agreement. In the 1- to 10-keV range the effect of multiple scattering for this sample is at a maximum and increases the observed fission yield by about 9%. However, because the multiple scattering and self-shielding corrections have opposite signs, the overall correction for sample thickness in this case changes by only slightly more than 2% over the complete energy range. For the thinner sample, the increase in yield due to multiple scattering is always less than 2%, and the overall correction for sample thickness changes by about 1% over the energy range.

3.2. Prompt neutron detector efficiency, $\epsilon(E)$

As the incident neutron energy increases, $\epsilon(E)$ will (a) increase because of the rise in temperature of the prompt neutron spectrum, and for a detector at 90° to the incident beam it will (b) decrease because of the increased forward peaking of neutron emission.

The increase in $\epsilon(E)$ with prompt neutron temperature is due simply to the increased number of neutrons above the detector bias. Since the effect is small at the present energies, it is adequate to represent the spectrum by a Maxwellian function with an average energy [7]

$$\overline{E}_p = 0.75 + 0.65 \left\{ \overline{\nu}_p(E) + 1 \right\}^{1/2}$$

It is also reasonable to ignore the gross structure in $\overline{\nu}_p(E)$ and assume $\overline{\nu}_p$ to be constant below 100 keV and to increase linearly between 100 keV and 1 MeV. With these assumptions and the experimental bias set at 1.5 MeV, $\epsilon(E)$ is constant below 100 keV and rises by $\sim 0.7\%$ at 1 MeV.

The measurements taken with the detectors at various angles to the beam showed the effect of anisotropic neutron emission to be quite small. Below ~ 200 keV, no difference in the energy dependence of the fission yield could be found between any of the angles. The only noticeable difference was an increase of about 2% in the ratio of the yield at 45° to the yield at 90° between ~ 200 keV and 1 MeV. Because the anisotropy is small and difficult to measure, the variation of $\epsilon(E)$ for the 90° detectors was calculated from the known fragment angular distribution

$$W(\alpha)/W(90°) = 1 + A \cos^2\alpha$$

$W(\alpha)$ being the fragment yield at an angle α to the incident neutron. Experimentally, A has been found to change from 0 at 180 keV to 0.11 at 1 MeV [8,9]. For simplicity, all the prompt fission neutrons were assumed to be emitted isotropically in the fragment rest systems. Allowance was made for the 1.5-MeV detector bias and for the angular resolution of the detectors. The calculation confirmed the experimental 45°/90° yield ratio

TABLE I. ESTIMATED ERRORS IN THE MEASURED RELATIVE CROSS-SECTIONS (%)

Energy range (keV)	Measurements		Corrections			Total error
	Spectrum	Fission yield[a]	Detector efficiency	Sample thickness	$\bar{\nu}_p(E)$	
1-3	3.6	0.2	0	1.0	1.0	3.9
3-5	3.2	0.2	0	0.5	1.0	3.4
5-100	2.8	0.5	0	0	1.0	3.0
100-500	3.1	0.9	0	0	1.0	3.4
500-700	3.3	1.4	0.3	0	1.0	3.7
700-1000	3.4	1.6	0.5	0	1.0	3.9

[a] Statistical errors in the fission yields are not included since they depend on the particular grouping of timing channels used to present the data. In Table II these statistical errors lie between 0.2% and 0.7% and are included with the above total errors.

and showed that in order to allow for anisotropy, the observed 90° yields have to be increased above 200 keV by a maximum of 1.1% at 1 MeV.

For the 90° detector used to determine the cross-section, the net change in $\epsilon(E)$ with energy due to the two effects is less than 0.4% at all energies.

3.3. Average number of prompt neutrons per fission, $\bar{\nu}_p(E)$

The energy variation of $\bar{\nu}_p$ was taken from the evaluation by Mather and Bampton [10]. The evaluated $\bar{\nu}_p(E)$ is almost constant below 100 keV and increases by 5% at 1 MeV. Gross structure is present in the form of a broad humped increase in $\bar{\nu}_p(E)$ of about 1% around 400 keV.

4. RESULTS

At low energies where the sample thickness correction is important, the measured relative fission cross-sections obtained for the two samples were found to agree to within about ±1% when the data were averaged over wide energy intervals to improve statistical accuracy. This agreement extended over the complete energy range, and because there was no apparent systematic difference between the two sets of data, the final cross-section was based on the measurements for the thicker sample which had superior statistical accuracy.

TABLE II. COMPARISON OF MEASURED AND EVALUATED CROSS-SECTIONS

Energy interval (keV)	Measured cross-section (barns)	Error (%)	Difference between measured and evaluated cross-section (%)		
			Sowerby et al. (1972)	ENDF/BIII	KEDAK (1972)
1-2	7.881	4.1	+5.7	+4.7	+5.8
2-3	5.722	3.9	+4.3	+2.5	+5.8
3-4	5.045	3.7	+3.7	+2.1	+3.8
4-5	4.474	3.5	+1.9	+1.1	+1.4
5-6	4.048	3.3	+2.7	+1.7	+2.9
6-7	3.379	3.2	-2.9	-5.3	-2.9
7-8	3.280	3.1	-2.9	-1.7	+1.8
8-9	3.071	3.1	0.0	+0.4	+2.2
9-10	3.153	3.1	-0.4	-1.5	+3.7
10-20	2.530	3.0	-0.8	-8.3	+2.5
20-30	2.166	3.0	+0.8	-7.2	+3.1
30-40	1.978	3.0	-1.7	-11.5	-4.8
40-50	1.893	3.0	-0.8	-7.3	-5.1
50-60	1.866	3.0	-0.3	-6.0	-2.4
60-70	1.815	3.0	+0.3	-3.6	-1.3
70-80	1.733	3.1	+1.1	-4.3	-2.3
80-90	1.612	3.2	-4.3	-8.1	-6.4
90-100	1.588	3.3	-2.8	-6.0	-4.8
100-200	1.444	3.4	-0.3	-3.8	-3.9
200-300	1.266	3.4	-2.1	-4.2	-5.3
300-400	1.196	3.4	-1.9	-3.0	-5.2
400-500	1.153	3.5	-0.5	-3.6	-4.4
500-600	1.134	3.6	+1.4	-2.2	-3.6
600-700	1.122	3.8	+0.3	-2.3	-3.5
700-800	1.122	3.9	-2.3	-0.7	-3.2
800-900	1.156	3.9	-2.2	+0.1	-2.1
900-1000	1.218	4.0	+0.1	+0.3	+0.2

4.1. Normalization

The measured relative cross-section has been normalized to an average value in the 10- to 30-keV energy interval of 2.349 b, to agree with the evaluation of Sowerby et al. [11]. This energy region was chosen for the normalization for two reasons:

(a) The errors in the present measurements are at a minimum.
(b) The 10- to 30-keV region contains measurements of several different types. The evaluation includes three linac time-of-flight continuous measurements using both fission fragment and neutron detection, one Sb-Be spot point measurement and two sets of Van de Graaff spot point measurements.

The measured cross-section is shown in Fig. 1. The full energy resolution is only shown above ~ 300 keV, at lower energies the channels have been grouped in such a way that the statistical error on any plotted point does not exceed ± 2%. The same data averaged over particular energy intervals are given in Table II.

4.2. Errors

The measurement errors, expressed as standard deviations, are summarized in Table I.

The errors in the spectrum measurement include both random and estimated systematic errors. These include contributions from uncertainties in the black detector and Harwell long counter relative efficiencies (both ~ ± 2%), uncertainties in the secondary standard cross-calibrations (± 2% to ± 3%), uncertainties in the background determination (± 0.2% to ± 1.5%) and uncertainties due to errors in the time-of-flight measurements (~ ± 1% from 500 keV to 1 MeV).

The fission yield errors given in Table I include only background and timing uncertainties. Random counting errors have been excluded since they depend on the particular grouping of timing channels used. For the data shown in Fig. 1, the errors due to counting statistics are generally between 1 and 2%, and for the energy intervals in Table II they vary from 0.2% to 0.7%. The errors given in Table II include the counting errors in the fission yields together with the total estimated error from Table I.

Liberal estimates have been made for the uncertainties in the first two correction factors. The uncertainty in the energy dependence of $\bar{\nu}_p$ is based on the evaluation of Mather and Bampton [10].

5. DISCUSSION

It can be seen from Fig. 1 that, even with grouped timing channels, the well known structure in the cross-section is very evident below 50 keV. At the full experimental resolution the structure is found to be in broad agreement with that observed by Bowman et al. [12].

From Table I it can be seen that the measurements are in close agreement with the evaluation of Sowerby et al. except at energies below about 2 keV. The agreement in the shape of the cross-section with the other evaluations is not so good. To normalize to the ENDF/B-III evaluation in the 10- to 30-keV range, the measured cross-sections would have to be increased by 7.8% and this would lead to discrepancies of between 8% and 12% at the highest and lowest energies respectively. Normalization to the KEDAK evaluation would require a reduction in the measured cross-sections of 2.8%, and although the general agreement is better than for ENDF/B-III, there are still discrepancies of around 8%.

The main difference between the evaluation of Sowerby et al. and that of others is in the region between 5 keV and 100 keV. In the region 30-100 keV the evaluation by Sowerby et al. has a relative shape which is determined by the bomb-shot data of Lemley et al. [13]. The close agreement between our data and this evaluation can then be taken to confirm the shape measured by Lemley et al. It can be seen from Fig. 1 that the present data are also generally in agreement with the Van de Graaff measurements of Szabo et al. [14, 15] and generally lie below the measurements of White [16].

ACKNOWLEDGEMENT

We should like to thank Dr. J.E. Lynn and the late Dr. E.R. Rae for their encouragement during the course of these measurements, and Drs. M.S. Coates, B.H. Patrick, M.G. Sowerby and C.A. Uttley for many helpful discussions. Mr. G.J. Hunt provided most of the computer programs used in the analysis of the time-of-flight data and Mr. M.C. Moxon carried out the Monte-Carlo calculations for sample thickness effects.

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DISCUSSION

Comparison with other measurements

J.L. LEROY: In your Fig.1, the 1971 data of Szabo et al. generally fall slightly below your data. These data of Szabo et al. will probably be raised by something like 4%, as I mentioned earlier, so the agreement with your results will probably become even better.

J.J. SCHMIDT: I have plotted some of Mr. Gayther's data on Fig.3 of Mr. Poenitz's presentation on ^{235}U (paper IAEA-PL-246-2/25 in these Proceedings). Gayther's data seem slightly higher in the valley around 800 keV, but the shape is exactly the same, rising to a kind of peak around 1 MeV.

W.P. POENITZ: I should stress again that my Fig.3 shows the shape only; the normalization is only approximate. Our absolute values are shown in Fig.6 and are about 1% higher than the rough normalization used in Fig.3. With correct normalization, our shape measurements will go upward in the direction of Mr. Gayther's values.

Normalization of the measurements

L. STEWART: Would it be possible for you to make an absolute measurement at some energy?

D.B. GAYTHER: No. We cannot determine the absolute efficiency of the detectors. Therefore, we must normalize our data somewhere.

As I mentioned in our status report, these measurements should not be considered only in isolation. We have measurements of the ^{239}Pu fission cross-section and therefore a ratio to the ^{235}U fission cross-section which is independent of our incident neutron spectrum. The ratio is useful in comparing our data with other measurements and in selecting the normalization. A possible complication is that the angular dependency of the fission fragment distribution is more extreme in ^{239}Pu than in ^{235}U.

H. LISKIEN: I notice that your Tables I and II do not include the normalization error.

D.B. GAYTHER: These tables show only the errors of the relative cross-sections. The error which Sowerby et al.[1] place on their evaluation in the energy range used for normalization of my values is 4.5%.

Change in detector efficiency

E.J. AXTON: You stated that the efficiency of the fission neutron detector increases with increasing energy of the neutrons incident on the ^{235}U because the temperature of the prompt fission neutron spectrum increases so that there are more neutrons above the bias. But doesn't the efficiency also decrease because the hydrogen scattering cross-section decreases with increasing neutron energy? I did a calculation some time ago and found that the bias had to be reduced to 0.8 MeV before the efficiency increased.

D.B. GAYTHER: We attempted to include the effect of the change in the hydrogen cross-section; the correction was very small.

Effects of changes in the angular distribution of fission fragments and fission neutrons

C.D. BOWMAN: You mentioned that since the angular distribution of the fission fragments was forward peaked, the neutron angular distribution might also be peaked in the same direction. Therefore, you make measurements at a number of angles. Can you tell me what anisotropy you actually observed?

D.B. GAYTHER: Our most extensive data are at 45° and 90°. Almost the entire series of measurements was made with two detectors at 45° and two at 90°, all of which were recorded simultaneously. At 1 MeV we saw only a 2% change between the detectors at 45° and the ones at 90°. The same is true at 135°, but the statistical accuracy was not so good. The effect becomes important only above 200 keV, just as the fission fragment angular distribution begins to become forward peaked only above this energy. Our calculations confirm the observed anisotropy, 2% ± 1% in the 45° to 90° ratio, if the uncertainty of the observations is considered.

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[1] SOWERBY, M.G., PATRICK, B.H., MATHER, D.S., UKAEA Rep. AERE-R 7273 (1973).

Extension of the measurements above 1 MeV and structure in the fission cross-section near 1 MeV

L. STEWART: Is there any possibility that Mr. Gayther could extend his measurements above 1 MeV?

D.B. GAYTHER: We have indeed taken measurements up to 1.5 MeV, but these have not been analysed yet. At these energies it becomes increasingly difficult to discriminate against scattered neutrons without at the same time eliminating much of the fission neutron spectrum.

I think the main improvement in these measurements will come from an improved knowledge of our neutron spectrum, especially at low energies. Although I would like to see improvement, I am not so concerned about the uncertainties at low energies because I think the principal value of these measurements is to bridge the gap between the energy range where measurements are usually made with a linac and the range where a Van de Graaff is usually used.

W.P. POENITZ: One reason why it would be interesting if you could extend your measurements a little above 1 MeV would be to investigate this 'step' in the cross-section more thoroughly. I have tried to determine where the cross-section begins to increase and the value to which it rises. My values tend to indicate that the cross-section has already reached the top of this step-like structure at 950 keV. This appears to be confirmed by ^{235}U to ^{239}Pu ratio measurements, which are easier to do and should be more accurate for this purpose.

D.B. GAYTHER: My values are still rising above 1 MeV, in fact much like the Sowerby[1] evaluation. I have not quoted numbers above 1 MeV because the bias settings which I used may have allowed a slight contribution from scattered neutrons.

All these measurements were made with a flight path of only 100 m. At 1.5 MeV, tiny errors become quite significant for such a short flight path and with long pulses of the length we used. It may be quite difficult to squeeze accurate numbers from the short flight times corresponding to 1.5 MeV, but we shall try.

In the future, we plan to make measurements in this important 1- to 5-MeV region using the synchro-cyclotron and measuring flux with a proton-recoil detector.

B.C. DIVEN: The lowest-energy points of measurements, which are reported in paper IAEA-PL-246-2/28 in these Proceedings, stop in just this energy range because the errors are becoming larger there. Therefore, we would not wish to make any comment about observation of a peak around 1 MeV.

[1] SOWERBY, M.G., PATRICK, B.H., MATHER, D.S., UKAEA Rep. AERE-R 7273 (1973).

MEASUREMENT OF THE NEUTRON FISSION CROSS-SECTION OF ^{235}U BETWEEN 0.5 AND 1.2 MeV

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Abstract

MEASUREMENT OF THE NEUTRON FISSION CROSS-SECTION OF ^{235}U BETWEEN 0.5 AND 1.2 MeV.

Because of the importance of the ^{235}U fission cross-section as a secondary standard, an absolute measurement was performed in the neutron energy range between 0.5 and 1.2 MeV. The method adoped was based on the hydrogen (n,p) cross-section. Monoenergetic neutrons from a fast pulsed source provided background discrimination by the time-of-flight method. A variety of runs was made to check the accuracy of sample masses and detector efficiencies. The consistency of the results confirmed that the experimental uncertainties of the seven absolute values lay between 2.6% and 3.4%. In addition, 12 relative values with statistical uncertainties of less than 2% were determined in order to establish the cross-section shape.

1. INTRODUCTION

Because of the increasing importance of the fission cross-section of ^{235}U as a standard in the fast neutron range, a number of experiments for its determination have been performed during the last twenty years. Nevertheless, the situation is unsatisfactory for neutron energies higher than a few hundred keV. Although most experiments claim an accuracy of a few per cent, the discrepancies between the results of different authors are three to four times larger than the given experimental uncertainties. Moreover, above 500 keV, there is no experiment which describes the shape of the fission cross-section sufficiently.

These difficulties have been the reason for a new effort towards an absolute measurement of the fission cross-section of ^{235}U. The method adopted is described in its first form in a previous paper [1]. In the meantime, the method has been improved in many details, so that a short description is given below.

2. SURVEY OF THE EXPERIMENTAL METHOD

In the case of thin samples, the relation for the fission cross-section is given by

$$\sigma_f = \frac{Z_u}{\theta \cdot N_u \cdot \epsilon_u} \qquad (1)$$

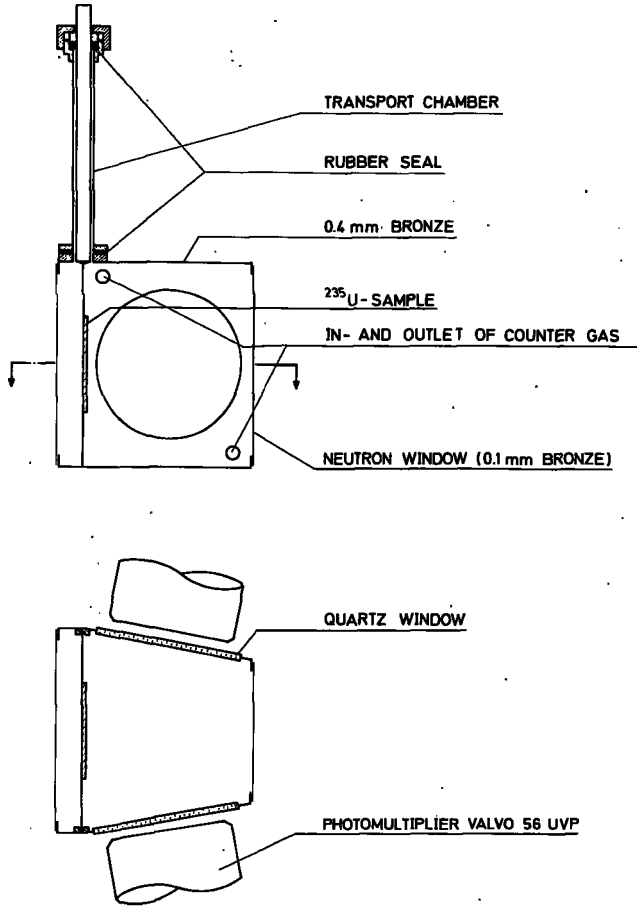

FIG. 1. Schematic view of the gas scintillation detector for the fission events.

where Z is the count rate, θ the neutron flux, N the number of fissile nuclei, ϵ the detector efficiency and u an index for the quantities related to the fission detector.

The experimental method of this measurement was designed to allow the determination of all quantities on the right-hand side of Eq. (1) with as few corrections as possible, and to check the reliability of the experimental results by the variation of all parameters on which these quantities may depend. By the consistency of both the estimated uncertainties and the experimental results, systematic or unrecognized errors could be excluded.

A fast pulsed, monoenergetic neutron beam, together with a fast fission detector, largely reduced background problems in the determination of the fission count rate. The fission detector, which is shown in Fig. 1, is a gas scintillation detector filled with a continuously flowing argon-nitrogen mixture of atmospheric pressure. Fission events were detected by

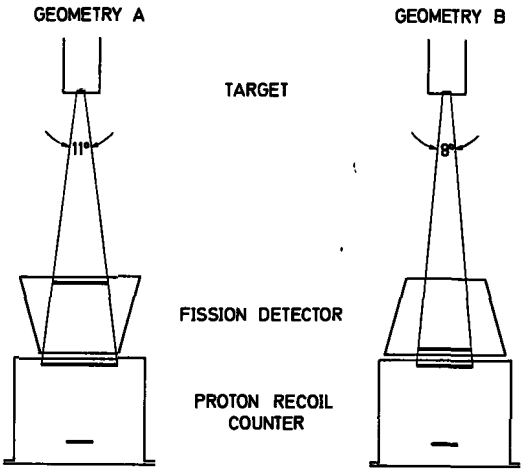

FIG.2. Arrangements of the detectors for fission events and neutron flux used in the experiment.

coincident pulses from two photomultipliers. By this, the multiplier noise was suppressed and the energy resolution was improved. For the determination of a background from (n, x)-reactions, the fissile samples could be withdrawn into a transport chamber. The detector efficiency ϵ_u was determined by comparing the measurements of samples of different thickness with a corresponding Monte-Carlo calculation. The number of fissile nuclei N_u follows from an accurate mass analysis of the samples. The determination of the neutron flux (which is described in paper IAEA-PL-246-2/14 in these Proceedings) was based directly on the hydrogen (n, p) cross-section as a standard and was free from threshold or extrapolation problems.

During the experiment, two arrangements of the proton recoil detector for the neutron flux measurement and the fission detector have been used, as is shown in Fig. 2. Geometry B served for the absolute measurement of the fission cross-section. In this back-to-back arrangement, the neutron flux is well defined for both samples and the flight path of about 30 cm between target and fissile sample allows the discrimination of most of the time-dependent background.

The advantage of geometry A is that the solid angle between target and fissile sample is doubled as compared to geometry B. Therefore, only half of the measuring time is necessary, as the fission count rate is much lower than that of the proton recoil detector. However, this advantage has to be paid for by much larger corrections and uncertainties. The finite size of the target caused deviations of about 10% in the neutron flux, especially in the outer zones of the samples. Moreover, the poorer discrimination of the time-dependent background causes uncertain corrections of several per cent. Therefore, only relative values of the fission cross-section have been determined from geometry A, which were normalized to the absolute values of geometry B. By means of the relative values, the energy dependence of the fission cross-section could be described in more detail. Because of the short

distance from the target and the relatively high neutron energies, no collimators were used in this experiment.

Altogether, 24 runs have been performed in geometry A and 27 runs in geometry B. All of these served to confirm the reliability of the experiment by the consistency of the results for varied parameters.

3. NEUTRON FLUX

The reader is referred to paper IAEA-PL-246-2/14 in these Proceedings, which gives a detailed description of the neutron flux determination.

4. EXPERIMENTAL DETAILS

4.1. Neutron source

Neutrons were produced by the bombardment of thin metallic ^{7}Li-targets with protons from the Karlsruhe pulsed Van de Graaff accelerator. All details are described in the above-mentioned paper.

4.2. Samples

The samples were fabricated and analysed by the Central Bureau for Nuclear Measurements (CBNM) of Euratom in Geel, Belgium. The fissile layers consisted of uranium acetate electrosprayed [2] onto thin polished backings of stainless steel. The average range of about 2.2 mg/cm^2 for fission fragments in uranium acetate limited the thickness of the samples to about 200 μg/cm^2; otherwise, the discrimination between α-particles and fission fragments could have been disturbed.

Although the range of fission fragments is about three times larger in uranium oxide, the acetate results in a better discrimination between alphas and fission fragments. The reason for this effect is that the light absorption of the black oxide sample is much larger for fission fragments than for α-particles as the specific ionization of fragments is largest near the sample while that of alphas is a minimum near the sample. Thus the bright, yellow acetate is preferred as a sample material for the gas scintillation detector.

To avoid systematic uncertainties of the detector efficiency by local differences in the absorption of fission fragments, a good homogeneity of the fissile samples was essential. The samples provided by Geel were on backings with a polish of 0.5 μm and had a homogeneity of better than 1%.

For the mass determination of the samples two methods were used. Before the samples had left Geel and after they had been returned to Geel for the final analysis, their α-activity was determined within 0.3% by low-geometry counting [3]. In this way it was ensured that no remarkable losses of the sample masses had happened during transport or during the experiment. From the known specific activity of the sample material the total mass was calculated with an uncertainty of 0.7%.

Finally, a destructive analysis of the samples by the isotopic dilution method [4] was performed with a total uncertainty of ± 0.4%. The results of the two methods agreed within 0.2%. Thus the sample mass is believed to be accurate to ± 0.4%. The characteristic data of the samples are given in Table I.

TABLE I. THICKNESS, TOTAL MASS AND UNCERTAINTY OF THE ^{235}U-SAMPLES

Backing thickness (mm)	Sample thickness ($\mu g/cm^2$)	Total mass (mg)	Uncertainty (%)
0.25	139	1.744	0.4
0.25	95	1.194	0.4
0.25	74	0.933	0.4
0.9	100	1.265	0.4

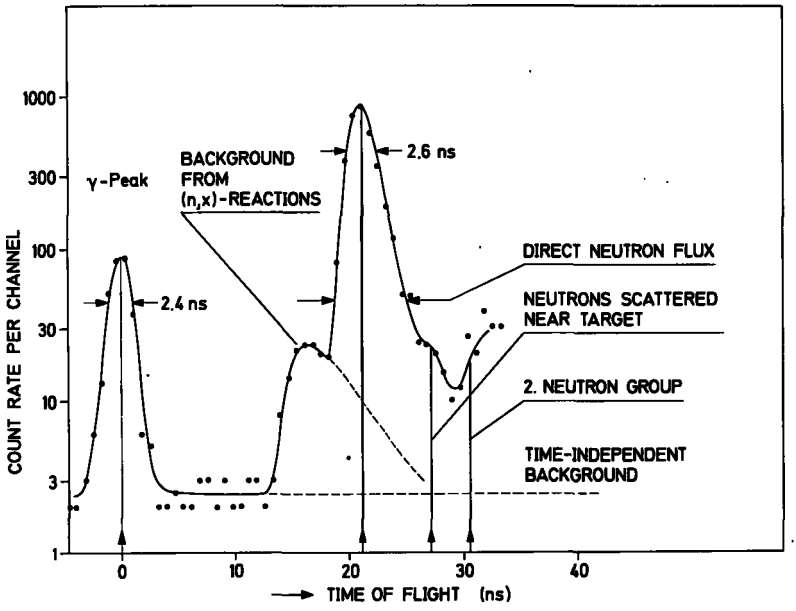

FIG. 3. Time-of-flight spectrum of the fission detector at a neutron energy of 911 ± 23 keV.

4.3. Electronics

The scintillations of the fragments were converted into pulses by the two multipliers. Fast pulses were taken from the anodes for the time-determining branch of the electronics, and integrated pulses were taken from one dynode of each multiplier for the pulse-height branch. A coincidence between the fast pulses furnished the start-signal for a time-to-amplitude converter and the gating for the summed pulse-height signals. Thus time-of-flight and pulse-height spectra were recorded simultaneously, both steered by the automatic sample changer as it is described in the above-mentioned paper in these Proceedings.

Figure 3 shows a time-of-flight spectrum of the fission detector in geometry B for a neutron energy of 911 ± 23 keV. The overall time resolution of 2.4 ns can be derived from the γ-peak. By means of the logarithmic scale the background discrimination is more clearly demonstrated. In addition to a time-independent background resulting from room-scattered neutrons, a background due to (n, x)-reactions arises. This was directly measured with the sample withdrawn into the transport chamber. It was found to be approximately constantly 2% of the main fission event (direct neutron flux in Fig.3) over the whole neutron energy range. This main peak is somewhat broader than the γ-peak because of the neutron energy distribution. Delayed with respect to the main neutron flux by several nanoseconds are events caused by slower neutrons of two different types. Both the neutrons scattered near the target and those from the second neutron group are discussed in the description of the neutron flux determination in the above-mentioned paper in these Proceedings. However, Fig.3 shows that their contribution to the fission count rate Z_u can be discriminated by the time-of-flight method. The uncertainty of the count rate Z_u is composed of the statistical uncertainties of the main peak and the subtracted background events.

4.4. Detector efficiency

The detector efficiency ϵ_u is defined as the ratio of detected fission events to all fission events which took place. For its determination it was necessary to know the fraction of fission events which could not be detected because of energy loss in the sample.

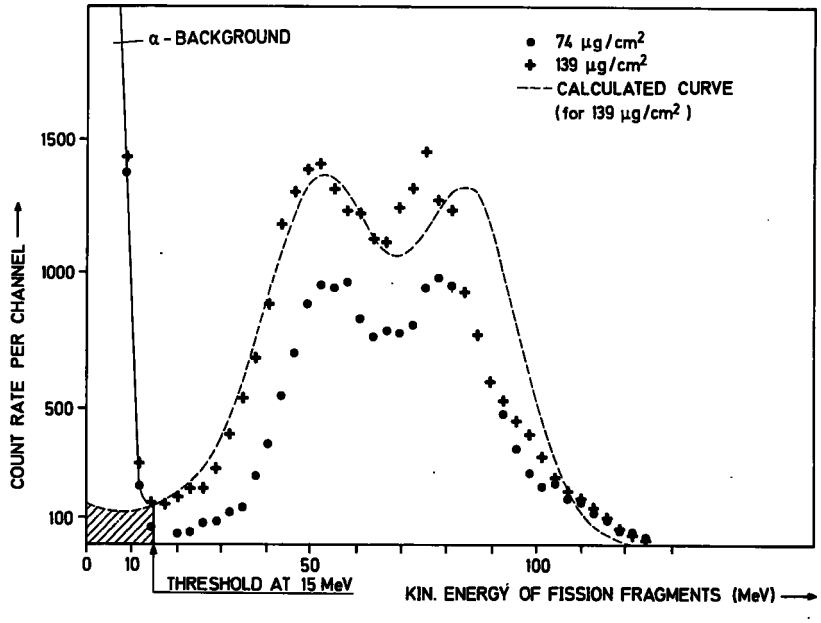

FIG.4. Measured and calculated pulse-height distributions for fission fragments.

TABLE II. EFFICIENCY OF THE FISSION DETECTOR

Sample thickness ($\mu g/cm^2$)	Efficiency		Uncertainty (%)
139	0.959	(0.957)	1.0
100	0.969	(0.969)	0.8
95	0.969	(0.968)	0.8
74	0.978	(0.976)	0.6

Figure 4 shows experimental pulse-height distributions for two sample thicknesses. The dashed line represents a calculated distribution. The deviation of the measured from the calculated distribution at energies above 70 MeV is due to the fact that the multiplier bases were designed for optimal time resolution. This resulted in non-linearities for large pulses, a detail which does not affect the results.

The adjustment of the energy scale was performed in the linear part below 70 MeV by means of the well known pulse-height distribution of a very thin ^{252}Cf-source [5]. It was confirmed by the fit of the calculated distribution to the experimental spectra of ^{235}U. In this way the position of the electronic threshold was determined to be 15 ± 5 MeV. Although the given uncertainty of ± 5 MeV is an extreme value, its influence on the efficiency is very small, as follows from the shape of the pulse-height spectra.

The fraction of fragments which are absorbed or leave the sample with energies below 15 MeV is calculated by fitting the experimental fission fragment distribution to a curve computed from the theory by Lindhard, Scharff and Schiott [6]. Since dE/dx values for uranium acetate are not available, approximate values were taken and adjustments made according to the above fits. A Monte-Carlo method was used to select the places and depths of fission events in the sample and the direction of fragment emission. The calculated distributions were also corrected for detector resolution.

The resulting values of ϵ_u are given in Table II for the samples used in the experiment. They agree well with the values in brackets, which were derived from an equation given by Rossi and Staub [7] for the absorption losses in a homogeneous foil with the adjusted dE/dx values as input data. The agreement confirms again the good homogeneity of the samples.

4.5. Corrections

In the discussion of the time-of-flight spectrum in section 4.3, one background component was not mentioned. Neutrons of the primary flux which are scattered in the detector or the nearby surroundings may cause fission events which cannot be discriminated by the time-of-flight method. There is a possibility to determine this background experimentally, i.e. as the difference between the values of the fission cross-section derived from normal runs and those with doubled detector masses. But this method is very uncertain because the resultant correction of about 6% came from the difference of two big numbers with statistical uncertainties of the same order. Thus this method was used only as a check on calculated values of the scattering correction.

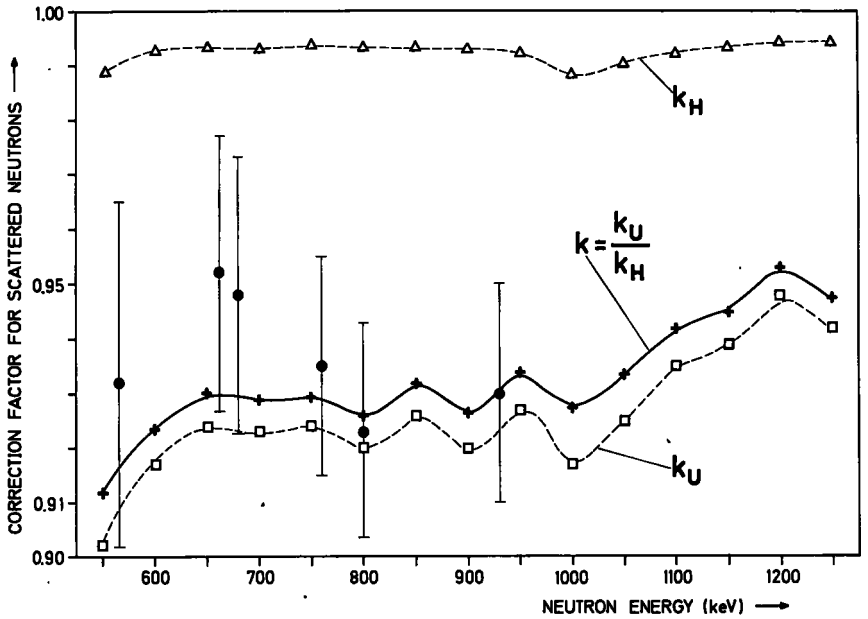

FIG. 5. Correction factors for neutrons scattered in the detectors.

For these calculations the detector geometry was approximated by cylinders and planes, and the scattering correction was determined for random scattering places and neutron directions from the differential cross-sections and angular distributions for the respective detector materials [8-10]. The correction factors k_U and k_H for the count rates of the fission and proton recoil detector are shown in Fig. 5 for the detector arrangement of geometry B.

The fluctuations are caused by corresponding fluctuations in the scattering cross-sections and the angular anisotropies. As was expected from the characteristics of the proton recoil detector, the correction k_H is much smaller than k_U, although it is increased by the presence of the fission detector. In Fig. 5 the computed resulting correction k is shown as a function of neutron energy. The experimental values are given by full circles.

The calculated contribution of scattered neutrons to the count rate has an uncertainty of about 20%. Thus the uncertainty of the correction factor is between 1.0% and 1.6%.

4.6. Uncertainties

In the same way as for the neutron flux determination it was ensured that the absolute determinations of the fission events were consistent when the experimental parameters were varied. By using different sample and backing thicknesses, the number of fissile nuclei, the counter efficiency, and a main part of the scattering correction were changed. The observed results were again consistent with the estimated uncertainties, which are listed in Table III, together with the overall total uncertainty for the absolute values of the fission cross-section.

TABLE III. EXPERIMENTAL UNCERTAINTIES FOR THE ABSOLUTE VALUES OF THE FISSION CROSS-SECTION DETERMINED IN GEOMETRY B.

Source of uncertainty		Uncertainty (%)
Number of fissile nuclei	N_u	0.4
Detector effeciency	ϵ_u	0.6 - 1.0
Fission count rate	Z_u	0.6 - 1.4
Scattering correction	k	1.0 - 1.6
Neutron flux determination		1.7 - 2.4
Overall total uncertainty		2.6 - 3.4

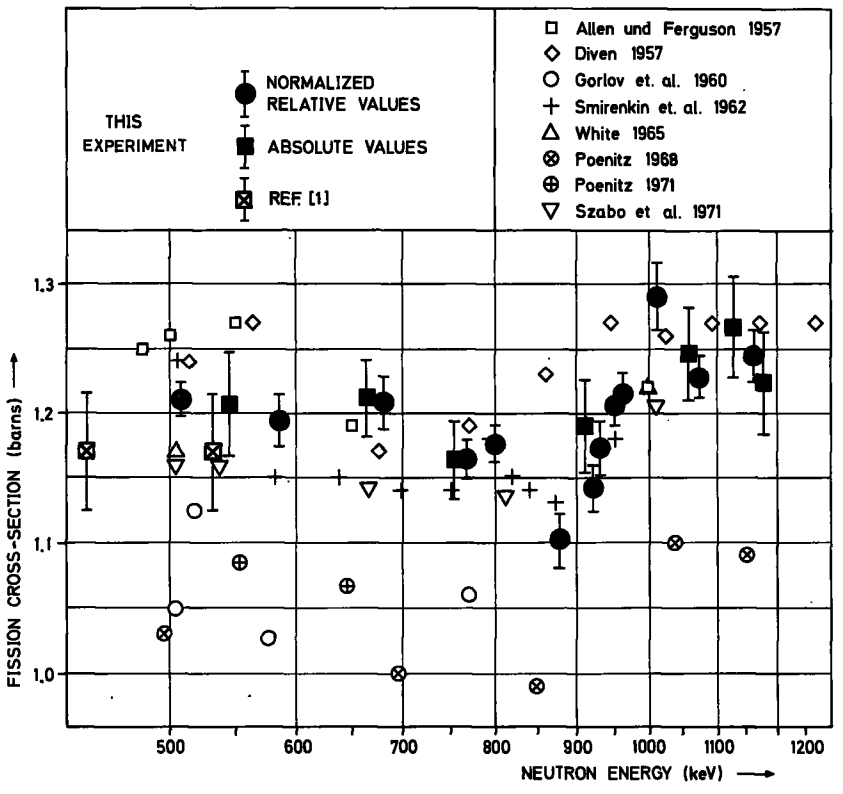

FIG. 6. Fission cross-section values from this experiment and from existing measurements.

It is important to notice that the main experimental uncertainties are related to the fission detector and not to the neutron flux determination. A reduction of these uncertainties caused by the scattering correction k_u and the efficiency ϵ_u may be achieved by using a neutron collimator and thinner samples. However, this can be done only with decreased overall efficiency of the fission detector and must be compensated for by a more intense neutron flux.

The relative values, measured in geometry A, were normalized by an optimal fit to the absolute values. The uncertainty of this fit is of the order of 3%, corresponding to the uncertainty of the absolute values. However, the shape of the fission cross-section is described with much better accuracy and is only dependent on the statistical uncertainties. Therefore, only the statistical uncertainties are quoted for the normalized relative values.

TABLE IV. NUMERICAL VALUES OF THE ^{235}U FISSION CROSS-SECTION

Neutron energy (keV)	Fission cross-section (b)	Overall uncertainty (%)
NORMALIZED RELATIVE VALUES		
513 ± 32	1.210	0.9
580 ± 20	1.193	1.8
678 ± 21	1.208	1.2
767 ± 20	1.165	0.9
795 ± 20	1.178	1.2
872 ± 20	1.12	2.1
920 ± 22	1.141	1.5
930 ± 20	1.172	1.9
945 ± 24	1.206	1.1
966 ± 21	1.213	1.2
1013 ± 20	1.293	1.9
1060 ± 22	1.227	1.2
ABSOLUTE VALUES		
546 ± 22	1.207	3.4
662 ± 23	1.215	2.6
758 ± 23	1.164	2.6
908 ± 22	1.193	2.9
1057 ± 26	1.248	3.0
1125 ± 25	1.256	3.4
1175 ± 25	1.221	3.4

5. RESULTS AND DISCUSSION

As a result of this experiment, 7 absolute and 12 relative values of the ^{235}U fission cross-section for neutron energies between 500 and 1200 keV were established. The overall uncertainties of the absolute values lie between 2.6% and 3.4% and those of the relative values between 0.9% and 2.1%. Figure 6 gives a survey of the results of this experiment and of the cross-section values measured by several authors. The numerical fission cross-sections are listed in Table IV.

The results of the previous measurements can be divided into two groups, which show a discrepancy of 10 - 15%, although their given uncertainties are of the order of 3 - 7%. The first group consists of the values of Allen and Ferguson [11], Diven [12], Smirenkin et al. [13], White [14], Szabo et al. [15] and two values of a preceding measurement [1]. The second group with lower fission cross-sections consists of the values of Gorlov et al. [16] and of Poenitz [17,18]. In general, the two groups can be distinguished by the fact that the experiments of the first group are more or less based on the hydrogen scattering cross-section as a standard, while in the experiments of the second group other methods for the neutron flux determination are used. As can be seen from Fig. 6, the results of the present experiment agree within the experimental uncertainties quite well with the data of the first group.

In detail, there is good agreement above 750 keV with White, Szabo et al., Allen and Ferguson, Smirenkin et al. and, at least above 1000 keV, with Diven too. For neutron energies from 500 to 750 keV the present results lie between the results of Refs [13-15] and those of Refs [11,12]. However, these differences can be explained by the respective experimental uncertainties.

In addition to the determination of absolute values, the results of this work also allow a more complete description of the fission cross-section shape in the investigated energy range. This is of importance for the use of the ^{235}U fission cross-section as secondary standard. Within the energy resolution of the experiment, the measured shape shows a smooth behaviour up to about 900 keV. At this energy, however, a 15% step-like increase with a width of 100 keV was observed. Above 1000 keV the cross-section seems again to be smooth with the energy.

This step in the fission cross-section was not unexpected because corresponding dips in fission cross-section ratios had already been observed at this energy [19-21]. An explanation may be that this increase marks the end of the energy gap in the compound nucleus ^{236}U. At this energy the fission width increases because of the onset of one-particle states. This assumption is supported by the behaviour of the fission fragment angular anisotropy [22] which shows a correlated decrease owing to the higher K-quantum numbers of the one-particle states. Also a correlated structure was found for the average kinetic energy of the fission fragments $\overline{E_{kin}}$ and for the average number of fission neutrons $\bar{\nu}$ [23].

ACKNOWLEDGEMENTS

I wish to thank Drs. Verdingh, Lauer, Delbino, Spernol and de Bièvre from CBNM, Geel, for the careful preparation and mass analysis of the ^{235}U samples and Dr. R.R. Spencer for his help with the manuscript.

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DISCUSSION

W.P. POENITZ: Since you normalized your relative values to absolute values, I would expect the error bars of your final results to be larger than those of the absolute values.

F. KÄPPELER: You are correct. I have quoted only the statistical errors of the relative measurements because they describe the shape of the fission cross-section. The normalization factor has an uncertainty which is at least as large as the uncertainty of the absolute values.

W.P. POENITZ: How did you separate the true fission count rate from the background?

F. KÄPPELER: I showed the time-of-flight spectrum in which contributions from various (n, x) reactions are included (x = charged particle). I subtracted the experimentally determined background and corrected for the effects of neutrons scattered in the detector itself to obtain the fission count rate.

W.P. POENITZ: Where did you set the threshold of the detector?

F. KÄPPELER: This is shown in Fig. 4. It was set in the valley between the alpha peak and the fission-fragment peak. If the threshold had been set higher, the extrapolated part of the fission fragment peak and the associated uncertainty would have been larger. The correction for contributions from (n, x) reactions was about 2%, so the uncertainty of this correction should be small.

W.P. POENITZ: Around 700-800 keV you have high values which deviate from other shape measurements in this range, e.g. those of Gayther et al.[1], Szabo et al.[2] and myself[3]. In this range, there are very strong peaks due to N(n, p) and N(n, α) reactions, whereas around 900 keV, where you show a dip in the ^{235}U fission cross-section, the nitrogen cross-sections also have a dip. I assume that you also used an argon-nitrogen mixture in the fission detector.

F. KÄPPELER: The correction for (n, x) reactions, including those you just mentioned, was measured over the whole energy region, so I do not believe they would cause a systematic error of this type.

R.W. PEELLE: Did you see a difference in the shape of the fission pulse-height spectrum when the uranium deposit faced toward versus away from the neutron source (geometries A and B in your Fig. 2)?

F. KÄPPELER: I observed no definite deviations between the two spectra.

J.L. LEROY: The uncertainty of the shape of the cross-section is not entirely statistical. For example, there is certainly some uncertainty in the energy-dependent scattering correction.

F. KÄPPELER: That is correct. In the error bars, which I called statistical in the text, are also included energy-dependent uncertainties for the scattering correction and for the scattering cross-section of hydrogen.

T.A. BYER: Since you are still waiting for the results of destructive analyses of the samples, should we perhaps regard these results as semi-preliminary.

F. KÄPPELER: Yes. However, the samples were made from a batch of uranium which had a well known specific activity and have been inter-compared by alpha counting. Within a year, we do not expect the final mass determination to be very different from that which we are able to calculate now. Perhaps Mr. Deruytter could comment on this question.

A.J. DERUYTTER: I agree with Mr. Käppeler that very little change should be expected as a result of the final mass determination. In this case, there are no discrepancies in the alpha count rate such as occurred with the foil used by Szabo[4]. Our alpha counting methods are very reliable in my opinion. By now they have been confirmed by measurements on hundreds of foils.

When more than one foil have been used in a set of measurements, the foils can be intercompared frequently by alpha counting so that it is possible to determine whether any one of them has changed. Since Mr. Käppeler used three foils, I believe it is unlikely that any additional systematic uncertainty will be added to his results as a result of the final destructive analyses.

F. KÄPPELER: I made measurements on three foils, and I believe that three more foils were prepared at the same time as the foils used in the measurements.

(Editor's Note: The question arose whether the data obtained by Szabo et al.[4] using the fission chamber and the data of White[5] should be raised by as much as 2.5% because of the change in the recommended value

[1] Paper IAEA-PL-246-2/26 in these Proceedings.

[2] Paper IAEA-PL-246-2/24 in these Proceedings.

[3] Paper IAEA-PL-246-2/25 in these Proceedings.

[4] SZABO, I., et al., Neutron Standards and Flux Normalization (Proc. Symp. Argonne, 1970), CONF-701002, USAEC Symp. Ser. 23 (1971) 257.

[5] WHITE, P.H., J. Nucl. Energy A/B 19 (1965) 325.

of the half-life of ^{234}U. Such a change would result in better agreement between these data and the data presented by Käppeler.

In the discussion it was pointed out that the mass determination used by White was based on four independent methods of analysis whose results agreed within their assigned errors. On the basis of private communications it was suggested that the mass determination used in the calculation of the cross-sections depended on the weights assigned to the several methods of analysis and that the final value was not particularly sensitive to the value assumed for the half-life of ^{234}U.)

PROGRESS REPORT ON LASL MEASUREMENT OF THE ^{235}U FISSION CROSS-SECTION FROM 1 TO 6 MeV

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Abstract

PROGRESS REPORT ON LASL MEASUREMENT OF THE ^{235}U FISSION CROSS-SECTION FROM 1 TO 6 MeV.

Measurements of the ^{235}U fission cross-section in the range 1 to 6 MeV, using a pulsed Van de Graaff accelerator, 2π fission counting and a proton recoil telescope to measure the neutron flux, are in progress. The product of the amount of ^{235}U on the fission foil and the efficiency of the 2π fission counting system was determined in a separate experiment by comparison with the fission rate of a standard ^{235}U foil counted in low geometry in a uniform thermal neutron flux. Corrections to the data and sources of error are described. Preliminary data contingent upon final mass determination of the standard ^{235}U foils are given.

In March 1970, a measurement of the ^{235}U fission cross-section was started. The experimenters were D. M. Barton, P. G. Koontz and R. K. Smith. Plans for the experiment were made in consultation with a task group consisting of the experimenters and R. L. Henkel, J. C. Hopkins, G. Hansen and B. C. Diven, chairman. In 1971, Koontz retired and the experimenters were joined by G. A. Jarvis. In 1972, D. M. Drake replaced Hopkins, and Hansen replaced Diven as chairman. Hansen also has made extensive calculations of all corrections and has studied carefully the consistency of the data.

Before the experiment was started, past measurements were studied and limitations on accuracy surveyed. It was decided that the neutron flux should be measured with a proton telescope using polyethylene as the proton source. The fission counter was to use nearly 2π geometry to avoid problems associated with poorly known angular distributions of fission fragments. The ^{235}U deposit and polyethylene foils were to have the same diameter and to be located as close together as possible. Solid-state detectors were chosen for both fission and proton detectors. The experimental arrangement is shown in Fig. 1.

The use of 2π geometry produced an unacceptable uncertainty in the fraction of fission events detected, and a separate calibration experiment was performed to determine the product of the amount of ^{235}U on the fission foil and the detection efficiency. In the calibration experiment the CH_2 foil was replaced by a thin deposit of ^{235}U whose mass was accurately known, and the proton detector was used to detect fission fragments from the standard foil. The assembly was placed in a uniform thermal neutron flux and the ratio of counts in the two detectors determined the effective mass of ^{235}U in the fission counting system. Since the actual mass of ^{235}U on both fission foils and the geometry factor of the proton counting system were all known, the efficiency of the fission counting system could be determined.

TABLE I. ^{235}U FISSION CROSS-SECTIONS – PRELIMINARY DATA
(Hansen, Barton, Jarvis, Koontz, Smith, 10 Nov. 1972)

Energy (MeV)	σ_f
1.0	1.238 ± 0.014
1.1	1.261 ± 0.016
1.2	1.254 ± 0.014
1.3	1.250 ± 0.015
1.4	1.233 ± 0.024
1.5	1.268 ± 0.014
1.6	1.240 ± 0.014
1.7	1.294 ± 0.016
1.8	1.275 ± 0.017
1.9	1.274 ± 0.017
2.0	1.276 ± 0.011
2.2	1.273 ± 0.014
2.4	1.253 ± 0.019
2.5	1.251 ± 0.020
2.6	1.219 ± 0.012
2.7	1.224 ± 0.014
2.8	1.210 ± 0.012
2.9	1.193 ± 0.014
3.0	1.216 ± 0.009
3.2	1.215 ± 0.016
3.4	1.183 ± 0.016
3.5	1.182 ± 0.011
3.6	1.182 ± 0.016
3.7	1.155 ± 0.015
3.8	1.164 ± 0.017
4.0	1.140 ± 0.011
4.2	1.143 ± 0.019
4.4	1.118 ± 0.016
4.6	1.098 ± 0.016
4.8	1.108 ± 0.016

TABLE I (cont.)

Energy (MeV)	σ_f
5.0	1.089 ± 0.015
5.1	1.085 ± 0.019
5.2	1.090 ± 0.014
5.3	1.085 ± 0.014
5.4	1.067 ± 0.012
5.5	1.063 ± 0.014
5.6	1.048 ± 0.014
5.7	1.068 ± 0.015
5.8	1.092 ± 0.025
5.9	1.129 ± 0.018
6.0	1.145 ± 0.023

Note: The uncertainties listed include: (1) 0.24% uncertainty in calibration factor; (2) ~0.3% uncertainty in secondary source factor, i.e. neutrons not from gaseous tritium in target cell (vacuum background); (3) ~0.3% uncertainty in detector assembly in-scatter factor; (4) ~0.2% uncertainty in room return factor; (5) 0.5% uncertainty in ^{235}U assay; (6) 0.2% uncertainty in fission fragment anisotropy factor; and (7) uncertainty due to counting statistics and lack of reproducibility. Not included are errors associated with hydrogen assay and hydrogen scattering cross-section.

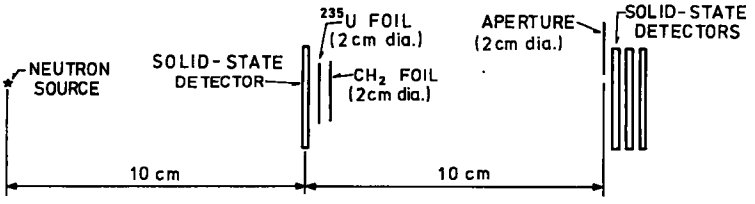

FIG. 1. Experimental arrangement (not to scale).

The efficiency is 90% at 10 MeV fission fragment bias. Except for small corrections, the efficiency of the fission counter and the geometry factor of the proton counter need not be known because the ratio of these quantities was used in both the calibration experiment with thermal neutrons and the fission cross-section measurement at high neutron energy.

The efficiency of the fission detector changes slightly as the neutron energy changes because of the changing angular distribution of fission fragments and centre-of-mass effects. This change, which is not monotonic, varies from zero to 1% in the neutron energy range from 1 to 6 MeV.

In some past measurements with counter telescopes and fission counters, large corrections were made necessary by scattering of neutrons in counter materials. The scattering affects the two counters differently. The design of the counter system was carried out in conjunction with calculations of the scattering corrections, so that errors in the corrections could be kept small. These corrections range from 1.3% to 2.4%.

Room-return neutrons contributed a negligible error in this experiment because a monoenergetic pulsed source of neutrons was used, and a time-of-flight spectrum determined the fraction of fissions caused by these neutrons. The fraction is less than 2% in all cases.

The concentration of hydrogen in the stock from which the polyethylene foils were prepared was determined by two laboratories, a private chemical laboratory and the National Bureau of Standards.

The cross-sections resulting from this experiment are dependent upon the knowledge of the absolute number of ^{235}U atoms on the standard foil used in the calibration experiment. Thirteen deposits of ^{235}U were prepared by vacuum deposition from a single supply of ^{235}U. Thicknesses were 125 and 550 $\mu g/cm^2$. All thirteen foils were intercompared by alpha counting in low geometry. The intercomparisons were made by two different groups at Los Alamos. The thirteen foils were also intercompared by fission counting in a thermal flux in a specially designed counting system. Four of the thirteen foils were retained as standards by the experimenters. One of the four foils was used in the calibration experiment. Two foils were sent to the U.S. National Bureau of Standards for analysis by isotopic dilution and coulombmetric techniques. Two foils were sent to the Central Bureau for Nuclear Measurements, Geel, for analysis by whatever means they prefer. Two foils were analysed at Los Alamos by a colorimetric method, and one by comparison with another local standard foil that had been prepared by stippling with aliquots of a ^{235}U solution prepared from weighed quantities of ^{235}U. These analyses are not yet all completed, so that the cross-sections given in Table I are preliminary and will be multiplied by an appropriate factor after the analyses are all in; otherwise, all corrections have been made to the data. Of course, changes may be made in the meantime, after collection of additional data.

DISCUSSION

Backgrounds from scattered neutrons and from reactions in the detectors

M. S. COATES: Have you investigated the effects of spurious neutrons which are scattered or produced other than in the target reaction?

B. C. DIVEN: I have mentioned the use of the monoenergetic pulsed source and time-of-flight to eliminate effects of room-return neutrons. Effects of neutrons scattered in the structural materials of the counters cannot be eliminated by time-of-flight because the spurious effects originate too close to the detectors. Corrections were made by calculation and were reasonably small. The spectrum of neutrons produced by scattering in this particular type of tritium target is well known from other time-of-flight experiments over the years, and appropriate corrections have been made.

B. D. KUZMINOV: The neutron energies were sufficiently high to cause reactions in the solid-state detectors. What interferences do these reactions cause?

B. C. DIVEN: These effects depend very much upon neutron energy. At low neutron energy there are very few reactions in solid-state detectors. The proton detectors in the proton-recoil telescope are of various thicknesses; the first is quite thin. At low energies near 1 MeV, the recoil protons do not go all the way through the first detector, but the background is also very low. As neutron energy is increased to 2-3 MeV, the protons go through the first detector and into the second. As neutron energy increases, the background also increases, but as soon as a coincidence can be obtained between the first two detectors most of the background can be eliminated by the coincidence requirement.

Backgrounds are measured after each run in which 10 000-20 000 fission events have usually been recorded while the polyethylene-foil proton radiator is in front of the proton detector. Then the polyethylene foil is switched out of the beam path, and another 10 000-20 000 fission events are collected during the background measurement. This background varies greatly with neutron energy.

Another effect also contributes to greater uncertainty as the neutron energy is decreased toward 1 MeV. As the proton range becomes shorter, the thickness of the polyethylene foil must be decreased, whereas the various backgrounds remain nearly the same. The uncertainty at 1 MeV must be significantly greater than at 2 MeV even though statistical accuracy is good.

Hydrogen content of polyethylene foils

H. LISKIEN: How well did the independent analyses of the hydrogen content of the polyethylene foils agree?

B. C. DIVEN: The hydrogen contents of the foils submitted for analyses agreed to better than the 1% accuracy claimed by each of the two laboratories. However, there may be other uncertainties in the hydrogen content.

The polyethylene foils which were actually used in the experiment might have differed in hydrogen content from those which were destructively analysed. However, at many energy points, more than one polyethylene foil were used.

It is somewhat difficult to determine the hydrogen content of the very thin foils, which must be used at low neutron energies, because of interferences such as absorbed hydrogen and significant changes in weight of the very light-weight foils owing to evaporation of absorbed substances. The masses of these light foils can be satisfactorily determined by 'neutron weighing'. In a high-energy neutron beam the relative proton count rate of a light foil is compared with the proton count rate of a heavy polyethylene radiator which can be accurately weighed. The hydrogen content of the light foil is then calculated from the mass of the heavy foil and the relative count rates.

H. LISKIEN: We also used this method and found that certain polyethylenes lost hydrogen with time.

F. KÄPPELER: Have you compared the hydrogen content of the polyethylene-foil radiators before and after use in the experiments?

B. C. DIVEN: The experimenters are aware of Mr. Liskien's investigations of absorption and loss of hydrogen from polyethylene. I do not believe they have made any investigation of these effects themselves, and I do not know their future plans.

Error due to uncertainty in the $^{1}H(n,p)$ standard cross-section

B. C. DIVEN: In some energy regions, the error in this experiment is due mostly to uncertainty in the hydrogen cross-section for production of protons at zero degrees. Although the total n-p scattering cross-section is quite well known in the MeV range, the angular distribution is not. In the higher energy ranges to be covered in these experiments, uncertainties in the angular distribution contribute an error of 2%. At 14 MeV, for instance, other errors are expected to be much smaller than that. In general, we expect the errors to be no greater than 3% although at energies where the angular distributions are very poorly known the error may be greater than 3%.

Pulse-height spectrum of the 2π fission chamber

A. J. DERUYTTER: Although the calibrations were made in low geometry with sharply defined pulse-height spectrum, the actual measurements were made with 2π fission chambers so that the stability of the fission-fragment pulse-height spectrum must be very important.

B. C. DIVEN: Yes, the shape of the pulse-height spectrum is one of the considerations which limit the thickness of the ^{235}U deposit. The shape of the fission-fragment pulse-height spectrum must permit the detector bias to be set so that small changes in the amplifier gains and similar electronic effects will not change the efficiency of the fission counter.

In this experiment the preferred bias was about 10 MeV because in that energy region there were few fission pulses so that the count rate was insensitive to changes in the electronics; however, there was greater danger of noise with such a low bias setting. Fission cross-sections were also measured with the bias settings of 20 and 30 MeV, and the ratios of the fission count rates thus obtained were monitored to ensure consistency.

Corrections must also be made for changes in the angular distributions of fission fragments with energy and for centre-of-mass motion.

A. J. DERUYTTER: Were data always recorded with a two-parameter system?

B. C. DIVEN: During the many months when backgrounds were being analysed and optimum experimental conditions selected, pulse heights and times were recorded simultaneously and analysed in an on-line computer. It turned out that some precautions were unnecessary. For example, the room-return background was so small that it was unnecessary to use the two-parameter system to record simultaneously the flight times and pulse heights. During the actual measurements, the time spectra and the pulse-height spectra were recorded but not correlated.

Radiation damage from fission fragments

B. D. KUZMINOV: The stability of the detector with respect to radiation damage is important. In the Soviet Union, we have found it possible to

record up to 500 000 fission fragments with target and detector immersed in the same flux without any noticeable changes in the counting characteristics of the detector. Could Mr. Diven comment on effects of radiation damage on the detector?

B. C. DIVEN: Two considerations turned out to be important in selecting the type of fission detector. First, the thickness was important because the detector was so close to the fission foil that scattering effects could have been significant. Second, the lifetime of the detector under bombardment by fission fragments was important.

The part of the experiment most damaging to the detector was the calibration in which the count rate of the detector was compared with that of a low-geometry fission counter in a thermal neutron flux. The low-geometry counter counted 100 times more slowly than the one to be calibrated so that the latter necessarily had to experience a very large number of fission events. The type of detector used can take some tens of millions of fission events without noticeable deterioration.

During the actual measurements the detector is monitored continuously by recording not only all of the fission fragments but also the alpha-particle peak. These various ratios are watched carefully to see that they stay constant. So far, in this experiment it has been necessary to change the detector once. That is why the calibration experiment was repeated once.

Anisotropy of the fission fragment distribution

R. W. PEELLE: How large were the corrections for anisotropy of fission fragments and for centre-of-mass motion at an energy of about 10 MeV?

B. C. DIVEN: We have not made the calculation at 10 MeV yet, and in that energy range it will be difficult because I know of no data on the angular distributions of fission fragments between 8 and 14 MeV. Between 1 and 6 MeV the errors in these corrections are very small because reasonably good angular distribution data are available and because there is little centre-of-mass motion.

Fission-foil geometry and backing

C. D. BOWMAN: Since a large part of the mass of ^{235}U is near the outside of the foil, it is important in 2π geometry to have the foil close to the solid-state detector. How close was it actually?

B. C. DIVEN: The distance between foil and detector was less than 1 mm but I do not remember the exact distance. The detector was significantly larger than the diameter of the fission foil. If the fissile material had zero thickness, so that there was no absorption in the ^{235}U, then the efficiency should have been 95%.

R. W. PEELLE: You mentioned that the standard foil used in the calibration experiment had a stainless-steel backing while all the other foils associated with the measurements had heavy-element (platinum) backings. Fission fragments which leave the uranium deposit in the direction of the backing interact differently with the different backings. Does the difference in backings affect the calibrated efficiency of the 2π fission counter?

B. C. DIVEN: I think the effect of the backing would not be important in a low-geometry system, and this is one of the reasons that the calibration experiment was done in low geometry. Let me clarify the procedure.

The measurements with the Van de Graaff are done with a particular fission counter of nearly 2π geometry, and that counter, including its foil, is never disturbed in any way until it fails and becomes useless. In the calibration experiment with thermal neutrons, a standard foil on a thin backing is placed adjacent to the fission chamber and counted in low geometry. Thereby the 2π fission chamber with all its bad scattering effects is calibrated against a standard foil counted in low geometry.

GENERAL DISCUSSION

OF STRUCTURE IN SYSTEMATICS OF ^{235}U FISSION AROUND 1 MeV

J.J. SCHMIDT: I would like to inquire about possible physical or theoretical reasons, such as channel effects, for structure in the ^{235}U fission cross-section around 1 MeV.

B.D. KUZMINOV: I think it is possible that some channel effects appear in this energy range. In this same energy range there are some irregularities in $\overline{\nu}$. If one analyses the variation of Ko^2, the square of the average value of the projection of the angular momentum on the fission axis, one finds corresponding irregularities. The possibility that such variation in the fission cross-section may be explained by channel effects is not excluded.

F. KÄPPELER: A possible explanation may be that this step-like increase marks the end of the energy gap in the compound nucleus ^{236}U and is due to the onset of single-particle states. This is probable because the angular anisotropy of the fission fragments shows a correlated decrease in anisotropy at the same energy. As Mr. Kuzminov mentioned, structure is also observed at the same energy in $\overline{\nu}$ and in the average kinetic energy of the fission fragments.

W.P. POENITZ: I looked at the reported fluctuations in $\overline{\nu}$. The bump, the deviation from smooth variation of $\overline{\nu}$ with incident neutron energy, seems to appear around 400 keV but has largely disappeared at about 1 MeV where the cross-section begins to rise.

There may be one, and possibly two, other 'steps' in the fission cross-section before it begins to decrease around 2 MeV.

J.L. LEROY: Should one expect a correlation between structure in $\overline{\nu}$ and structure in the fission cross-section also below 100 keV?

E. MIGNECO: I think there are two quite different phenomena which may account for various fluctuations in the fission cross-section. At energies lower than about 10 keV, fluctuations are due to the effects of the double-humped fission barrier. At high energies, about 1 MeV, one would not expect to see a peak because the damping of any new channel should be strong, and any resonance-like effect would be strung out over a large energy interval. A 'step', however, would be understandable.

J.W. BOLDEMAN: A review such as that of Manero and Konshin[1] shows that there may possibly be several humps in the $\overline{\nu}$ curve below 2 MeV. However, careful examination shows that there are inconsistencies among the various data sets. Values of $\overline{\nu}$ derived from measurements of the kinetic energy of the fission fragments show a different divergence from linearity than direct measurements. The discrepancies among the measurements suggest unusual effects, but it is difficult to determine exactly what is happening.

One trouble with the early interpretation in terms of channel effects was a mistake in the determination of the fission threshold of ^{235}U based on (d, p) fission studies. Since the ground-state of ^{235}U has negative parity, only compound nuclear states of negative parity were populated. It was very difficult in that experiment to see the ground-state for fission, which is a positive parity state. The irregularity in $\overline{\nu}$ was interpreted by some people

[1] MANERO, F., KONSHIN, V., At. Energy Rev. 10 (1972) 687.

as being due to the availability of a K^{2+} state; others thought that there were only a couple of fission channels available and that the irregularities in $\overline{\nu}$ were due to a change from s-wave to p-wave fission. These arguments no longer hold because it is now known that the fission threshold is 600-700 keV lower.

For ^{233}U we have measured both the kinetic energy of the fission fragments and $\overline{\nu}$; we observed some strange structure in both as a function of neutron energy. The structure in these parameters seems to fit well with the change in probability for s- and p-wave fission.

We thought that if we were going to see channel effects in ^{235}U, they would be similar to those in ^{233}U. The bump in $\overline{\nu}$ for ^{235}U, which occurs at 400 keV should really occur about 300 keV lower, but we observed no rapid rise around 100 keV.

The results of Meadows and Whalen[2] are the ones which require structure in $\overline{\nu}$ for ^{235}U in order to be correct. I think that Mr. Soleilhac's new results suggest structure but could be explained without it.

M. SOLEILHAC: It is difficult to think of structure in $\overline{\nu}$ for ^{235}U in addition to that at 400 keV, which is found in our results of 1970[3] and in those of Meadows and Whalen[2]. Since the $\overline{\nu}$ data are statistically much less precise than the fission cross-section data, I do not think it possible to make meaningful correlations between structure in $\overline{\nu}$ and structure in the fission cross-section. It is important to study other systematics such as variation in kinetic energy of the fission fragments and in gamma emission.

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[2] MEADOWS, J.W., WHALEN, J.F., J. Nucl. Energy 21 (1967) 157.

[3] SOLEILHAC, M., FREHAUT, J., GAURIAN, J., MOSINKI, G., Nuclear Data for Reactors (Proc. Conf. Helsinki, 1970) 2, IAEA, Vienna (1970) 145.

IV. FISSION AND CAPTURE STANDARDS

B. Fast neutron capture cross-sections of ^{197}Au and other appropriate capture standards

MEASUREMENTS OF THE CAPTURE CROSS-SECTION OF ^{197}Au BETWEEN 75 keV AND 500 keV

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Abstract

MEASUREMENTS OF THE CAPTURE CROSS-SECTION OF ^{197}Au BETWEEN 75 keV AND 500 keV.

The capture cross-section of ^{197}Au has been measured between 75 keV and 500 keV by two methods. (1) Prompt γ-ray cascades immediately following neutron capture were detected with a liquid scintillator. The total-energy weighting technique of Maier-Leibnitz was used to render the detector response insensitive to details of the γ-ray cascade. Neutrons were produced with a pulsed Van de Graaff using the $^{7}Li(p,n)^{7}Be$ reaction, and neutron flux was monitored with a ^{10}B - NaI(Tl) detector. (2) In the activation measurements, neutrons were produced with a continuous proton beam on a lithium target by the $^{7}Li(p,n)^{7}Be$ reaction. The neutron flux was monitored with a BF_3 counter of known efficiency. The activities of gold samples irradiated at different distances from the neutron source were determined absolutely by β-γ coincidence counting and relatively by γ-counting. The measurements by the two methods disagree for unknown reasons. The cross-sections obtained by prompt gamma counting are more consistent with existing data and at present appear more reliable than the activation data.

1. INTRODUCTION

(E. Fort).

Before a cross-section can become a working standard, it has to be measured by as many different techniques as possible.

The cross-section of $^{197}Au(n,\gamma)^{198}Au$ can be determined by the three following techniques:

(a) Spherical shell transmission method. The capture cross-section is directly related to neutron absorption. This method has been rarely used with gold (Schmitt and Cook [1], Belanova [2]) because of multiple scattering and self-shielding (Froehner [3, 4], Semler [5]) for which an exact knowledge of the total and elastic cross-sections is needed. Systematic errors can develop when processes other than radiative neutron capture occur.

(b) Detection of γ-ray cascades which immediately follow neutron capture. These methods entail the use of detectors which are independent of the decay scheme. This independence is obtained either by giving the detectors an efficiency as near as possible to 100% (large detectors: Diven et al. [6], Gibbons et al. [7], Haddad et al. [8], Iliescu et al. [9], Carlson et al. [10], Poenitz et al. [11] or by making the efficiency proportional to the total gamma energy (neutron energy + binding energy). This is the principle of the Moxon-Rae detector (Moxon and Rae [12], Macklin et al. [13]) and also of the Maier-Leibnitz detectors because of a weighting function applied at each pulse. The weighting function is dependent only on the pulse size (Macklin and Gibbons [14], Czirr [15],

Le Rigoleur et al. [16]). The Maier-Leibnitz detectors have a greater efficiency than those of Moxon-Rae. All detectors of this type provide a fast response, so they can be used in conjunction with the time-of-flight technique. If $(n,n'\gamma)$ processes are properly accounted for, the combination of these methods is capable of producing the most reliable cross-section values and the most accurate energy values (Le Rigoleur et al. [16]).

(c) Activation methods can be applied to gold because of a decay constant of 2.698 days and because of the favoured transition (99%) in the decay of $^{198}Au \xrightarrow{\beta^-} {}^{198}Hg$. The two techniques generally employed are absolute measurements of the β^--activity (β^- of 962 keV) or the γ-activity (412 keV) (Harris et al. [17], Miskel et al. [18], Lyon and Macklin [19], Cox [20], Johnsrud et al. [21], Gibbons et al. [22], Bame and Cubitt [23], Weston and Lyon [24]). This method can be easily applied. Since only small amounts of material are required, corrections for multiple scattering and self-shielding are small. The method is suitable for energies approaching thermal energy. At higher energies it is less useful because it gives mean values corresponding to a wide spectrum of incident neutrons. We describe recent work carried out at Cadarache using (i) measurements of the γ-activity following neutron capture by means of a total-energy detector, and (ii) the activation method.

2. CAPTURE MEASUREMENT FROM PROMPT γ-COUNTING

(C. Le Rigoleur, A. Arnaud, J. Taste)

2.1. Principle of the method

In these measurements we used the total-energy weighting technique proposed by Maier-Leibnitz [25] and first used by Macklin and Gibbons [26].

The efficiency of the detector for capture γ-rays is proportional to the total energy released, i.e. neutron energy plus binding energy:

$$E_n + B_n = \sum_{i=1}^{m} \gamma_i \qquad (1)$$

The detector response was rendered insensitive to details of the cascade of individual γ-rays following neutron capture by applying a weighting function W(I) to each pulse from the detector. This weighting function, which is a function of pulse size only, is defined as follows:

$$\sum_{I=1}^{N} P(E_\gamma)S(I,E_\gamma)W(I) = E_\gamma \qquad (2)$$

$P(E_\gamma)$ is the probability for detection of a γ-ray of energy E_γ, and $S(I,E_\gamma)$ is the probability to have a pulse of amplitude I if the γ-ray has been detected.

First, the pulse-height response $P(E_\gamma)\,S(I,E_\gamma)$ of our detector for a γ-ray of energy E_γ was calculated with a Monte-Carlo code and then W(I) was calculated.

The validity of this theoretical weighting function W(I) has been checked with calibrated γ-sources (^{51}Cr, ^{137}Cs, ^{54}Mn, ^{22}Na, ^{60}Co, ^{24}Na). It was

found that W(I) had to be corrected by 2.5% in order that the energy of the sources can be calculated correctly with Eq.(2). For energies below 3 MeV, W(I) was established with 1% precision and for energies above 3 MeV it was estimated with 2% precision. Strictly, the method is applicable to samples of separated isotopes or to isolated resonances, as for instance to gold, which is monoisotopic.

2.2. Experimental set-up (see Fig. 1)

The data were obtained with time-of-flight techniques. Pulsed protons (3.5 MHz), bunched (~ 1.2 ns) full-width-at-half-maximum (FWHM), accelerated by the 5.5-MeV Van de Graaff at Cadarache, interacted with ^{7}Li to produce 1.2 ns (FWHM) neutron pulses with a broad energy spectrum. Neutrons at 0° were passed through a collimator of ^{6}LiH, Li_2CO_3 plus paraffin, and lead. Gold samples, 25 mm dia., were exposed at the centre of the prompt γ-ray detector at a distance of 85 cm from the target. The neutron beam covered a transverse area of 28 mm dia. The γ-ray detector was a 1.441-litre C_6F_6 liquid scintillator contained in a quartz cell. The experimental time resolution was better than 1.8 ns.

The neutron flux was measured with a ^{10}B, NaI(Tl) detector at a distance of 170 cm from the ^{7}Li target. The detector had an experimental time resolution of 2.5 ns. The efficiency of this detector was measured by comparing it with a flat-response detector of well-known efficiency (1.8%) [27].

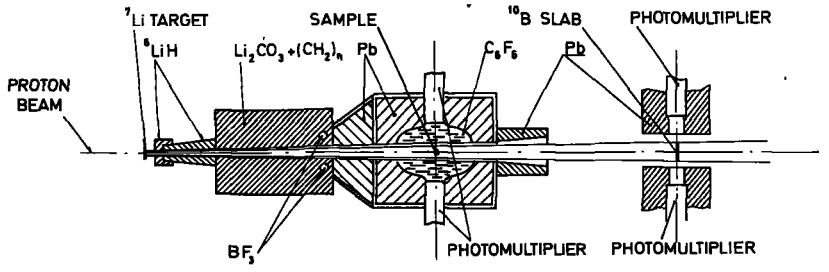

FIG. 1. Experimental set-up for measurement of the ^{197}Au(n,γ) cross-section by detection of the prompt gamma cascade which immediately follows neutron capture.

The parameters time and pulse height were digitized for each neutron event and transmitted to a CII 90 10 computer for on-line processing. The events were sorted according to the identification code of the γ-ray detector or neutron flux detector and were stored. For the events coming from the γ-ray detector, W(I) was stored in the time spectrum of this detector instead of I, the amplitude of the pulse from the detector.

2.2.1. Samples

The gold samples (99.99% purity) used were discs of 25 mm dia. and 1 mm or 0.5 mm thickness. For background determination, carbon samples were used, the thickness of which was chosen so as to match the scattering from the gold samples to be analysed.

2.2.2. Analysis

Time-of-flight spectra were transformed into energy spectra with energy intervals ΔE. Non-linearities in the time digitization and photon flight time were taken into account. An uncorrected neutron radiative capture cross-section (in barns) can be calculated as follows:

$$\sigma_{n,\gamma}(E) = \frac{\Omega_2}{\Omega_1} \frac{1}{n} \frac{\epsilon(E_n)}{N} \frac{S_T(E_n)}{E_n + B_n} \qquad (3)$$

where

Ω_2 = the solid angle of the ^{10}B slab subtended at the ^{7}Li target

Ω_1 = the solid angle of the gold sample subtended at the ^{7}Li target

n = thickness in nuclei/barn of the gold sample

$\epsilon(E_n)$ = efficiency of the ^{10}B NaI(Tl) detector at energy $E_n + \Delta E_n/2$ (efficiency known with 2.5% precision)

N = count rate of the ^{10}B NaI(Tl) detector in the energy range $(E_n, E_n + \Delta E_n)$ after correction for the attenuation of a few per cent by the capture sample (calculated from the total cross-section)

$S_T(E_n)$ = calculated count rate, corrected for background measured with the carbon sample, of the γ-ray detector in the same energy range

B_n = binding energy of the neutron in ^{198}Au

Calculated corrections were applied for the following sample effects:

(a) Neutron scattering and resonance self-shielding. Analytical calculations adjusted by Monte-Carlo calculations at several energies were used.

(b) γ-attenuation and non-linearity of the weighting function. Our γ-ray detector has a high efficiency (34% for γ-rays of 1 MeV, 20% for γ-rays of 4.5 MeV) so that two γ-rays from neutron capture cascades can be detected simultaneously. Because of the non-linearity and the shape of the weighting function, more weight is assigned to the sum of two pulses than would be to each of them separately.

We have calculated the excess weight assuming that the spectral distribution of the γ-rays in the cascade has the same shape and multiplicity at neutron energy as at thermal energy. γ-attenuation in the gold sample is also related to spectral distribution. The γ-attenuation for individual γ-rays of energy E_γ was calculated with a Monte-Carlo code. The effect of both corrections was calculated. Calculation made with drastic changes in the shape and multiplicity of the spectral distribution led to confidence in the calculated corrections. It is estimated that these corrections introduce an additional error of 1.6%.

(c) Several other minor corrections were applied, e.g. for scattering of neutrons in air and for γ-rays from inelastic scattering of neutrons in gold. The agreement between the results obtained with the 0.5-mm-thick sample and the 1-mm-thick sample gave some confidence that corrections dependent on the thickness of the gold sample were calculated correctly.

2.3. Results and comparison with other data

Figure 2 gives the experimental values. Typically, we get 4.5% precision including 2.8% statistical error. Our results agree very well with those of Poenitz [28], i.e. within 1-2%. Extensive comparisons of existing capture cross-section measurements on gold in the keV and MeV energy range have been made. Our results agree well with the evaluation of Poenitz [29] except above 450 keV where we found lower values. One of the interesting results of our measurements is that the gold capture cross-section is not smooth and that structure is observed below 200 keV.

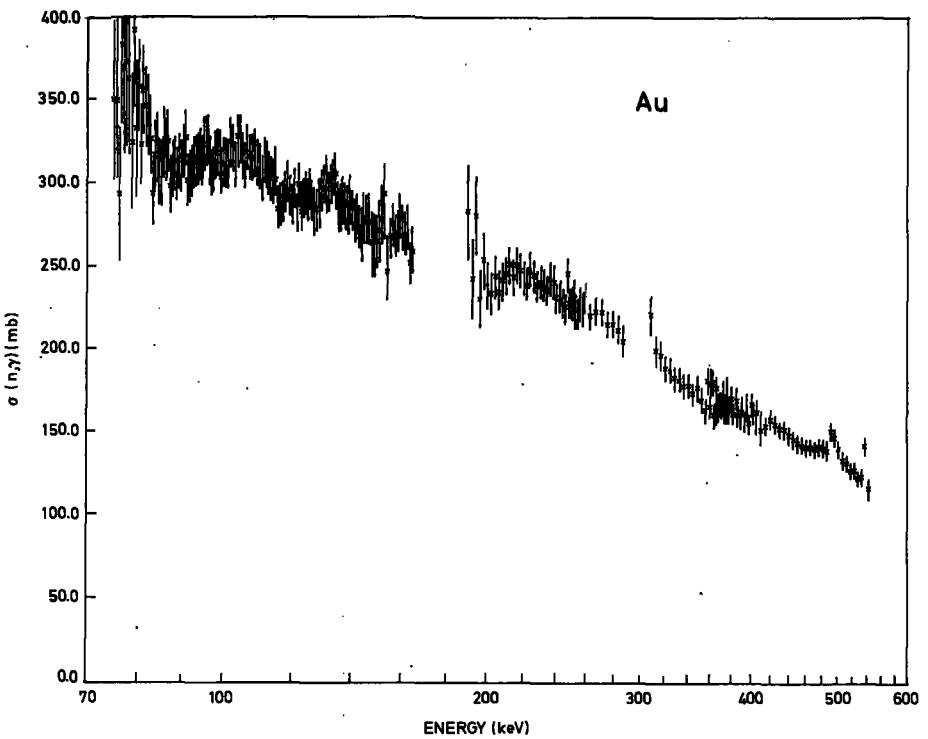

FIG.2. The gold capture cross-section measured in Cadarache by counting prompt capture γ-rays.

3. ACTIVATION METHOD

(E. Fort, J. L. Huet, J. Maloizel)

3.1. Description of the apparatus

Neutrons were produced by means of the $^{7}Li(p,n)^{7}Be$ reaction with a continuous beam. The mean current was about 15 μA.

^{7}Li was evaporated in vacuum on a copper backing (0.2 mm thick) or a tantalum backing (0.5 mm thick). The thickness of the evaporated ^{7}Li deposit is between 10 and 15 keV. The target was mounted on a rotating support and was air-cooled. A diaphragm of 5 mm dia. was placed 50 cm in front of the target. In front of the diaphragm, on the trajectory of the protons, was mounted a system of 6 tungsten wires, 0.35 mm in dia., placed at 60° angles from each other. Each wire penetrated more or less deeply into the proton beam and collected a certain amount of current. By means of a recorder, the value of this current could be established as a function of time. Thus it was possible to have at each moment an approximate idea of the charge distribution in the proton beam. The gold samples were discs of 16 mm dia. and 0.1 mm or 0.05 mm thick. The samples were placed at 20° with respect to the proton beam, on different trajectories. The sample assembly is made of hollow steel tubes (exterior diameter: 3 mm, thickness: 0.1 mm) and allows simultaneous irradiation of three gold targets (1), (2), (3), placed respectively at distances of 5 cm, 10 cm and 15 cm from the target (see Fig. 3). The neutron flux was measured by a directional BF_3 counter

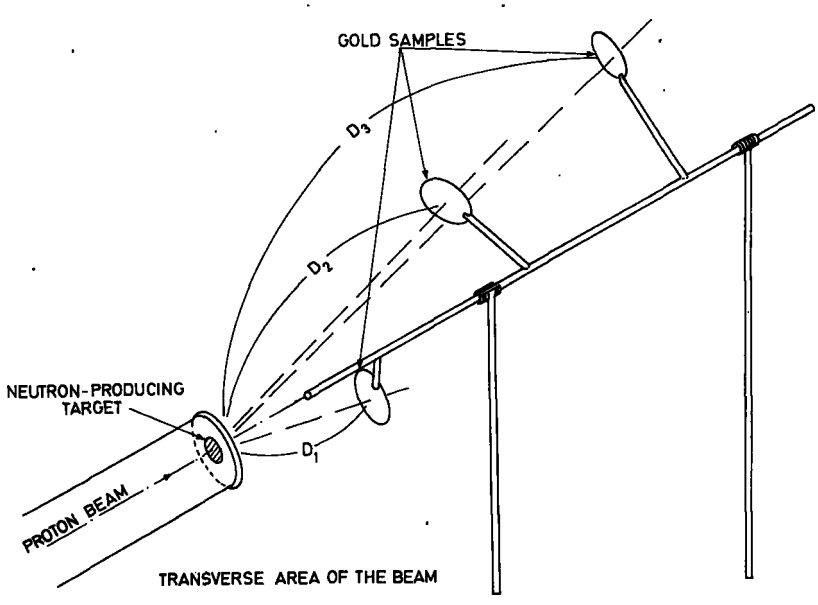

FIG. 3. Experimental arrangement of samples for measurement of the gold capture cross-section by the activation method.

with flat response whose efficiency had been previously determined by the $MnSO_4$ bath technique and also by the 'associated-particle' technique using the $T(p,n)^3He$ reaction. This reference detector was placed at 1.72 m from the target and at 20° with respect to the proton beam. The neutron flux was recorded on a multi-scale basis — each time unit was 15 mm.

The activity of sample (1) was absolutely determined by β-γ coincidence counting. The β's were counted with a proportional counter filled with pure methane. The γ-counter was composed of two NaI(Tl) scintillators with photomultipliers operated in coincidence. The activities of the samples (2) and (3) were measured relative to sample (1) (by γ-counting) or absolutely as in the case of sample (1).

The activities of samples (2) and (3) were needed for determining the contribution of background neutrons to the activity of sample (1), assuming that background flux does not depend on the sample position. This hypothesis was verified by the relative count rate of the samples and also by a supplementary experiment. This experiment involved the determination by the time-of-flight technique of the response, according to the distance, of a ^6Li glass scintillator and of a thin-walled grid ionization chamber containing boron. The response of these two detectors varies according to energy, much in the same way as the (n,γ) gold cross-section. The scintillator was used for distances greater than 10 cm and the ionization chamber for shorter distances.

3.2. Corrections

Corrections were applied for the following effects:

(a) Neutron scattering in air. Scattered neutrons affect the BF_3 counter. The mean value of the correction was about 6%.

(b) Target effect. The incident flux N(E) is contaminated by neutrons scattered by the backing, at energies $E' < E$. The scattered neutrons have a spectrum $n(E')$. The correction can be written as:

$$\frac{\int_{\Delta E'} n(E')\sigma(E')\,dE'}{\int_{\Delta E} N(E)\sigma(E)\,dE} \Bigg/ 1 + \frac{\int_{\Delta E'} n(E')\,dE'}{\int_{\Delta E} N(E)\,dE}$$

Values of the correction were calculated from the curve given by Vaughn and Grench [30] for $\sigma(E)$ and from the spectrum $n(E)$ calculated by Filippi [31] using a Monte-Carlo method and were between 3% and 11%.

(c) Multiple scattering in gold targets. This effect was calculated by Taste [32] using a Monte-Carlo method. Values of 0.6% for samples 0.05-mm thick and of 1.4% for samples 0.1-mm thick were obtained.

(d) Effect of the finite dimensions of the proton beam. This effect concerned the ratio of the solid angles. It was evaluated from data obtained with the apparatus described in section 3.1 in the following way:

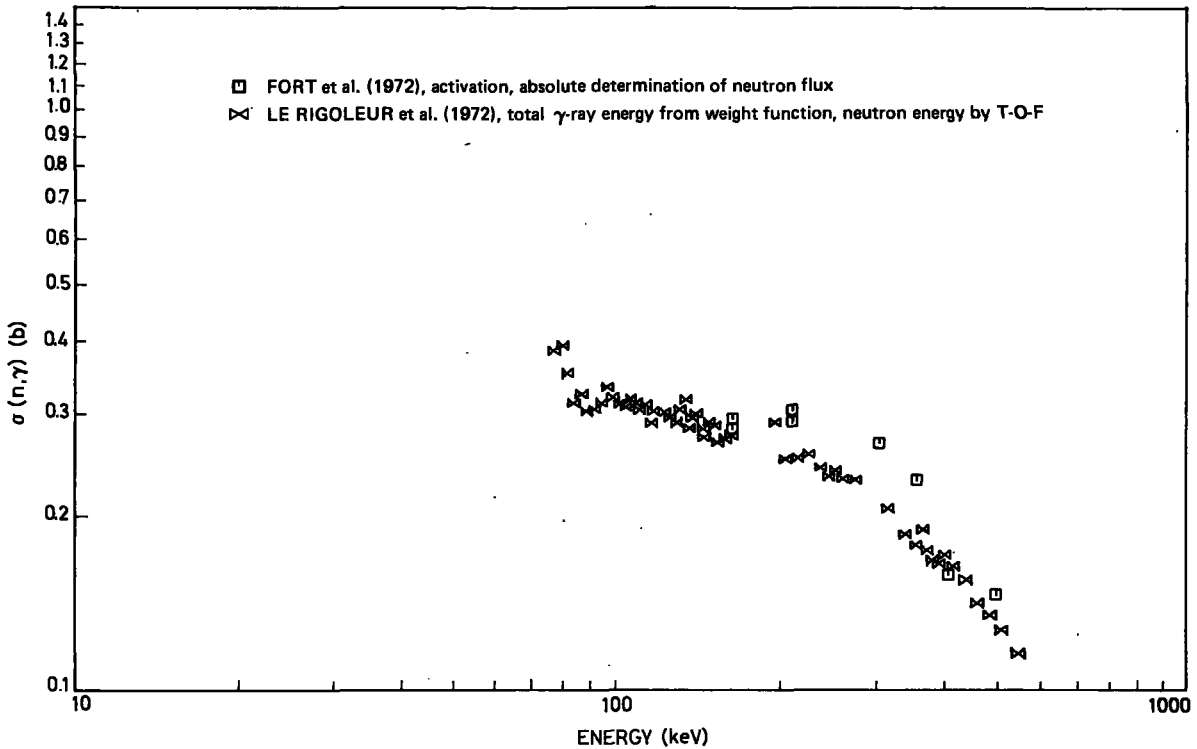

FIG.4. Comparison of the capture cross-section of gold measured at Cadarache (1) by counting prompt capture γ-rays and (2) by the activation method.

The transverse area of the beam was divided into six parts. The charge density inside each part was believed to vary lengthwise along a ray. The shape of the linear variation was the same for each part and was determined by the value of the total current.

We have also taken into account the time distribution of the neutron spectrum and the decay of ^{198}Au. This correction fluctuated between 0.05% and 1.5%.

3.3. Results

From the activation measurements were obtained values of the gold capture cross-section averaged over the incident neutron spectrum. The shape of the incident spectrum was calculated from the measured thickness of the ^{7}Li target using kinematic laws. The effect of the distribution of incident neutron energies on the activity observed at the end of the irradiation period has been taken into account. The energies reported here are the most probable values, and the uncertainties are the half-widths of the distributions at half maximum.

Uncertainties in the cross-section values obtained from the activation measurements are as follows:

(a) Uncertainties concerning the measurement of the target-to-detector distance are evaluated at 2%, thus giving a 4% error on the cross-section.

(b) The uncertainty of the count-rate measurement of the irradiated sample varies between 1% and 4%.

(c) The uncertainty of the count rate of the reference detector is equal to 2%.

If a 10% error is assumed for the correction factors (section 3.2), a mean error of 5% for the cross-section values is obtained by combining the partial errors quadratically.

4. CONCLUSIONS

The capture cross-section of ^{197}Au has been measured at Cadarache by two different methods, (1) by detection of prompt γ-ray cascades and (2) by activation. The results are compared in Fig. 4. Activation measurements were made at neutron energies of 115 keV, 164 keV, 202 keV, 302 keV, 355 keV, 406 keV and 498 keV. The values of the capture cross-section obtained at 202 keV, 302 keV and 355 keV disagree appreciably with values for the same energy range obtained by detection of γ-rays. The Cadarache activation values, however, are reproducible, and checks were made to ensure that the target backing was not responsible for the discrepancy.

Other values obtained by the activation method are plotted with the Cadarache activation data in Fig. 5. Agreement among the various data is poor. In the energy range 160-166.8 keV the apparent agreement between the Cadarache data and those of Gibbons et al. [22] and Harris et al. [17] cannot be regarded as significant because of the serious disagreement at other energies.

The similarity in shape, but not absolute value, of the data of Harris et al. [17] and Cox [20] should be noted. It would help to determine

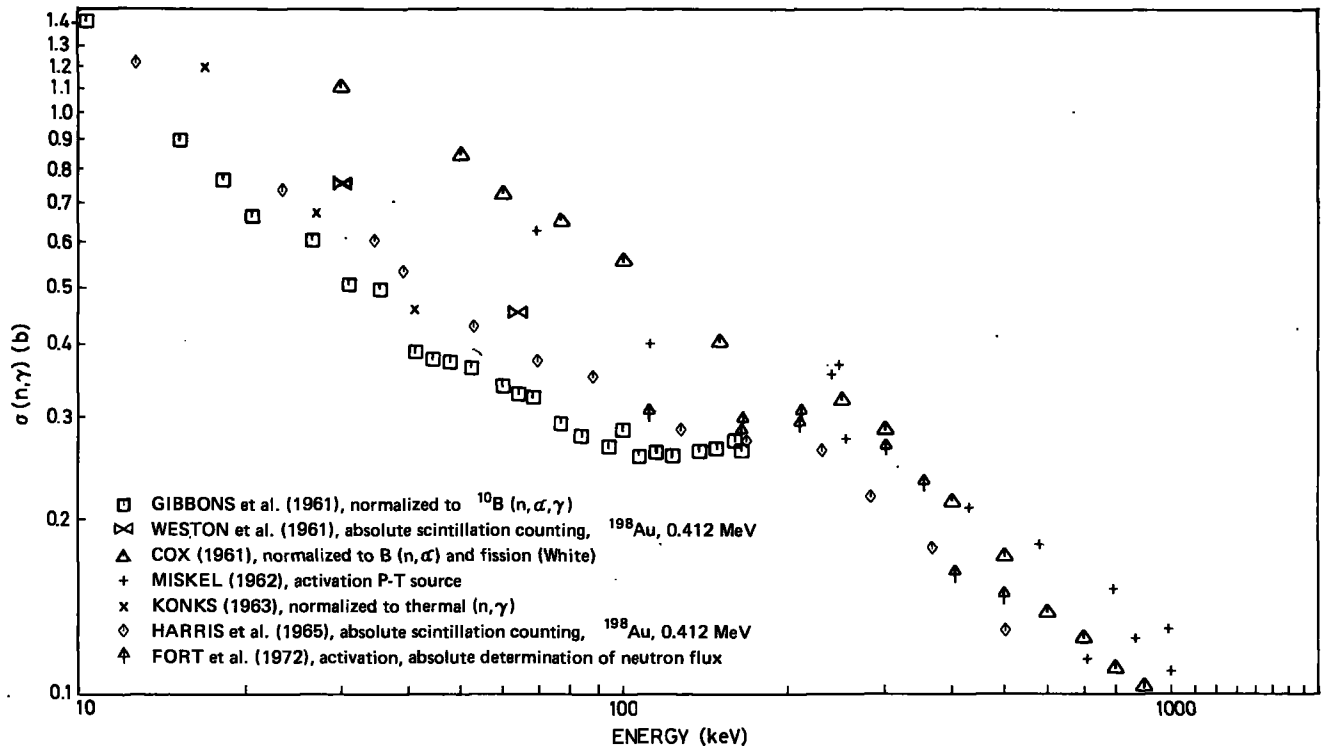

FIG. 5. The capture cross-section of ^{197}Au measured by the activation method.

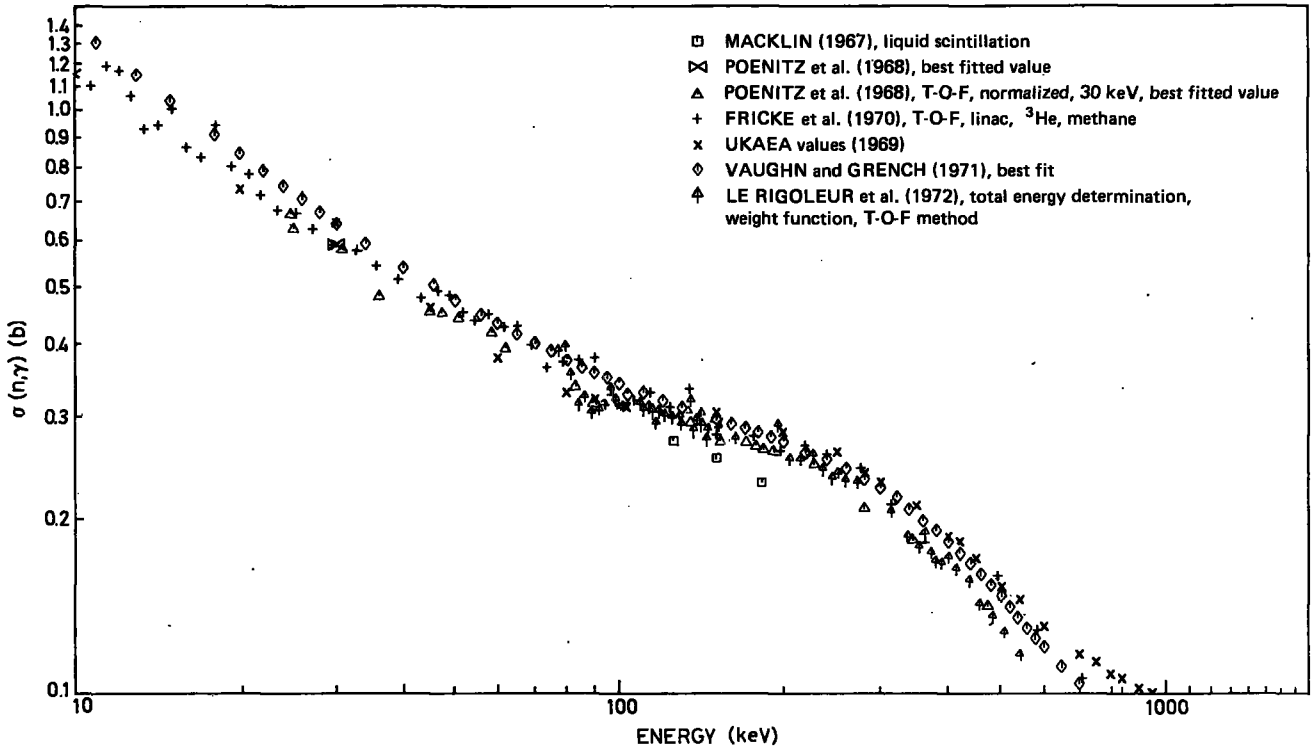

FIG.6. The capture cross-section of ^{197}Au measured by counting prompt capture γ-rays.

the physical origin of the systematic errors. Possible sources of systematic error are the neutron flux measurements and the values used for normalization (Gibbons et al. [22]).

Figure 6 shows that cross-section values obtained from measurement of the prompt γ-rays are more consistent. The values of Fricke et al. [33] (except those above 400 keV) and those of Poenitz et al. [11] are in good general agreement with the present Cadarache data of Le Rigoleur et al. The Cadarache data were obtained with excellent timing resolution and show structure at least in the lower part of the energy range. These fluctuations occur around 30 keV at which energy Poenitz used an averaged value for normalization. The fluctuations show that activation methods do not yield reliable values for normalization at that energy. The fluctuations are probably the cause of the 1-2% discrepancy between the present Cadarache values (Le Rigoleur) and those of Poenitz et al. [11]. Except at lower energies, good agreement exists between the values of Harris et al. [17] (activation method), Fricke et al. [33], Poenitz et al. [11] and the Cadarache values of Le Rigoleur.

As a first attempt to establish practical standard values for the cross-section, the data of the latter four groups of authors could be treated in the manner of Vaughn and Grench [30]. However, better agreement between data obtained by activation methods and data obtained by direct measurements of the radiative capture cross-section is required in order to establish reliable standard values of the gold capture cross-section.

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DISCUSSION

Present accuracy of the ^{197}Au capture cross-section

C. D. BOWMAN: I would like to know how accurately the Cadarache group considers the gold capture cross-section to be known at present and what accuracy they eventually expect to achieve in their measurement programme.

J. L. LEROY: Since we were beginning an extensive programme at Cadarache to measure capture cross-sections of many nuclei, we thought it important to check our techniques by measuring the gold capture cross-section before undertaking measurements on other nuclei.

The accuracy of the data given in section 2 of the paper is 4-4.5%, including 2.8% statistical error, 2% on the flux determination (on the average), 1% on the weighting function and about 1.6% error from other corrections. These data are somewhat preliminary and their accuracy can be improved. If the agreement between these data and other recent absolute measurements using prompt gamma counting is considered, one might conclude that the gold capture cross-section is known to 3-4%.

Before a definitive statement about the accuracy of the gold cross-section is made, I think the discrepancy between the prompt gamma counting data and the activation data should be resolved. One problem with the activation method is to eliminate the effects of slow neutrons. This is possible if the experiment is performed in a very large room. Incidentally, the National Physical Laboratory in the United Kingdom has such a facility, and their activation measurements should be of very high quality. It is possible that a more careful consideration of the effects of slow neutrons may lead to an explanation of the apparent discrepancy of our activation measurements.

L. STEWART: Is anyone planning an experiment in which the cross-section is measured simultaneously by the two methods using the same sample? For example, could the prompt gamma rays be observed at the same time the sample was being activated, with the induced activity being counted at a later time?

J. L. LEROY: We are doing that now.

L. STEWART: What is the highest incident neutron energy at which you expect to be able to make measurements?

J. L. LEROY: We are not expecting to go much above 700 keV where inelastic scattering begins to cause trouble.

Ratio $^{235}U(n,f)$: $^{197}Au(n,\gamma)$ and derived fission cross-sections

J. J. SCHMIDT: Restricting my remarks to the energy range above 100 keV, it seems that the apparent agreement of the new results of Le Rigoleur with those of Poenitz [1] revives an old discrepancy. If the ^{197}Au capture cross-sections are combined with measurements of the ratio $^{235}U(n,f)$:$^{197}Au(n,\gamma)$, the ^{235}U fission cross-section thus derived supports the older measurements of $^{235}U(n,f)$ by Poenitz [2] which were quite low. If measurements of the ratio $^{239}Pu(n,f)$:$^{235}U(n,f)$ are combined with absolute measurements of $^{239}Pu(n,f)$, the derived $^{235}U(n,f)$ cross-section is in much better agreement with other existing $^{235}U(n,f)$ data and with most of the data presented at this meeting. There are no ^{239}Pu-fission to ^{197}Au-capture measurements, and such measurements might be useful. Would anyone care to comment on the discrepancy between the $^{235}U(n,f)$ cross-sections derived by these two methods?

W. P. POENITZ: It is very simple. Either the ^{235}U data, the gold data or the ratios are wrong. All measurements since 1968 which I know of, including the data just presented, confirm our measurements of the ^{197}Au capture cross-section. The ^{235}U fission cross-section is now much better known, and it is lower than previously believed, although not enough so to remove the discrepancy. We have also confirmed the ratio measurements in which, by the way, activation methods have always been used. As a result of much work, all the data are better known, but the discrepancy remains.

Incidentally, I tend to question one of the techniques used by Byer and Konshin [3] for the ^{239}Pu fission cross-section. If the ^{235}U fission data of Szabo [4] or White [5] are used with a set of ratio measurements to derive a ^{239}Pu fission cross-section, it seems reasonable to expect the derived plutonium cross-section to agree with Szabo's [4, 6] experimentally measured ^{239}Pu fission cross-sections, provided the ratio measurements are correct. After all, Szabo's data on both ^{235}U and ^{239}Pu were measured with the same apparatus and flux-measuring technique, and I do not think the ratio measurements should be much in error.

J. L. LEROY: My explanation of the conflicting results is that the ratio measurements involving gold are probably wrong. Some time ago, the most important problem in measuring these cross-sections was to measure the neutron flux, and therefore ratio measurements were considered more reliable than absolute measurements. Today, flux measurement is only one of a number of equally important methods such as absolute fission counting, determination of sample mass, etc. Therefore, two absolute measurements with the same apparatus and flux-measuring

technique are equivalent to a ratio measurement and are equally reliable so that a direct ratio measurement should not be assigned more weight.

T.A. BYER: I would like to reply to Mr. Poenitz concerning the review of the ^{239}Pu fission cross-section by Konshin and myself [3]. Above 300 keV, the data of Szabo et al. [4] are essentially the only absolute measurements which exist. In the 300- to 900-keV region, there is very good agreement, within 2.5-3%, among four or five completely different sets of measurements of the fission cross-section ratio of ^{239}Pu to ^{235}U. When our average fission-ratio curve is used with Szabo's ^{239}Pu data, a fission cross-section for ^{235}U is obtained which agrees with the data of White [5] and of Szabo et al. [4]. But I do not agree with Mr. Poenitz's point that one should necessarily expect such an agreement.

I would now like to comment on Mr. Leroy's remark concerning capture-to-fission ratio measurements. In Sowerby's evaluation [7] of ^{235}U(n,f) using the simultaneous technique, which includes ratio data for ^{238}U(n,γ) and ^{197}Au(n,γ) and for ^{239}Pu(n,f), the recommended curve for the ^{235}U fission cross-section dips rather low at 600-700 keV. When I discussed this with Sowerby, he said that he had not given dominant weight to the ratio of the fission cross-sections of ^{239}Pu and ^{235}U. As a result, his recommended curve tended to be pulled down because of the capture-to-fission ratio measurements. Sowerby was coming to almost the same conclusion as Mr. Leroy, namely that one should perhaps doubt the capture-to-fission ratio measurements but not necessarily the fission-to-fission ratios.

W. P. POENITZ: I think that it is incorrect to say that Szabo's absolute measurements on ^{239}Pu prove that his absolute results for ^{235}U are correct. In an absolute measurement it is necessary to measure the flux, the efficiency of the detectors, the fission rate, and the mass of the sample. If two absolute measurements use the same flux measurement techniques and detectors, then the only new information obtained from the second absolute measurement is a comparison of the fission count rates and the mass determinations. Since the flux measurement and detector efficiencies cancel out, the two absolute measurements are exactly equivalent to one absolute measurement and a ratio measurement. In an evaluation, an equivalent information should be used only once.

T.A. BYER: I understand your point. The idea was to attempt to draw conclusions about the absolute value of, for example, the ^{235}U fission cross-section based not only on absolute measurements of ^{235}U but rather on a much larger number of independent data sets including ratio measurements.

Cross-sections and integral experiments

J.J. SCHMIDT: The results of Monte-Carlo calculations [8] of parameters of small fast-critical assemblies seem to be in much better agreement with experiment when data like those presented here by Mr. Gayther and Mr. Poenitz are used in the calculations. If instead the older results of Poenitz [2] are used with the ratio of the fission cross-sections of ^{239}Pu and ^{235}U, for which there are about ten experiments in agreement, then the multiplication factor is not predicted correctly for any plutonium fast-critical assembly.

To me this seems to be additional indication that Szabo's absolute fission cross-section measurements for ^{239}Pu, the ratios of the fission

cross-sections of ^{239}Pu to ^{235}U, and the newer values of the ^{235}U fission cross-section are correct and consistent. I think this agreement lends support to Mr. Leroy's suggestion that either the capture-to-fission ratio measurements or the capture measurements themselves are wrong.

W. P. POENITZ: I am glad that the question of integral measurements has arisen because I doubt that integral experiments can give significant information about differential data. As we all know, White [5] reported a low value for the ^{235}U fission cross-section at 5.4 MeV, and some re-evaluated Los Alamos data later agreed with his value. As a result, all evaluated data files contain low values for the ^{235}U fission cross-section over most of the whole high-energy range. In the Monte-Carlo calculations by Benzi et al. [8], essentially unmoderated fast-critical assemblies with very hard spectra were considered. It therefore seems difficult to draw conclusions about differential data in the energy range around 1 MeV and below, when the cross-section data used to cover the most intense part of the fission neutron spectrum in these critical assemblies were values which we now believe to be much too low.

I do not doubt that my old, low values for ^{235}U fission produce low values of K_{eff}. However, my old data had quite large errors assigned to them. If the old, high data in the high-energy range above 1 MeV had been used with my older low data at the upper limits of their assigned uncertainties, then values of K_{eff} much closer to unity would have been obtained.

L. STEWART: Two remarks: (1) Calculations have also been made for fast critical assemblies which have very low energy spectra, and raising the ^{235}U spectrum in the high-energy range will not help to reduce discrepancies in K_{eff} for them. (2) ENDF/B was always 5% above White's value [5] at 5.4 MeV for the ^{235}U fission cross-section.

I think that the ^{235}U fission cross-section in ENDF/B will be reduced at low energies to agree with new measurements, but it will be ^{238}U capture cross-section, not ^{235}U fission, which must be changed most drastically in order to obtain $K_{eff} = 1$ from critical-assembly calculations.

W. P. POENITZ: From critical assemblies, which have soft spectra, it may be even more difficult to draw conclusions about a single differential cross-section because of the many moderating materials which they contain. I think that in particular the effects of capture and inelastic scattering in ^{238}U are causing problems.

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NEUTRON FLUX MEASUREMENT IN THE keV ENERGY REGION
Suitability of indium as an activation standard for keV neutrons

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Presented by E.J. AXTON

Abstract

NEUTRON FLUX MEASUREMENT IN THE keV ENERGY REGION: SUITABILITY OF INDIUM AS AN ACTIVATION STANDARD FOR keV NEUTRONS.

For measuring monoenergetic neutron fluxes in the energy range 100-1000 keV by activation of small discs, the accepted standard material, gold, can be unsuitable because of inconveniently long half-life and insufficient intensity of the induced activity. The suitability of indium as an activation standard for keV neutrons and the required improved knowledge of the ^{115}In capture cross-section are discussed.

The measurement of monoenergetic neutron fluxes in the neutron energy region 100-1000 keV by the activation of small disc samples is a simple and accurate method when the neutron capture cross-section as a function of energy is known. Gold has often been used for these activation measurements, but from an experimental aspect, the 2.7-d half-life of the ^{198}Au produced is much too long for many purposes. Thus, in general, the activity produced is rather low and the gold samples remain active and unusable for several weeks after irradiation.

For recent measurements at the National Physical Laboratory (NPL) on the keV neutron capture cross-section of ^{238}U [1], it was desirable to use an intermediate activation flux standard, and it was decided to use indium rather than gold, because the accelerator runs had perforce to be short (~1 h) and the samples had to be activated daily. The activation of indium foils to form the 54-min ^{116m}In produces far higher relative levels of activity than gold in short irradiations (~1 h) and the foil activity decays quickly enough for the samples to be used again within a few hours. Thus in experiments using an accelerator to produce a point source of 10^8 monoenergetic neutrons $\cdot s^{-1}$, several large indium foils can be quickly activated at distances from 5 to 50 cm from the target and the specific activity plotted against 1/distance^2 in order to estimate the background activity in the usual way, whereas with gold foils the activity at the larger distances from the source would be prohibitively low unless day-long irradiations could be made.

The 54-min indium can be assayed in a $4\pi\beta$ counter, and it is found that the competing reactions are either very small (50-d ^{114m}In, 4.5-h ^{115m}In) or have decayed within several minutes before counting starts. The indium

foils can be calibrated beforehand by β-γ coincidence counting, making allowance for the K-correction, which includes the effects of the complex decay scheme and the interaction of γ-rays in the foil and β-counter used. The half-life is known to about 0.1%.

Thus it follows that the foil activities can be measured absolutely to better than ±1% after allowing for the long-lived component, and the neutron background estimated to the same order of accuracy. Once the neutron capture cross-section has been established accurately, the monoenergetic neutron flux can be measured routinely.

The neutron capture cross-section has to be established before measuring neutron flux. The shape of the cross-section is generally agreed, it falls 40% between 100 and 200 keV and then remains flat to within ±20% up to 1 MeV, but the resonance structure is not well known, nor is the absolute magnitude of the cross-section. Recent measurements at NPL [1] are about 10-15% below the evaluation in BNL-325 [2] with an error of less than ±2% (standard deviation). Clearly, it is most desirable to obtain other confirmatory data before the indium activation method can be used to measure the neutron flux.

REFERENCES

[1] RYVES, T.B., HUNT, J.B., ROBERTSON, J.C. (1972), to be published.
[2] BROOKHAVEN NATIONAL LABORATORY, UPTON, N.Y., Rep. BNL-325, 2nd Edn., Suppl. 2 (1966).

DISCUSSION

W.P. POENITZ: This paper introduces the old question of whether another capture standard should be adopted. One argument in favour of retaining gold is that, to establish another standard to the same reliability, the necessary effort may ultimately be equivalent to or exceed that which has already gone into the measurement of the absolute capture cross-section of gold.

The properties of indium seem to me to be generally suitable for a standard material although there are some disadvantages. One possible inconvenience is that the softness of the metal might cause mechanical problems in the fabrication and use of large, thin foils.

Another possible complication is that the activation cross-section of indium is not identical with the capture cross-section as is the case with gold. In addition to the 54-min state of ^{116}In, a higher-energy 2.16-s level, which decays entirely to the 54-min level, and the ground-state of ^{116}In, which decays differently than the 54-min level, are also excited.

If standards are selected according to their importance in practical applications, perhaps ^{238}U capture would be a useful standard. A possible disadvantage is that, because of the low neutron binding energy, the energy available to a gamma-ray cascade following capture is very low.

E.J. AXTON: In the work which I just described where the neutron flux was measured as a function of distance in order to estimate the background activity, indium was used in preference to ^{238}U because ^{238}U samples

had to be chemically separated every time before they were used and could only be used for a few hours after chemical separation. In this experiment, gold could not be used for the reasons stated in the paper.

J. L. LEROY: If there is great interest in indium as a capture standard, we would want to include it in our programme of capture measurements which I mentioned previously.

IV. FISSION AND CAPTURE STANDARDS

C. The value of $\bar{\nu}$ for the spontaneous fission of ^{252}Cf

THE VALUE OF $\bar{\nu}$ FOR ^{252}Cf

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Abstract

THE VALUE OF $\bar{\nu}$ FOR ^{252}Cf.

A number of absolute measurements of $\bar{\nu}$ for ^{252}Cf have been reported with reputedly high accuracy. At the time of the IAEA review in 1969, the results appeared to divide into two distinct groups at about 3.7 and 3.8 neutrons/fission. In the intervening period, some new measurements have appeared and the older measurements have been subjected to an objective scrutiny in an attempt to find an explanation for the discrepancy. In this paper, a brief description of each experiment is given and in some cases suggestions are made for revising the value and/or the estimated uncertainty. The measurements now appear to form a consistent set with a weighted mean of 3.733 ± 0.008 neutrons/fission.

1. INTRODUCTION

Absolute values of $\bar{\nu}$, the average number of neutrons emitted per fission, have been requested at high accuracy by reactor designers for ^{233}U, ^{235}U and ^{239}Pu. The use of ^{252}Cf as a standard in this field has many advantages; for example, the spontaneous fission half-life is such that sources of negligible mass can be made, and absolute neutron emission rates can be measured in ideal surroundings far from the disturbing influences of reactors and accelerators. A number of absolute measurements with reputedly high accuracy have been reported and in 1969, when the IAEA review was published [1], the results appeared to divide into two distinct groups at about 3.7 and 3.8 neutrons/fission. In the intervening period, some new measurements have appeared, and the older values have been subjected to an objective scrutiny in an attempt to find an explanation for the apparent discrepancy. This paper gives a brief description and criticism of each experiment, and in some cases suggestions are made for revising the value and/or the estimated uncertainty. The measurement techniques fall into two categories:

(a) Delayed coincidence experiments in which a neutron 'gate' is opened for a finite time after each detected fission event. This method has the advantage that no absolute fission counting is required and that the same equipment can be used subsequently to measure $\bar{\nu}$ for neutron-induced fission. However, in the case of liquid scintillators, there is the disadvantage that intermediate stages of the experiment (e.g. absolute neutron counting) cannot be confirmed by comparison of sources with other laboratories.

(b) Direct measurements in which absolute fission rate measurements and absolute neutron emission rate measurements are made separately. In this type of measurement, in principle if not in practice, the two separate operations can be the subject of external comparisons.

The values and uncertainty estimates derived in this paper have been accepted as provisional input data for the current IAEA review of 2200-m/s constants, pending further information on the problems discussed and discussions with the authors.

2. DELAYED COINCIDENCE EXPERIMENTS

2.1. Liquid scintillator (Hopkins and Diven [2])

The californium sample is placed in a fission counter situated at the centre of a cylindrical liquid scintillator tank, 1 m long and 1 m dia., light pulses from neutron capture γ-rays being detected by banks of photomultiplier tubes situated in the cylindrical wall. The neutron detection efficiency is determined by replacing the fission chamber with a plastic scintillator and observing neutron capture pulses in delayed coincidence with recoil proton pulses from the scattering of neutrons in the plastic scintillators, in a reaction with $\bar{\nu} = 1$.

The description of the absolute measurement of $\bar{\nu}$ for californium is rather short, forming only two or three pages of Ref. [2a], and consequently there are some aspects of the experiment which warrant further explanation. It appears that the work with the T(d, n) reaction involving neutrons in the 0- to 2-MeV and 6- to 8-MeV energy ranges, described on page 435 of Ref. [2a], is the experimental check of the Monte-Carlo calculations referred to on page 434 of Ref. [2a]. The absolute efficiency on which $\bar{\nu}$ is based is thus determined only from the runs with 0- to 1.3-MeV neutrons using the D(d, n) reaction, as described on page 436 of Ref. [2a]. It would be interesting to know why the efficiency is given as 'about 94%' on page 435, and 'about 86%' on page 436. What is the explanation of this difference?

Further, it is assumed, that the efficiency E is independent of neutron energy except for the effects of variation in the leakage fraction and escape through the axial hole. Whilst this assumption is probably approximately true, it is likely that there is some preferential absorption at the lowest energies in the structural materials of the bath and counting systems. Since there are more low-energy neutrons in the 0- to 1.3-MeV calibration group than there are in the californium spectrum, this would lead to an over-estimation of $\bar{\nu}$. How dependent is the efficiency on the position of the neutron capture? In general, low-energy neutrons will finish up nearer the axis (further from the photomultipliers) than higher-energy neutrons. Isotropic neutrons will finish up nearer the axis than calibration neutrons emitted at 80-90°. Although it is stated on page 1014 of Ref. [2b] that no difference in the capture pulse-height spectrum was observed between calibration neutrons and fission neutrons, it would be necessary to estimate how large the difference would have to be for it to be detected. In any case, this statement applied to a smaller tank with many more light detectors.

The Stockholm group [3] referred to three processes which reduce the neutron energy for a proton pulse of a given size, namely, neutrons scattered by a proton after having been scattered by a carbon nucleus of the plastic scintillator, neutrons scattered by a proton after previous scatter by another proton, and protons escaping from the scintillator

before delivering all their energy. Other effects mentioned by the Stockholm group are: (i) the escape of neutrons after double scattering in the crystal or in structural material, and (ii) detecting α-particles from C(n, α) reactions in the crystal and mistaking them for proton pulses. The latter effect only applies to the part of the experiment concerned with leakage check. It would be desirable to see uncertainty estimates in the form of upper limits for all these possibilities.

The leakage estimate in Ref. [2] of 1.67% for isotropic fission neutrons appears to be too high. At the National Physical Laboratory (NPL), the leakage from the 1-m spherical $MnSO_4$ bath with a spherical cavity of 8.8 cm in diameter is about 0.25%. The leakage from the bath under discussion (1 m dia., 1 m length; axial hole: 7 cm dia.) should be on the one hand greater because of the reduced hydrogen density in the liquid scintillator compared with $MnSO_4$ in water, but on the other hand less because there will be almost no thermal leakage and because the average escape path for a cylinder is greater. The difference does not seem to be accountable by the axial hole. Monte-Carlo calculations have been carried out using the actual dimensions for the bath and the axial hole and the formula of Reines et al. [4] which gave the atom densities as 5×10^{22} cm^{-2} for hydrogen, 3.72×10^{22} cm^{-1} for carbon and 0.003 for Cd/H. Inelastic scattering in carbon was omitted (time did not permit its inclusion) but anisotropic scattering in carbon up to 5 MeV was included by means of Legendre polynomial coefficients which were kindly provided by J. L. Leroy of Cadarache. The neutron energy scale was divided into 200 equal bins and each bin was allocated a number of neutrons proportional to $E^{\frac{1}{2}} \exp(-E/1.43)$ (1.43 being a weighted average of recent measurements of the Maxwellian temperature), such that the total number came to 15 000. These neutrons were then started at random angles to the axis and tracked until they were captured by hydrogen or cadmium or else escaped. The hydrogen capture was about 0.6% of the cadmium capture and the leakage came out at 1.13%. This was reduced to 1.11% by a small correction for the missing carbon inelastic scattering, and it compares with the value of Hopkins and Diven of 1.67%. The run was repeated with a spherical hole of the same diameter at the centre of the bath, giving 0.52% leakage. The difference of 0.6% is attributable to the distortion produced by the axial hole. For the efficiency measurement a rectangular distribution of neutrons from 0 to 1.3 MeV was started off at 72° to the axis and gave 0.05% leakage, compared with the value of 0.4% by Hopkins and Diven. Thus, after making the appropriate corrections and assuming that the efficiency is independent of neutron energy apart from leakage variations, $\bar{\nu}$ should be reduced by 0.22%. However, the leakage values given on page 435 of Ref. [2a] were also checked. Rectangular distributions of neutrons from 0 to 2 MeV at 79° and 6 to 8 MeV at 45.9° were introduced, giving leakage fractions of 0.075% and 4.03% against the estimates of Hopkins and Diven of 0.5% and 6%. If the approximate figures on page 435 of Ref. [2a] are taken as exact (which they are not), one can deduce that the true efficiency after correction for leakage is 0.936 for 0 to 2 MeV and 0.9245 for 6 to 8 MeV.

It would be desirable for the authors to re-examine their experiment with the following purposes:

(a) To estimate upper limits to the various effects and include them as uncertainties, bearing in mind that several of them are 'one way'.

(b) To supply supporting evidence for the assumption that the efficiency is independent of neutron energy and direction apart from leakage and the effect of the axial hole, i.e. to make measurements at more than one calibration energy band and to show that the various efficiencies agree after correction for leakage. Another possibility would be to observe the effect of probing with a γ-source.

The authors accounted for most of the queries raised by demonstrating that the effects were negligible. They re-calculated the leakage correction using slightly different atom densities (hydrogen: 0.0453×10^{24} cm^{-3}, carbon: 0.0396×10^{24} cm^{-3}, cadmium: 0.00009×10^{24} cm^{-3}) and obtained significantly lower leakage fractions, but confirmed their original correction for the difference between californium neutrons and calibration neutrons. A trial run with Reines' mixture confirmed the calculations described above. The authors also made a reduction in $\bar{\nu}$ of 0.3% for the effects of delayed γ-rays. The value published in Ref. [1] was raised by 0.38% because of a change from 1.4 MeV to 1.59 MeV in the value of T. Later evidence (see, for example, Ref. [5]) indicates that the authors' original choice was nearer the truth. Therefore, the authors' original value of 3.771 (prompt) is adopted provisionally, reduced by 0.3% as described above, and not adjusted for the 1969 spectrum change. The authors' estimate of uncertainty of $\pm$ 0.83% is retained.

2.2. Liquid scintillator (Asplund-Nilsson et al. [3])

The principal features of this experiment are the same as in Ref. [2] but there are considerable differences in experimental detail. The scintillator tank was a 60-cm-dia. sphere with a 6-cm-dia. axial hole. The efficiency determination was carried out with an anthracene crystal instead of a plastic scintillator and was measured for many more neutron energies. Also, the authors went into considerable detail in their paper to discuss possible causes of error. The scintillator solution is given as cadmium octoate in tri-ethyl benzene, but the atom densities are not provided.

The crux of the experiment is the determination of the effective efficiency of the liquid scintillator neutron detector for the detection of californium fission neutrons emitted isotropically.

A study of the results of the measurement of this efficiency as a function of energy at fixed angles reveals apparent systematic differences between the sets of results in the four groups of Table I. For example, over the important range from 0.65 MeV to 1.07 MeV, there is a difference of from 1.75% to 2.7% between the Van de Graaff results and those of the low-energy accelerator. A similar situation exists in the 2.5-MeV to 3.01-MeV range between the D-T results and the Van de Graaff results. It appears that the true efficiency could differ by up to 2% in the important region around the californium spectrum peak, and up to 1% in the range 2-5 MeV.

It is probable that some, if not all, of these differences are traceable to differences in leakage. The bath radius is only 30 cm.

TABLE I. LEAKAGE CALCULATIONS FOR EFFICIENCY MEASUREMENTS

Energy (MeV)	Angle	ΔE	Rectangular distribution leakage (%)
0.24	73.6	0.25	0.35
0.65	62.3	0.25	0.75
1.07	53.3	0.25	1.95
0.15	79.5	0.25	0.30
0.50	70.5	0.25	0.35
0.97	62.3	0.25	0.98
1.40	56.1	0.25	2.75
1.86	50.0	0.25	5.45
2.43	42.7	0.25	10.90
2.75	38.6	0.25	13.72
2.93	36.2	0.25	13.68
0.21	77.2	0.25	0.20
0.58	68.5	0.25	0.58
1.00	61.2	0.25	1.45
1.82	49.4	0.25	5.48
3.17	30.8	0.25	17.88
0.48	79.7	0.75	0.35
1.65	70.6	0.75	4.33
1.79	69.7	0.75	4.65
2.18	67.5	0.75	6.80
2.49	65.9	0.75	9.23
3.01	63.3	0.75	11.70
3.46	61.2	0.75	13.03
4.27	57.6	0.75	19.83
5.42	52.8	0.75	27.03
6.69	47.9	0.75	32.57
7.94	43.1	0.75	36.4
10.75	31.9	0.75	48.3

With an axial hole of 3 cm radius the neutrons have only to traverse 27 cm of liquid scintillator to reach freedom, even when emitted at 90° to the axis. Thus the bath does not seem to be large enough to justify the assumptions made by the authors. For example, on page 127 of Ref. [3a] it is stated that "the efficiency of the spherical detector is assumed to be the same in all directions except where the cylindrical channel through the tank influences the symmetry".

To investigate the leakage, Monte-Carlo calculations similar to those described above were carried out using the correct dimensions for the spherical bath and the axial cavity. Unfortunately, the exact composition and density of the liquid are not stated in Ref. [3] so the material used by Hopkins and Diven as specified by Reines et al. [4] was used with an atom density of 5×10^{22} cm^{-3} for hydrogen, 3.72×10^{22} cm^{-3} for carbon, and 0.003 for Cd/H. The density of

Reines' mixture is estimated at 0.94. The solution used here is believed to have a similar hydrogen density and rather less carbon and cadmium. Whilst the leakages from the two solutions will be different, they will not be sufficiently so to affect the argument. The leakage above about 7 MeV will be slightly over-estimated as inelastic scattering in carbon was not included. The following calculations have been made:

(a) The energy scale was divided into 200 equal intervals from 0 to 10 MeV, and each interval was allocated a number of neutrons proportional to $E^{\frac{1}{2}} \exp(-E/1.43)$ (1.43 being an average of recent measurements of this quantity), such that the neutron total was 15 000. These neutrons were started off at random angles to the axis and followed until they either escaped or were captured. The run gave 9.9% for the leakage of isotropic fission neutrons. The calculation was repeated with a spherical cavity of the same radius (3.0 cm) to give 6.6%, the difference being attributable to the effect of the axial cavity.

(b) Neutrons with a rectangular distribution whose width was equal to the error bars in Fig. 3 on page 129 of Ref. [3a] were introduced to assess the leakage for each of the measured efficiency points. The results are shown in Table I of this paper, the points above about 6 MeV having been roughly corrected for the effect of the missing carbon inelastic scattering. The measured efficiency corrected for leakage then appeared to be flat at about 0.78 from 10 MeV down to about 1.8 MeV and then reduced slowly to about 0.73. Table II shows some examples of leakage as a function of angle to the axis for single energies.

There is some external evidence that the leakage fractions were not an order of magnitude too high in these calculations, viz. (i) the same calculation for the Hopkins and Diven 1-m cylindrical tank gave results significantly lower than those quoted by these authors, and (ii) the leakage for isotropic californium neutrons is similar to the measured fraction for the 25-cm radius NPL manganese bath. The latter has a spherical cavity instead of an axial hole, and a greater hydrogen density, but this is offset by greater thermal leakage.

TABLE II. LEAKAGE VERSUS ANGLE

Angle	% Leakage	
	E = 2.18 MeV	E = 1.5 MeV
15	25.13	16.45
30	11.05	5.5
50	8.00	
56.1		2.75
67.5	6.80	
70		2.6
90	6.35	2.1

It is evident from the foregoing argument that certain assumptions made by Asplund-Nilsson et al. [3] are not justified. It is hoped that they will investigate this problem, possibly by extensive Monte-Carlo calculations based on the correct liquid specification. But in any case, with such a small bath, it is problematical whether the small uncertainty of 0.9% could be retained in view of the large leakage corrections involved.

Apart from the main problem of the leakage, it would be desirable to see all the corrections and uncertainties listed. The total uncertainty of 0.9% appears to be made up of 0.77% on the average californium efficiency and 0.05% for the spectrum uncertainty (page 130 of Ref. [3a]) but this does not appear to include, for example, any uncertainty in the 1.5% correction for pile-up (page 126 of Ref. [3a]).

The effective efficiency for californium neutrons will certainly not be a smooth curve because of the effects of the carbon resonances. One wonders whether the efficiency after correction for leakage would be flat or whether there are other effects present. For example, the light collection efficiency may not be the same for neutrons of low energy thermalized near the centre or for neutrons emitted at small angles to the axis. This may explain the fall-off at low energies of the measured efficiency corrected for leakage.

At the bath diameter is only 60 cm, the system will be more sensitive to errors in the mean energy of the assumed californium spectrum. Table III shows the calculated leakage from isotropic californium fission neutrons for various assumed Maxwellian temperatures both for this system and for the 50-cm-dia. cylindrical tank described in Ref. [3a].

TABLE III. VARIATION OF LEAKAGE WITH ASSUMED SPECTRUM

Maxwellian temperature	% Leakage	
	60-cm-dia. sphere	50-cm-dia. cylinder
1.3	8.98	11.88
1.43	9.87	12.79
1.5	10.85	13.60

The authors provided a detailed table of uncertainties, and the quoted $\bar{\nu}$ value was reduced by (0.6 ± 0.3)% for the French effect and by (0.2 ± 0.2)% for the effects of delayed γ-rays. The total uncertainty was expanded to ± 1.5%, pending re-evaluation of the leakage corrections.

Meanwhile, for the purposes of the current evaluation, the authors' original $\bar{\nu}$ value of 3.799 is retained, reduced by 0.8% as described above, and not corrected for the 1969 spectrum change. The uncertainty has been increased to 1.5%.

2.3. Liquid scintillator (Boldeman [6])

A recent measurement by J. Boldeman using the liquid scintillator technique has been reported as

$$\bar{\nu} = 3.735 \pm 0.014$$

This measurement was made with the full benefit of hind-sight, and a well documented account of the experiment is presented in Boldeman's paper IAEA-PL-246-2/33 in these Proceedings. At the consultants' meeting on the 2200-m/s parameters in November 1972 it was decided to accept the measurement and its estimate of uncertainty.

2.4. Boron pile (Colvin et al. [7a, 7b])

In this experiment, the neutron detector was a 220-cm cube of graphite surrounded by a 35-cm reflector of graphite and containing a lattice of 240 BF_3 counters to detect thermalized neutrons. The pile efficiency was determined by another reaction with unit $\bar{\nu}$, namely photodisintegration of deuterium. The system has the advantage (unlike liquid scintillators) that it can be used for independent neutron source comparisons. In fact, an alternative $\bar{\nu}$ result has been derived from sources calibrated by the NPL manganese sulphate bath.

The independent result is based on some very accurate efficiency measurements derived from the D(γ, n) reaction supported by calculations of the pile efficiency as a function of energy carried out by E. Pendlebury of Aldermaston using the Carlsen tin technique. The Pendlebury curve is not a very good fit to the experimental data (see Fig. 2 of Ref. [7b]). No explanation is given for the sharp rise in this curve below 1 MeV.

For the Carlsen calculation the pile was idealized to a homogeneous sphere of graphite, boron-10 and aluminium, the aluminium being used to simulate copper.

As a result of Monte-Carlo calculations, using the actual dimensions for the core and reflector but with the core replaced by a homogeneous mix of 97.8% graphite, 2.2% copper and $1.3 \times 10^{-3}\%$ ^{10}B, the leakage is estimated as 0.56% for the spectrum $E^{\frac{1}{2}}\exp(-E/1.43)$ (1.43 being the average of recent measurements of the Maxwell temperature). The values for the captures are: carbon 4.41%, copper 19.29% and boron 75.74%. The efficiency as a function of energy can be obtained by taking a constant fraction of the boron capture as a function of energy, the constant being a weighted mean of the values obtained from the four experimental points. This constant, applied to the overall boron capture for the spectrum (75.74%), gives 0.64145 for the pile efficiency compared with 0.6428 quoted by Colvin et al., representing an increase of 0.2% in $\bar{\nu}$. A similar calculation can be carried out by correcting the individual measurements for the appropriate leakage and carbon capture and then applying the overall leakage correction at the end. This gives 0.6429 in agreement with the result of Colvin et al. and confirming the value of 3.713 ± 0.015. The alternative value obtained from the source calibration of the pile is 3.700 ± 0.031.

The authors have carefully investigated all the various criticisms that have been made of their experiment and found no cause to change their result or their uncertainty. The original value and uncertainty of Colvin et

al. are therefore retained for this paper. At the consultants' meeting on the 2200-m/s parameters it was decided to retain both values because there are facilities for the fitting programme to accept the common manganese bath uncertainty of ± 0.013 on the one hand and the common boron pile uncertainty of ± 0.009 on the other.

3. DIRECT MEASUREMENTS

3.1. Moat et al. [8a], Fieldhouse et al. [8b]

In this experiment the fission rate of a californium sample is compared with its neutron emission rate using two cylindrical wax detectors of different dimensions, each containing several BF_3 counters for the detection of thermalized neutrons. The authors obtained a value of 3.77 ± 0.07, the more significant uncertainties being due to differences in the energy spectra of ^{240}Pu and ^{252}Cf and to self-multiplication effects. The result has had a rather chequered career. Fieldhouse et al. [8b], after recalibration of the Harwell standard ^{240}Pu source and re-evaluation of the corrections, re-valued the result down to 3.675 ± 0.040, the reduction in the uncertainty reflecting the improvement in neutron source calibrations. In the 1969 2200-m/s revision [1] the value was pushed up by 1.2% to 3.727 ± 0.056 after the ^{240}Pu - ^{252}Cf energy difference had been revised, the total uncertainty now including a component owing to the 1% uncertainty in this correction. The same value and its uncertainty are retained for the evaluation, although there may be arguments, based on spectrum differences, to reduce it slightly.

3.2. Axton et al. [9a, 9b], Ryves and Harden [9c]

This experiment is not complete and the details are incompletely published. Therefore, rather more detail is given in this paper. The experiment is based on separate absolute measurements of the fission rate and neutron emission rate of many californium samples. One problem normally associated with this type of measurement is the incompatibility of neutron and fission counting efficiencies so that a source which will produce adequate statistics in a manganese bath will overload a fission counter if 2π or 4π geometry is used. Consequently, either the neutron source is inadequate or the fission counting must be performed in low geometry.

To obviate this problem, a system of aliquotting is used. From a stock solution of californium chloride purified on an ion-exchange column, a series of small sources is prepared on thin foils and counted in a pill-box type gas-flow proportional counter. If N_1 and N_2 are the respective count rates recorded in the top and bottom 2π sections of the counter, and N_c is the coincidence rate, it can be shown that the true fission rate N is given as follows [9b]:

$$N = N_2 + 2K (N_1 - N_c)$$

where K is a constant which has a value between 0.5 and 1 depending on the method of source preparation. If there is no scattering of fission fragments, then K = 1, whilst K = 0.5 when there is so much scattering that no event

goes undetected on at least one 2π section of the counter. In practice, a straight line is fitted to a plot of N_2 against $N_1 - N_c$, and N and K are obtained from the slope and the intercept. The remainder of the californium chloride is then used as a neutron source for the determination of the neutron emission rate in the manganese sulphate bath [9a]. The technique for the calibration of the bath contains a number of features which ensure the long-term stability and repeatability of neutron source measurements. The bath is calibrated by stirring in a known amount of ^{56}Mn which has previously been standardized by $4\pi\beta$-γ coincidence counting. Each batch of ^{56}Mn sources is counted twice, once by the neutron group on its own equipment and once by the radionuclide standards group. Also, the same solution is used to calibrate a very stable high-pressure γ-ionization chamber. Such calibrations are carried out monthly, and over a period of 8 years the ion chamber calibrations show a spread of less than 0.3%. Thus any weighing errors or malfunction of counting equipment are instantly detected. Also the bath detector system has two independent counting channels. A change in the channel ratio immediately shows up any faults in the bath counting equipment. Finally, the bath solution is circulated continuously through the counters and growth (source in bath) count rates are compared with decay (source removed) results. Deviations of results in the early stages of growth and near the change-over point immediately reveal faults due to change in pumping speed, timing uncertainties, etc.

After the first round of $\bar{\nu}$-measurements had been completed, the solution was opened up to provide more 4π fission sources and a new neutron source for the manganese bath. This process has been repeated up to seven times with the same solution, 10 fission sources being prepared each time. Furthermore, the experiment has been repeated using a solution from four different californium samples. Thus over 20 separate neutron sources and over 200 separate fission sources have been measured. Three of the californium samples were obtained in 1968-1969 which then contained $\sim 70\%$ ^{252}Cf. The other source was an earlier sample which then contained 35% ^{252}Cf. A $\bar{\nu}$-value of 3.70 was originally obtained with the old sample whilst all the newer samples give about 3.725. A recent remeasurement of the old sample appears to give ~ 3.71. It is the resolution of this apparent discrepancy which is delaying the conclusion of this experiment. Meanwhile, for the purposes of this paper, a $\bar{\nu}$-value of 3.725 is adopted, with a 0.5% uncertainty provisionally allocated for fission counting. Hopefully, the uncertainty will be reduced in the final report.

3.3. White and Axton [10]

In this experiment, the fission counting was carried out in low geometry at Aldermaston. The neutron source emission rate was obtained as described above from the NPL manganese sulphate bath. After consultation with White, some adjustments have been made to the estimated uncertainties, as discussed below.

The size of the alpha peak (0.6% - 0.7% instead of the expected 0.3%) can perhaps be explained by multiple scattering of α-particles so that $\sim 0.3\%$ of the particles initially emitted in other directions eventually find their way through the collimator. White is therefore correct not to include this component in his fission estimate, but a further 0.3% uncertainty should be added for the uncertainty in the fission-product spectrum.

The uncertainty on the background variation should be enlarged to 0.6% instead of 0.4%, while that on the solid angle should be 0.5% instead of the 0.15% published in error.

The uncertainty table should now read as follows in percentage:

	Random	Systematic
Neutron measurements	0.2	
Cross-section ratios		0.25 [a]
Efficiency	0.15	0.15 [a]
Escape		0.02 [a]
Charged-particle reactions		0.10 [a]
Source capture		0.05 [a]
Manganese resonance		0.10
Fission measurements	0.2	
Solid angle		0.5
Backscatter		0.5
Self-transfer		0.6
Fission-spectrum uncertainty		0.3
Other		
Delayed neutron fraction		0.03
Chemical and isotopic purity		0.1
Total		1.12
Independent uncertainty		1.03
Common to all NPL bath measurements		0.33

[a] These uncertainties are common to all NPL manganese bath measurements of ^{252}Cf.

The result (3.797 total, 3.788 prompt) is 1.9% higher than the current NPL estimate of 3.725 total. The measurements are therefore not considered to disagree significantly.

3.4. De Volpi and Porges [11]

In this experiment, the neutron emission rate from three californium samples is measured using the 'on-line' manganese bath technique with, in some cases but not all, variable manganese concentration. The fission rate is determined in a separate experiment in which neutrons are detected with a Hornyak detector, the fission counting efficiency being determined from fission-neutron coincidences.

These authors have done a great deal of work in establishing their neutron source calibration facilities in conjunction with their novel method of fission counting for the purpose of measuring $\bar{\nu}$ for ^{252}Cf. It is therefore rather unfortunate that certain disturbing aspects of the neutron source measurements make it necessary to reconsider the estimated uncertainties. The final $\bar{\nu}$-value is heavily weighted by the measurements with fission chamber number 3, the neutron calibrations of which were performed during the two-year period when weighing errors occurred in the dispensation of ^{56}Mn aliquots.

In 1968, as a result of an international comparison of ^{56}Mn, a systematic uncertainty was revealed in the Argonne National Laboratory (ANL) system for dispensing liquid samples. In the original ANL technique, the mass of a liquid sample was determined by weighing a vial with a narrow neck before and after depositing the sample in the vial. The evaporation from the vial occurring during the act of deposition was neglected. Subsequently, the samples were weighed in a pycnometer before and after deposition; this is believed to be the more accurate method. In Ref. [11] (Metrologia), an uncertainty of 1% is implied[1], but in an earlier report (ANL-7642) a figure of 2% is quoted. Further, it was found that this discrepancy reduced to 0.2% if the vial contained some alcohol, but no alcohol was used in the vials for the measurements under discussion. Using similar vials, Goodier at NPL has found a discrepancy of about 0.2% between vial measurements and pycnometer measurements for samples of about 100 mg irrespective of whether the vial contained alcohol or not [12]. One is thus faced with possible corrections of 0.2%, 1% or 2%. As indicated above, De Volpi and Porges in Ref. [11] (Metrologia) corrected all weighings by 1% and stated that this caused a lowering of the neutron emission rate of 0.5% and an upward shift in the observed value of σ_H/σ_M of 1%. The originally observed value of σ_H/σ_M was in agreement with the ratio calculated from the individual cross-sections. The suggestion that the corrected, and now discrepant, value of σ_H/σ_M could be due to impurities in the water supply does not appear to the evaluator to be sufficient to explain the difference, particularly as no neutron-absorbing material was found in the water. Moreover, the presence of an impurity in such abundance would invalidate the equations.

Another relevant factor is that it takes some considerable time to do source measurements with a large number of bath concentrations, at each of which the bath efficiency must be measured. If the weighing error varied (for example with temperature and humidity) over this period, the effect would tilt the fitted line, thus changing both the apparent source strength and the apparent cross-section ratio. Nevertheless, it must be assumed that the authors are in the best position to determine the magnitude of the correction and hence the 1% value should be accepted, but it would seem only prudent to include an uncertainty of 0.5% on the neutron emission rate to allow for aliquotting errors.

Two other factors affect the slope of the line obtained with the higher-energy sources (Ra-Be, Am-Be, ^{252}Cf). The first of these is the change in the neutron escape fraction by nearly a factor of two as H/Mn changes from 30 to 300. The effect would be to increase the apparent cross-section ratio. The effect upon the apparent source strength would depend on the concentration at which the (constant) applied leakage correction was valid.

The second effect is the variation in the oxygen and sulphur fast neutron capture of also nearly a factor of two over the same range. This experiment cannot determine the oxygen loss in water which accounts for about 55% of the effect. Thus the experiment seeks to determine the variation of the loss as the concentration varies, a change of the order of 0.25% for ^{252}Cf and 1.4% for Am-Be. The effect of this loss variation is to change the slope of the line in the opposite direction to the change produced by the leakage variation. The estimate of the oxygen and sulphur loss variation obtained

[1] A 1-mg deficiency in a sample of about 0.1 ml ≃ 1%.

by comparing the slopes of the fast neutron source lines with that of the photoneutron source line is therefore the difference between the slope changes produced by the leakage and fast neutron capture effects. This may explain why the derived values for the latter are so small in most cases. The high value for Am-Be remains a mystery.

The authors quote the calculations of Louwrier in support of their derivations. Whilst calculations by Ryves and Harden [9c] are known to be too high because the $O(n,\alpha)$ corrections used are too high, the Louwrier results are too low because their cross-sections are too low (see Ref. [13]). The effects of oxygen scattering have not been neglected by Ryves and Harden as stated by Louwrier. A concentration-dependent correction of 6% - 7.6% was made by them for this effect. The Ryves and Harden calculations have now been repeated using all the latest cross-section data (including Dandy et al.) which mainly affect the oxygen effect above 7 MeV.

Unfortunately, the only published experimental measurement of the fast neutron capture is Ryves' value for Ra-Be sources for which the spectrum is not very well known; especially at low energies. This makes a comparison of calculation and experiment uncertain. Calculations for Ra-Be sources with the Geiger spectrum using the low-energy group estimates of Zill [14] and Kluge [15] give 2.52% compared with the measurement of Ryves and Harden of 3.05%. The measurement is supported by an unpublished result by M. G. Sowerby and E. J. Axton, based on a comparison of the boron pile and the manganese bath. The new calculations give for ^{252}Cf ($N(E) = E^{\frac{1}{2}} e^{-E/1.43}$) 0.80% for H/Mn = 30 and 0.45% for H/Mn = 300. These values are slightly higher than the original Ryves and Harden calculation because the assumed spectrum used is harder. The values would have to be normalized upwards by about 20% to obtain agreement between the calculation and the measurement for Ra-Be. It is therefore felt that this effect has been underestimated by the authors.

In Table II of Ref. [11] (Metrologia), the Am-Be source value and the correction factors are the same for both treatments. This produces a ratio of 2.4 between the losses for Am-Be and Ra-Be sources, whereas the ratio expected from leakage measurements is about 1.3.

The (constant) leakage correction was derived from the BIPM fit. However, the measurements which contributed to this fit were for bare spheres of $MnSO_4$ and for a variety of concentrations. The leakage from a sphere surrounded by a water bath would be lower by about 20% or more.

Because the source strengths are all obtained from least-squares fits to unpublished data, it is very difficult to re-value the result. At NPL, the ^{56}Mn solution which is used to calibrate the manganese bath is also used to calibrate a highly stable sealed high-pressure ionization chamber. In this way, an accurate check on the long-term reproducibility of the $4\pi\beta$-γ coincidence measurements is achieved so that fluctuating weighing errors, if present, would be revealed. There is no such check on the reproducibility of the ANL measurements other than through a study of repeated measurements on a source. There is a spread of 7% in the results shown for the source in Fig. 6 of Ref. [11] (Metrologia).

If the sources involved in the concentration series were all measured using the same efficiency matrix, it would be possible to obtain two independent evaluations of the californium source 0 by using alternative correction factors and evaluating the bath efficiency as a function of concentration by means of the NPL and NBS sources in turn. However, this

would not be relevant to the all-important californium source 3. Under the circumstances it is recommended that the authors' published value be retained for the purposes of the current evaluation with the overall uncertainty increased to 0.80% to cover additional uncertainties in the correction factors and in aliquotting. However, in fairness it must be stated that the authors claim that their original estimate of uncertainty is justified.

4. CONCLUSIONS

A strenuous effort has been made to re-evaluate the important ^{252}Cf measurements, the results of which are given in Table IV. The listing is provisional in that it is subject to further information being forthcoming from some of the authors. Also, the final NPL value is not yet available. To the gated measurements must be added the delayed neutron contribution of 0.0086 ± 0.004 [16]. The uncertainty of 0.001 as published by Cox has been increased because there is some uncertainty in the definition of 'delayed'. There are a number of nanosecond groups listed by Nefedov et al. [17], and it is not clear how these reconcile with the Cox measurement.

To obtain a final value, a weighted mean of the NPL-bath-dependent group is first calculated with the bath uncertainty excluded. The weighted mean so obtained is then averaged with the other five measurements with the bath uncertainty included (see Table IV). The final result is 3.734 ± 0.0082. The previous pattern of two independent groups seems no

TABLE IV. RE-EVALUATION OF MEASUREMENTS OF $\bar{\nu}$ FOR ^{252}Cf

	Prompt neutrons	Total neutrons
NPL-bath-dependent group[a]		
White and Axton [10]		3.797 ± 0.038
Axton et al. [9b]		3.725 ± 0.019
Moat et al. [8a]	3.719	3.727 ± 0.055
Boron pile		3.700 ± 0.028
Other measurements		
Asplund-Nilsson et al. [3]	3.771 ± 0.060	3.778 ± 0.060
Hopkins and Diven [2]	3.761 ± 0.031	3.770 ± 0.031
Boldeman [6]	3.735 ± 0.014	3.744 ± 0.014
Colvin and Sowerby [7a]		3.713 ± 0.015
De Volpi and Porges [11]		3.729 ± 0.030
NPL-bath-dependent average		3.728 ± 0.0186
Overall weighted mean		3.734 ± 0.0082

[a] The weighted mean of the NPL bath group is 3.728 ± 0.0138. To this must be added the common uncertainty of the Mn bath, 0.0123, giving a total uncertainty of 0.0186. The external standard uncertainty for this group is 0.0168. The boron pile value is retained in this group since the correlation with the other boron pile value is small.

longer to exist, since the external standard error for this group is ± 0.0088. Thus, the results are no longer discrepant. However, the indirect value of $\bar{\nu}$ for ^{252}Cf obtained from η and α for ^{235}U combined with the $^{235}U/^{252}Cf$ ratio is still about 3.78. It therefore appears that there is some error in either α, η or the $\bar{\nu}$ ratio. Since $1 + \alpha$ is involved in the calculation, a large error in the cross-sections would be necessary to explain the discrepancy. The Boldeman value and the Colvin and Sowerby value of $\bar{\nu}$ for ^{235}U are unlikely to be in error, and there are no problems of spectrum conversion. It is possible that the defect lies in the η measurements [18, 19]. Both of these experiments involve a correction of > 3% for the complex opposing effects of fast fission in the sample produced by both outgoing and incoming neutrons and absorption of thermalized neutrons in cadmium. It is a matter of opinion as to whether this effect can be calculated to the accuracy claimed.

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SUMMARY OF THE ABSOLUTE MEASUREMENTS OF $\bar{\nu}$ FOR THE SPONTANEOUS FISSION OF ^{252}Cf

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Abstract

SUMMARY OF THE ABSOLUTE MEASUREMENTS OF $\bar{\nu}$ FOR THE SPONTANEOUS FISSION OF ^{252}Cf.

The experimental techniques used in recent absolute measurements of the average number of fission neutrons, $\bar{\nu}$, for spontaneous fission of ^{252}Cf are reviewed. The status of each experiment is discussed in relation to recent corrections and experimental effects currently being investigated. Results are summarized in tables.

1. INTRODUCTION

The absolute $\bar{\nu}$-value for the spontaneous fission of ^{252}Cf has been recognized as a suitable standard for $\bar{\nu}$-measurements. Thus the ^{252}Cf $\bar{\nu}$-value is requested in RENDA 72 with accuracies between 0.1% and 0.5% depending on its use as a standard in energy-dependent $\bar{\nu}$-measurements of the main uranium and plutonium isotopes. The absolute measurements of $\bar{\nu}$ for ^{252}Cf have recently been reviewed by several authors. The most recent reviews are made by Hanna et al. (1969), De Volpi (1971) and by Manero and Konshin (1972).

In principle, two different methods have been used in accurate measurements of the absolute $\bar{\nu}$-values for ^{252}Cf. The coincidence method is characterized by the use of a fission counter with high efficiency. A relatively low fission rate is used and the neutrons are detected in coincidence with each fission event. In the non-coincidence method, two separate measurements are made, one of the spontaneous fission rate and one of the neutron output of a relatively strong ^{252}Cf-source. Furthermore, the $\bar{\nu}$-value for ^{252}Cf can be calculated from measurements of α and η (capture-to-fission ratio and number of neutrons per neutron absorbed) of a fissile isotope and the $\bar{\nu}$-ratio for that isotope and ^{252}Cf ($\bar{\nu} = (1+\alpha)\eta$).

The original results of recent accurate measurements of the $\bar{\nu}$-value for ^{252}Cf are given in Table I, together with characteristic data of the experimental arrangements. The experiments by Hopkins and Diven (1963), Asplund-Nilsson et al. (1963), Colvin and Sowerby (1965), Colvin et al. (1966) and Boldeman and Walsh (1972) (in progress) were all based on the coincidence method, while the experiments by Moat et al. (1961), Fieldhouse et al. (1966), De Volpi and Porges (1969), White and Axton (1968) and Axton (in progress) were based on non-coincidence methods.

The experiments by Hopkins and Diven (1963), Asplund-Nilsson et al. (1963) and Boldeman and Walsh (1972) were made with a large liquid scintillator as the fission neutron detector, while Colvin and Sowerby (1965) used the boron pile, a graphite stack containing 240 BF_3-counters, as the neutron detector. De Volpi and Porges (1970), White and Axton (1968) and Axton

TABLE I. CHARACTERISTIC DATA OF ABSOLUTE $\bar{\nu}_t$ MEASUREMENTS FOR ^{252}Cf

Experiment	Neutron detector	Method of calibration	Approximate detector efficiency for ^{252}Cf neutrons	ν_t (^{252}Cf)
Coincidence measurements				
Hopkins and Diven (1963)	Cd-loaded liquid scint. 1 m long, 1 m dia.	Monte-Carlo calc. checked against (n, p) scattering in plastic scintillator	86%	3.780 ± 0.030
Asplund-Nilsson et al. (1963)	Cd-loaded liquid scint. sphere, 60 cm dia.	(n, p) scattering in anthracene crystal	69%	3.808 ± 0.034
Colvin and Sowerby (1965)	Boron pile: 220 cm^3 graphite containing 240 BF_3 counters	d(γ, n)p reaction in ion chamber	64%	3.713 ± 0.015
Colvin et al. (1966)	Boron pile	NPL $MnSO_4$-bath	64%	3.700 ± 0.031
Boldeman and Walsh (in progress)	Gd-loaded liquid scint. sphere, 76 cm dia.			3.73[a]

[a] Preliminary value.

TABLE I (cont.)

Experiment	Neutron detector	Method of calibration	Fission rate counter	ν_t (^{252}Cf)
Non-coincidence measurements				
Moat et al. (1961) also Fieldhouse et al. (1966)	Waxcastle cylinder of paraffin containing BF_3-counters	^{240}Pu standard source calibrated in boron pile	Fission fragment counting in ionization chamber	3.684 ± 0.040
White and Axton (1968)	NPL $MnSO_4$-bath sphere 98 cm dia.	β-γ coincidence measurements of ^{56}Mn activity	Solid-state detector in low geometry	3.796 ± 0.031
De Volpi and Porges (1969)	ANL $MnSO_4$-bath sphere 96 cm dia.	β-γ coincidence measurements of ^{56}Mn activity	Fission ionization chamber in coincidence with Hornyak button neutron counter	3.729 ± 0.015
Axton (in progress)	NPL $MnSO_4$-bath sphere 98 cm dia.	β-γ coincidence measurements of ^{56}Mn activity	4π-proportional counter to detect fission fragments in coincidence, from thin foils	3.72[a]
$\bar{\nu}$ deduced from α- and η-measurements				
Hanna et al. (1969)				3.784 ± 0.014

[a] *Preliminary value.*

TABLE II. CORRECTED $\bar{\nu}_t$-VALUES OF ^{252}Cf IN VIEW OF RECENT OBSERVED SYSTEMATIC ERRORS

Authors	Laboratory	Year	Corrected value	Remarks
Coincidence measurements				
Hopkins and Diven	LAS	1963	3.772 ± 0.030	(a)
Asplund-Nilsson et al.	FOA	1963	3.778 ± 0.060	(b)
Colvin and Sowerby	HAR	1965	3.713 ± 0.015	d(γ,n)p calibration
Colvin et al.	HAR	1966	3.700 ± 0.031	NPL $MnSO_4$-bath calibration
Boldeman and Walsh (in progress)	LHL		3.73	(c)
Non-coincidence measurements				
Moat et al.	ALD	1961	3.727 ± 0.056	
White and Axton	ALD	1968	3.796 ± 0.031	(d)
De Volpi and Porges	ANL	1969	3.729 ± 0.015	
Axton (in progress)	NPL		3.72	(c)
Weighted mean (excluding the preliminary values by Boldeman and Walsh and by Axton)			3.731 ± 0.009	
De Volpi	ANL	1971	3.735 ± 0.008	(e)

(a) Corrected for delayed gamma rays (- 0.2 ± 0.2)%.
(b) Corrected for delayed gamma rays (- 0.2 ± 0.2)%, the 'French effect' (- 0.6 ± 0.3)% and leakage ± 1.5%.
(c) Preliminary values.
(d) Error in fission counting of 2% discussed.
(e) Deduced from adjustment of 2200-m/s fission constants retaining the IAEA constant for $\bar{\nu}\sigma_f$.

(1972) have all used manganese sulphate baths as neutron detectors but different techniques when measuring the fission rates. Finally, Moat et al. (1961) used two cylindrical neutron detectors consisting of BF_3-counters embedded in paraffin wax.

A large spread in the original results compared to the claimed accuracies was observed (Table I). The liquid scintillator measurements (accuracies about 1%) by Hopkins and Diven (1963) and Asplund-Nilsson et al. (1963) gave about 1.5% - 2% higher $\bar{\nu}$-values than those reported by Colvin and Sowerby (1965) and De Volpi and Porges (1970) (accuracies about 0.5%). This discrepancy has initiated investigations of possible systematic errors. These investigations have resulted in the recognition of some additional corrections and errors to be applied to the originally reported $\bar{\nu}$-values. The current status of the different experiments is shortly discussed in section 2, and the corrected $\bar{\nu}$-values are given in Table II. The recommended ^{252}Cf $\bar{\nu}$-values of the different experiments in the review by Hanna et al. (1969) are given in Table III.

TABLE III. NEUTRON YIELD PER FISSION ($\bar{\nu}_t$) FOR ^{252}Cf FROM THE IAEA REVIEW BY HANNA et al. (1969)

Authors	Laboratory	Year	Reassessed value	Adopted mean
Liquid scintillator				
Asplund-Nilsson et al.	FOA	1963	3.830 ± 0.037	3.807 ± 0.024
Hopkins and Diven	LAS	1963	3.793 ± 0.031	
Boron pile calibrated with d(γ, n)p reactions				
Colvin and Sowerby	HAR	1965	3.713 ± 0.015	3.713 ± 0.024
Dependent on NPL manganese bath				
Moat et al.	ALD	1961	3.727 ± 0.056	
Colvin et al.	HAR	1966	3.700 ± 0.031	
White and Axton	ALD	1968	3.796 ± 0.031	3.713 ± 0.024
Axton et al.	NPL	1969	3.700 ± 0.020	
ANL manganese bath				
De Volpi and Porges	ANL	1969	3.725 ± 0.017	3.725 ± 0.024
			Weighted mean:	3.740 ± 0.016
			Fitted value:	3.765 ± 0.010

The $\bar{\nu}$-value for ^{252}Cf deduced from 2200 m/s data adjustments and least-squares procedures is discussed in section 3.

2. CURRENT STATUS OF THE ABSOLUTE ^{252}Cf $\bar{\nu}$-EXPERIMENTS

Recent measurements of the fission neutron spectrum for ^{252}Cf yield an average value of the Maxwellian temperature of about 1.4 in accordance with the spectra used in the efficiency calculations by Hopkins and Diven (1963) and Asplund-Nilsson et al. (1963). Hanna et al. (1969) used the fission spectrum obtained by Meadows (1967) with T = 1.59 MeV, which resulted in a + 0.4% and 0.6% correction for the Hopkins and Diven and Asplund-Nilsson results, respectively.

The delayed fission gamma rays from ^{252}Cf have been studied by Boldeman and Walsh (1972) who report a correction to their $\bar{\nu}$-values of (-0.2 ± 0.05)%. The experimental arrangements used by Boldeman and Walsh (1972), Hopkins and Diven (1963) and Asplund-Nilsson et al. (1963) are expected to have the same dependence on delayed gamma rays. Thus it is reasonable to apply the same correction but with a larger error, (-0.2 ± 0.2)%, also to the Hopkins and Diven and Asplund-Nilsson results (Table II).

The so-called 'French effect' suggested by Soleilhac et al. (see Colvin, 1970) or the dependence of the prompt pulse detection efficiency on the number of neutrons detected per fission has been studied by several groups using different liquid scintillators. The effect has been observed by all groups but the magnitude is reported to be small in most cases. It is very likely that the results depend on the light collection efficiency of the liquid scintillator tanks. Measurements by Signarbieux et al. (1971), Mather (see Colvin, 1970), Boldeman and Walsh (1972) and Diven (1970) yield a correction of the order of - 0.2% or less for the French effect, while measurements by Soleilhac et al. (see Colvin, 1970) and Condé et al. (1971) gave larger corrections. The measurement by Condé et al. (1971) was made on the same scintillator as used by Asplund-Nilsson et al. (1963) but with a gadolinium-loaded scintillator instead of a cadmium-loaded one. The correction to the absolute $\bar{\nu}$-measurement by Asplund-Nilsson et al. was calculated to be - 0.6%. Because of the uncertainty introduced by the use of different liquid scintillators, an error of ± 0.3% has been attributed to this correction when applied to the Asplund-Nilsson $\bar{\nu}$-value (Table II).

The leakage correction, i.e. the difference in leakage between a spherical symmetric scintillator tank and a tank with central channel, was relatively large in the experiment by Asplund-Nilsson et al. (1963). It was estimated by a Monte-Carlo calculation. Neutrons of energies of 2 MeV and 3 MeV were started from the centre of the spherical detector at small angles to the axis of the central channel. If the neutrons were slowed down to 1 keV by successive collisions within the scintillator they were registered as capture. The capture probability of these neutrons was compared with that of neutrons started in a direction corresponding to neutrons scattered from an anthracene crystal. The difference in escape of neutrons emitted isotropically from the fission of ^{252}Cf and neutrons scattered from the anthracene crystal was calculated and resulted in a correction in $\bar{\nu}$ of (1.3 ± 0.3)%.

Axton (1972) has recalculated the leakage for a scintillator similar to the one used by Asplund-Nilsson et al. and got a different result. The possibility to do a new careful Monte-Carlo calculation of the Asplund-Nilsson experiment will be looked into. As recommended by Axton, the error of the Asplund-Nilsson ^{252}Cf $\bar{\nu}$-value in Table II was meanwhile increased to 1.5% because of the uncertainty in the leakage correction.

The boron pile experiment was examined in detail for systematic errors by Colvin et al. (1966). No errors could be found in the corrections which changed the $\bar{\nu}$-value or the errors originally stated. The efficiency of the boron pile was measured with the d(γ, n)p reaction and independently with a standard neutron source calibrated with the NPL manganese bath. This resulted in two independent boron-pile $\bar{\nu}$-values (Tables II and III).

Fieldhouse et al. (1966) made a recalibration of the Harwell ^{240}Pu-source used by Moat et al. (1961) for the calibration of their neutron detectors. The recalibration, based on the NPL manganese bath, resulted in a revised $\bar{\nu}$-value of 3.675 ± 0.040. Furthermore, recent experiments of the fission neutron energy difference between ^{252}Cf and ^{240}Pu give a value of 0.40 MeV (Hanna et al., 1969) instead of 0.27 MeV as used by Moat et al. (1961). Using this higher value of the energy difference, the $\bar{\nu}$-value by Moat et al. increases by 1.1% and, assuming an uncertainty in this correction of ± 1%, the error increases from ± 0.040 to 0.056 (Tables II and III).

In the experiment by White and Axton (1968) the neutron counting was made with the NPL manganese bath facility, and the fission fragment detection was made at AWRE, Aldermaston, in a low-geometry counting set-up. The overall systematic error was estimated to be about 0.8%. Recently, White (1970) reported that there is a 2% discrepancy between the low-geometry counting at AWRE and the fission-rate evaluations done at the National Physical Laboratory (NPL) by Axton. It is also stated in the same report that "it is not known whether this discrepancy is significant, and no reason has been found to account for the difference".

De Volpi and Porges (1969, 1970) have very carefully looked into several systematic errors which apply to their manganese bath and fission counting measurements. Three different fission counters were used having efficiencies from 60% to 99%. The absolute fission-rate determination was based on a coincidence measurement between pulses from a fission ionization chamber and a Hornyak-button neutron counter. The fission counter with 99% efficiency was the primary basis for the results of the ^{252}Cf $\bar{\nu}$-value. The estimated systematic error of the fission-rate determination with this counter was 0.13%.

De Volpi and Porges have made several independent measurements to verify the accuracy of their neutron detection manganese bath system. Furthermore, three different neutron sources, i.e. Ra-Be(γ,n), Ra-Be(α,n) and Am-Be(α,n), having a wide range of emission spectra, were studied both at the National Physical Laboratory, the National Bureau of Standards (NBS) and Argonne National Laboratory (ANL). A discrepancy of the order of 1% or more between the NPL and ANL results was observed and the discrepancy increases as the neutron spectrum hardens; therefore, it was suggested that there is some connection with neutron escape or high-energy neutron capture. De Volpi (1971b and 1972) reported correction factors for neutron escape and source and cavity absorption. When these are applied, the results of NPL and ANL differ by no more than 0.5% (for the soft-spectrum source). The error claimed by De Volpi and Porges for the neutron rate measurement was 0.4%. The final value of the ANL ^{252}Cf $\bar{\nu}$-measurement is given in Table II.

The manganese bath measurement at NPL by Axton is in progress. The value given in the review by Hanna et al. (1969) (Table III) was made with a sample containing only about 30% of ^{252}Cf and should be considered as provisional. Measurements with a new sample are in progress and also a comparison of the fission rate per milligram of aliquots of the californium sample between the Central Bureau for Nuclear Measurements at Geel and NPL. The technique for the absolute counting of the fission rate used by Axton (Axton et al., 1969) differs from that used by De Volpi. The fission sources were evaporated on thin plastic films and the two fission fragments were detected separately and in coincidence. The absolute fission rate was determined from the three counting rates with an error of about 0.2%.

A preliminary value of $\bar{\nu}$ for ^{252}Cf from the measurement of Axton is given in Table II.

3. $\bar{\nu}$ (^{252}Cf) DEDUCED FROM 2200-m/s DATA ADJUSTMENTS AND LEAST-SQUARES FIT PROCEDURES

The outcome of ν_t (^{252}Cf) from the revised IAEA least-squares fit of 2200-m/s fission parameters by Hanna et al. (1969) was 3.765 ± 0.010.

This value can be compared with their weighted mean of the absolute $\bar{\nu}$-measurements, i.e. 3.743 ± 0.016 (Table III). The deviation between these two values points to the fact that the least-squares fit $\bar{\nu}$-value was mainly based on α- and η-measurements including $\bar{\nu}$-ratios for ^{233}U, ^{235}U and ^{239}Pu to ^{252}Cf. When abandoning completely all the absolute $\bar{\nu}$-measurements, Hanna et al. obtained a value of $\bar{\nu}$ (^{252}Cf) of 3.784 ± 0.014.

De Volpi also points out that some reduction in the averaged value of η is experimentally justified. The recent measurements with manganese baths at ANL and NPL indicate that the corrections for neutron escape, capture in sulphur and oxygen and resonance absorption in manganese should be revised in the η-experiments by Macklin et al. (1960) and Smith et al. (1966).

Furthermore, if the recent low measurements of the ^{233}U and ^{234}U half-lives are accepted, the 2200-m/s fission cross-section values for ^{233}U and ^{235}U will be about 1% higher.

From the above considerations, De Volpi arrives at a different set of data from which he works out an adjusted set of 2200-m/s data confined by the restraints $\nu\sigma_f = \eta\sigma_a = \eta(1+\alpha)\sigma_f$ = constant. The 'adjusted' value by De Volpi of $\bar{\nu}$ for ^{252}Cf is 3.735 ± 0.008.

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DISCUSSION

(Papers IAEA-PL-246-2/31 and 32)

B.C. DIVEN: I would like to make some comments about the effect which the position in which a neutron is absorbed in a liquid scintillator has on the efficiency for observing that neutron. The pulse-height distribution in a liquid scintillator, which contains a very clear solution so that the mean free path of a gamma ray is much greater than the radius of the scintillator tank, is almost entirely determined by the leakage of gamma rays from the scintillator. If a ^{60}Co source is placed at the centre of a large scintillator, two scintillation peaks which are not completely, but almost, resolved are observed. The higher-energy peak at 2.5 MeV corresponds to absorption within the scintillator of both gamma rays resulting from decay of ^{60}Co. The other peak at about 1.25 MeV corresponds to escape of one gamma ray and absorption of the other.

It is true that if a gamma ray undergoes one Compton scattering, there is a very high probability that the remaining gamma-ray energy will be absorbed in the scintillator. In a neutron capture cascade, some of the gamma rays almost certainly escape, whereas if a gamma ray undergoes one interaction, its energy is almost entirely absorbed in the scintillator.

If a gamma-ray source such as ^{60}Co is moved along the axis of a scintillator, the efficiency for absorbing the gamma ray decreases as the edge of the scintillator is approached. In the double-peaked pulse-height distribution the higher-energy peak decreases and the lower-energy peak increases as the probability of losing one of the gamma rays increases. The efficiency for detecting a neutron by observing scintillation light produced by capture gamma rays is therefore lower when the neutron is captured farther from the centre of the scintillator.

As a result of a letter which I received from Mr. Axton, we made Monte-Carlo calculations to find the mean position in the scintillator at which fission neutrons from ^{252}Cf were absorbed and the mean position at which calibration neutrons were absorbed. The mean radii were fairly similar, as I recall, 18 cm and 14 cm, with the calibration neutrons being captured a little closer to the centre than the ^{252}Cf neutrons. As Mr. Axton has pointed out, this effect should be in the direction to increase $\bar{\nu}$, but I think it is a very small effect.

We repeated some of Mr. Axton's calculations using the same geometry and atomic densities that he used. Our results confirmed his very well so that we have confidence in his calculations. The actual atomic densities occurring in our experiment were somewhat different, so we repeated the calculations using the correct values.

Our final conclusion, which confirmed Axton's, was that no change in our old value of $\bar{\nu}$ was required. This result may be purely academic because we assigned a rather large error to our value of $\bar{\nu}$. Boldeman, for instance, has new measurements with a much smaller error so that our old measurements would probably receive rather low weight in an evaluation.

I have an observation based on the data reported in Mr. Condé's review. I notice that in 1969, Hanna et al. reported a value of $\bar{\nu}$ for

^{252}Cf of 3.784 ± 0.014 based on the thermal constants of the fissile nuclei and excluding direct measurements of $\bar{\nu}$. De Volpi in 1971 derived a value of 3.735 ± 0.008 from essentially the same data, at least the same values of η. The change of 0.049 is six times the quoted error of 0.008. I would estimate the uncertainty, or lack of knowledge, of $\bar{\nu}$ as derived from measurements of η and other thermal constants more by the difference between the analyses of, for example, Hanna and De Volpi. I do not understand how an error as small as 0.008 can be assigned when the change in the reported value is six times greater from one year to the next.

W.P. POENITZ: From the tables presented by both Mr. Axton and Mr. Condé, I notice that the spread among the direct measurements of $\bar{\nu}$ used in various evaluations is of the order of about 2%. The final error quoted for several evaluations is of the order of 1/3 of 1%. Also in the case of the direct measurements I would have expected the final uncertainty to reflect the spread of the experimental data more closely.

H. LISKIEN: I have a question concerning Mr. Axton's own work described in section 3.2 of his review. In the determination of the true fission rate by measuring the fission count rate for ^{252}Cf sources of various thicknesses, should not the factor K in the equation in section 3.2 be a function of the specific activity, i.e. of the thickness of the fissile deposit (californium chloride)? If K is not constant, then the plot would not be expected to be linear.

E.J. AXTON: There are two ways to determine the counting efficiency. One way is to use a single fissile source and to vary the counter voltage, which changes the collection efficiency. The second way is to count a series of sources of different strengths using the same counter voltage. When the count rates are plotted as described in the paper, both methods give the same intercept, i.e. the true fission rate, within experimental error. I am prepared to admit that there is no theoretical proof that the line should be straight. We have measured something like 200 sources, and it always seemed to be straight.

In order for a fission fragment to reach a counter it must be emitted at an angle such that the distance through which it must travel in the fissile deposit and backing is not greater than its range in these materials. The count rates of the detectors are functions of the thicknesses of the deposit and the backing.

There are many other effects which can be introduced into the expressions for the count rates. For example, the range of a fission fragment in a material depends on its mass and its energy. Another consideration is that in order to be detected, a fission fragment must enter a counter with sufficient energy to produce a pulse. Various assumptions about scattering of fission fragments can be used to place limits on the count rates obtained by extrapolation to zero thickness. For example, it can be assumed either that there is no scattering or that there is so much scattering that at least one fragment from each fission is scattered into one of the detectors. If it is assumed that the absorption of fission products in the source is exponential, then the count-rate expressions contain exponential integrals of the second and third orders. We have tried to study many such effects.

It is necessary to make an extrapolation to zero thickness in order to obtain the true fission rate. A mathematician advised us to plot parameters which were uncorrelated, and so we finally decided to plot N_2 versus $(N_1 - N_c)$ as described in my paper.

The final error of the extrapolation will be based on the deviation of the intercepts obtained from counting series of samples prepared from a large number of californium sources.

B.D. KUZMINOV: What is the size of the correction for neutrons absorbed in the source, and does this effect limit the accuracy with which you can measure $\bar{\nu}$?

E.J. AXTON: The magnitude of the correction is about 0.3% and we think the uncertainty of the correction is about 33%. The correction is based on a measurement of the thermal neutron flux at the cavity boundary using manganese foils and on a calculation by Ryves which agrees with the foil measurements within about 8%. A knowledge of the capture cross-sections of the materials comprising the source is also required.

A.T.G. FERGUSON: In Table III of Mr. Axton's paper, which refers to the relatively small bath used in Mr. Condé's work, the neutron leakage from the bath shows significant sensitivity to the assumed spectrum of neutrons. Have similar calculations been made for the large tanks and baths of, say, 1 m diameter?

E.J. AXTON: I think the various authors have made such estimates. From time to time, values of $\bar{\nu}$ are recalculated because of changes in the assumed shape of the ^{252}Cf fission neutron spectrum. In the 1969 IAEA evaluation[1], all the liquid scintillator values were raised, and now we have lowered them again. The change assigned to Diven's[2] measurement was 0.38% and to Condé's[3] 0.6%, which may indicate the relative sizes of the correction for larger and smaller baths, respectively.

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PROMPT NEUTRON YIELD FROM THE SPONTANEOUS FISSION OF ^{252}Cf

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Abstract

PROMPT NEUTRON YIELD FROM THE SPONTANEOUS FISSION OF ^{252}Cf.

The average number of prompt neutrons emitted per fission in the spontaneous fission of ^{252}Cf has been measured using the liquid scintillator method. A value of 3.735 ± 0.014 has been obtained.

1. INTRODUCTION

A serious discrepancy exists between different determination methods of the average number of prompt neutrons ($\bar{\nu}_p$) emitted in the spontaneous fission of ^{252}Cf. The discrepancy is important because ^{252}Cf is used as the standard in $\bar{\nu}_p$ measurements, especially in those for the thermal neutron fission of ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu. Any error in the standard appears directly in relative measurements. The discrepancy first became apparent with the publication of the data of Colvin and Sowerby [1]. Using the boron pile, they obtained a value of $\bar{\nu}_p$ for ^{252}Cf which was approximately 2% less than the two accurate values existing at that time, both of which were obtained using the large liquid scintillator method (Asplund-Nilsson et al. [2] and Hopkins and Diven [3]). Subsequent discussion and an exhaustive search for systematic errors failed to resolve the disagreement.

Recent measurements [4-7] using $MnSO_4$ baths for neutron counting are in good agreement with the boron pile. Previous experimental data from the recent review by Manero and Konshin [8] are summarized in Table I. The discrepancy between values based on the liquid scintillator method and other methods is obvious from the table. Reference to Hanna et al. [9] in Table I is to a survey of the 2200 m/s fission parameters for ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu (σ_f, σ_γ, σ_a, α and η). They made a multi-parameter least-squares fit to reassessed experimental values and included $\bar{\nu}_p$ for the spontaneous fission of ^{252}Cf in the fitted data.

Careful analysis of the $MnSO_4$ bath technique has not resolved the discrepancy. In the liquid scintillator method, several sources of possible systematic error (delayed gamma rays, French effect, multiple photo-multiplier tube pulsing) have been investigated and eliminated as the source of the discrepancy. However, the last liquid scintillation measurements were made in 1963 and it was felt that a new measurement by this method would be valuable. Such a measurement is reported.

TABLE I. PROMPT NEUTRON YIELDS PER FISSION FOR ^{252}Cf SPONTANEOUS FISSION

Experiment	Year	Value	Adopted Mean[a]
Liquid scintillator			
Asplund-Nilsson et al.[2]	1963	3.821 ± 0.037	3.798 ± 0.024
Hopkins and Diven [3]	1963	3.784 ± 0.031	
Boron Pile calibrated with d (γ,n)p reactions			
Colvin and Sowerby [1]	1965	3.704 ± 0.015	3.704 ± 0.024
Dependent on NPL manganese bath			
Moat et al. [10]	1961	3.718 ± 0.056	
Colvin et al. [4]	1966	3.691 ± 0.031	3.704 ± 0.024
White and Axton [5]	1967	3.787 ± 0.031	
Axton et al. [6]	1969	3.691 ± 0.020	
ANL manganese bath			
De Volpi and Porges [7]	1969	3.716 ± 0.017	3.716 ± 0.024
Weighted mean Manero and Konshin [8]			3.731 ± 0.016
" " Hanna et al. [9]			3.734 ± 0.016
Hanna's fitted value			3.756 ± 0.010

[a] Input data from Hanna et al. [9].

2. DETERMINATION OF $\bar{\nu}_p$ FOR ^{252}Cf

The experimental system was basically similar to that used by Hopkins and Diven and Asplund-Nilsson et al. Collimated monoenergetic neutrons were scattered from a hydrogen target located at the centre of the scintillator, and the probability of detecting the scattered neutron was determined. By measuring the recoil proton energy, the energy of the scattered neutron and its entry angle into the scintillator can be determined.

In principle, the experiment requires the neutron detection efficiency of the scintillator to be known for all neutron energies at all angles with respect to the scintillator axis. The detection efficiency for ^{252}Cf fission neutrons can then be obtained by integrating over 4π and the ^{252}Cf fission neutron spectrum. For the required experimental accuracy, time would not allow the exhaustive set of measurements needed to provide the complete basic data set. It was necessary to compromise as follows:

(a) The relative efficiency of the scintillator as a function of neutron energy and angle of emission from the scintillator centre with respect to the scintillation axis was calculated by a Monte-Carlo method.
(b) The calculated efficiency values were normalized at the low-energy end of the neutron spectrum in a measurement in which low-energy neutrons were scattered.
(c) The shape and absolute calibration of the energy dependence of the efficiency curve were checked in a second independent experiment.

Details of the experimental system are given below.

2.1. The liquid scintillator and electronics

The use of liquid scintillators for fission neutron counting was originally developed by Reines et al. [11] and Diven et al. [12]. Subsequent developments were made by Mather et al. [13]. The present experimental system has been described in detail in a number of previous publications [14-16].

The liquid scintillator tank was 76 cm in diameter and held approximately 240 litres of NE 323, a trimethyl-benzene-based scintillator containing approximately 0.5 wt% of gadolinium. A 7.62-cm-diameter tube ran through the centre of the tank and allowed entry and exit of a neutron beam. Neutrons entering the liquid are moderated principally by the hydrogen content of the scintillator and detected by neutron capture in the gadolinium loading. Scintillations produced by the capture gamma rays were viewed by twelve photomultiplier tubes arranged in three coincident banks, each containing four tubes. The average neutron lifetime in the scintillator system before capture is 11 μs.

The system had been in operation for a number of years, and prior to the present measurement the detector was replenished with fresh scintillator liquid and all photomultiplier tubes were replaced.

The ^{252}Cf spontaneous fission counters were identical with that described in Ref. [14]. Two were used during the measurements and had count rates of approximately 8 counts/s and 55 counts/s. A coincidence between pulses from the ^{252}Cf ion chamber and the liquid scintillator (from prompt fission gamma rays and neutron-proton recoil events) was used to initiate the neutron counting cycle. This consisted of a 40-μs gate, initiated 585 ns after fission, to count the associated fission neutrons. A second gate, opened 100 μs after the completion of the first gate, counted the background. The electronics are shown in Fig. 1(a). The system was identical with that previously used, except that data were now recorded event by event on nine-track magnetic tape and the new system allowed up to 15 events to be counted during each gate. The neutron gate and background gate counts for each cycle were written on the magnetic tape in a single eight-bit word. During the course of the measurements, the scintillator was operated at approximately 84.5% efficiency for ^{252}Cf fission neutrons.

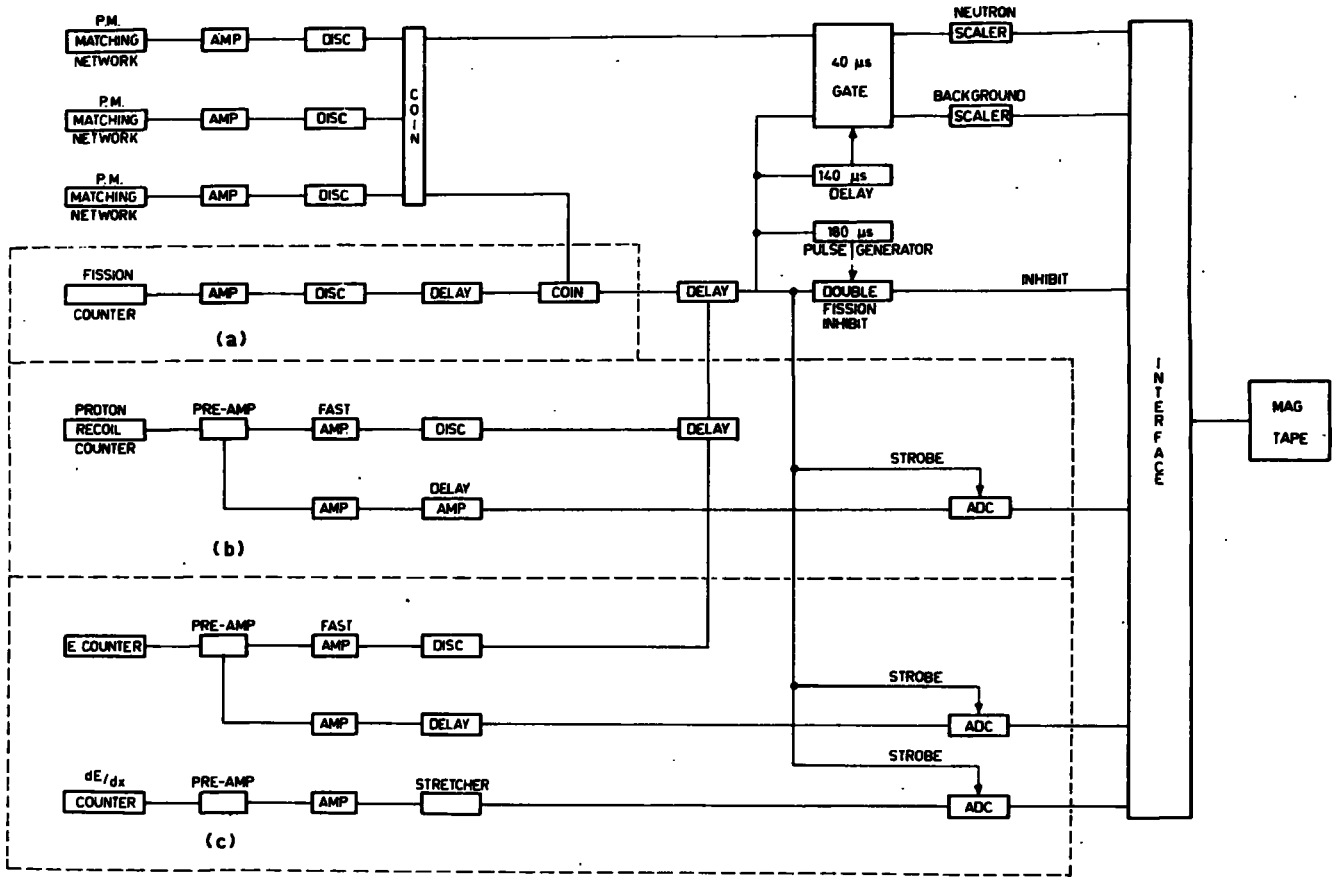

FIG. 1. Block diagram of electronics.

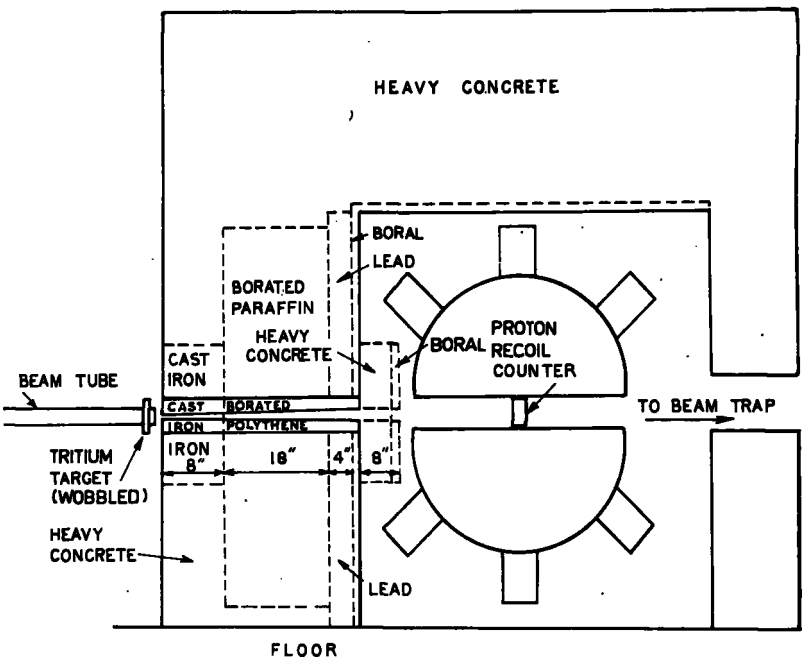

FIG. 2. Neutron collimator and shield for scintillator tank.

For the calibration runs the system was used in conjunction with a Van de Graaff accelerator. Details of the neutron collimator and shielding arrangement are given in Ref. [16] and shown in Fig. 2.

2.2. Monte-Carlo calculation

A special Monte-Carlo code, in which the exact geometry of the scintillator could be specified, was written to calculate the relative efficiency of the scintillator.

In the code, source neutrons were started from the centre point of the system from a variety of distributions in initial energy and starting direction and were tracked through subsequent collisions until they escaped from the system. Standard weight reduction techniques were used, but no other variance reduction techniques were implemented so that code debugging would not be a major problem.

Cross-section data were taken from version III of the ENDF/B data file, and the energy variations of the cross-sections were retained explicitly down to 0.05 eV. Below this energy, a single thermal group was introduced in which cross-sections were averaged over a Maxwellian energy spectrum. Inelastic scattering from carbon was treated explicitly as being from a single level of energy 4.81 MeV, with the angular distribution of the scattered neutron being isotropic in the centre-of-mass frame. Elastic scattering from carbon was treated as isotropic in the centre-of-mass frame for

neutron energies below 0.15 MeV, but above that energy the angular distributions compiled in the UKAEA data file were used. For both gadolinium and hydrogen the scattering was always treated as isotropic. The thin walls of the scintillator were ignored in the calculation.

The code allowed the starting conditions to be specified, and among the options available were the following:

(a) Isotropic Maxwellian neutron energy distribution with specific temperature.
(b) Monoenergetic neutron source within limits (ΔE) at a specific angle to the scintillator axis.

The output from the program included the absorption probability in gadolinium and hydrogen. The neutron detection efficiency of the scintillator for neutrons with the specified starting conditions is then this absorption probability multiplied by the probability of detection of the capture gamma rays so produced. Hydrogen capture is included as the capture gamma-ray energy of 2.2 MeV is well above the scintillator threshold and the relative probability of hydrogen and gadolinium capture is effectively constant for starting neutron energies in the significant energy range. It is assumed that the probability of detecting the capture event is independent of the neutron starting conditions.

2.3. Normalization of the relative efficiency scale

The normalization was made to a measurement in which 2-MeV neutrons (±50 keV) from the T(p, n) reaction were scattered from a hydrogen gas target located at the centre of the scintillator. Scattered neutrons considered for the calibration were restricted to the energy range 0 to 1 MeV for reasons that are given later. The neutron absorption probability of the scintillator system was calculated for a number of scattered neutron energies in the range considered, i.e. 1-MeV neutrons at 45° to the scintillator axis, 0.01-MeV neutrons at essentially 90° and a number of other cases between these limits. The calculated difference between the two extreme cases was 0.3%.

The proton recoil counter is shown in Fig. 3. To minimize neutron reactions of all kinds at the centre of the scintillator, the aluminium body of the detector was made as thin as could be tolerated and the system included the use of aluminium entry and exit windows for the neutron beam.

The proton counter was a standard surface-barrier detector collimated to an active area of 1 cm diameter. The intention was to ensure that all recoil protons considered in the experiment (2 MeV to 1 MeV) originated from a uniformly irradiated target volume (i.e. $2\sqrt{2}$ target thickness + diameter of collimator < diameter of beam). The neutron beam for the experiment had a diameter of 2.5 cm.

The entire chamber was filled with hydrogen gas at 1 atm. The target thickness (±20 keV) was defined by an aluminium foil placed in front of the surface barrier detector. The aluminium was 0.014 g/cm^2 thick and prevented recoil protons from reactions outside the target volume entering the detector. The hydrogen target thickness was determined by the average proton energy loss in the target volume. There are no problems with multiple scattering in this experiment. The multiple scattering probability is minute (5×10^{-5}) and of no significance since the variation in efficiency over the entire energy range is only 0.3%.

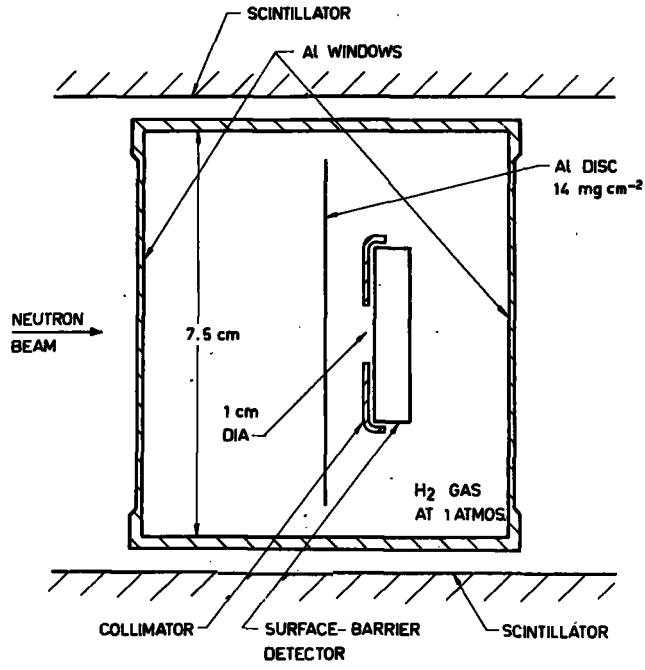

FIG. 3. Proton recoil counter for low-energy run.

Typical recoil proton spectra for incident neutron energies of 2 MeV have the theoretical shape down to at least 700 keV when the gamma-ray background starts to become significant. The neutron energy resolution (i.e. proton energy resolution plus spread in incident neutron energies) was of the order of ±70 keV. To determine the background contribution from gamma-ray detection in the surface barrier detector, measurements were made with the chamber evacuated. The principal background in the experiment arose from recoil proton events in the edge protection of the surface barrier detector. This was apparent as the majority of background events had an associated neutron. With the range of proton energies restricted to the region 1 to 2 MeV, the background correction to the neutron count rates was 0.33%. We have pessimistically assigned an error of ±30% to this correction.

The electronics for the low-energy neutron runs are shown in Fig. 1(b). For each recoil proton detected in the surface barrier detector, the digitized proton recoil energy was recorded on magnetic tape together with the neutron and background counting data from the scintillation tank. Considerable care was taken to ensure that the neutron counting gate opened at the same delay after the origin of the neutrons for both ^{252}Cf fission neutron counting and proton recoil counting. It is considered that the maximum systematic error was ±5 ns. The proton recoil measurements were made with average backgrounds of 0.7 counts per gate.

Seventeen low-energy measurements were made. Each consisted of a series of proton recoil energy measurements interspersed with a number of ^{252}Cf measurements. High statistical accuracy for each ^{252}Cf measurement allowed drifts in the system to be detected, but none were experienced. For each proton recoil measurement, a background measurement was also made with the chamber evacuated. Each proton recoil run was analysed separately. Analysis included dead-time correction (although an accurate average correction was finally used – see later), unfolding of the neutron and background distribution and an assessment of the statistical accuracy. Two tests were made to assess the reliability of the data.

(a) Since the neutron emission for each proton recoil must be one neutron, the unfolded neutron distribution should have a significant probability only for zero or one count. Within the statistical accuracy of each measurement, this was always the case.
(b) The assessed statistical accuracy can be compared with the distribution of data of the 17 different results. A chi-squared test indicated that the comparison was acceptable.

As mentioned previously, the maximum variation in calculated efficiency was 0.3% from effectively zero-energy neutrons at 90° to 1-MeV neutrons at 45°. This was less than could reasonably be determined experimentally (although it was noted that there was no statistically significant variation), and there is no error in treating all data for E_n between 0 to 1 MeV together and relating the value obtained to an average calculated value. Thus, if it can be assumed that the calculation of the relative efficiency of neutron detection is an accurate description of the energy and angular variation, and if it can further be assumed that, once a neutron is absorbed within the scintillator, the efficiency of detection is independent of the original energy, then the neutron detection efficiency for a ^{252}Cf fission neutron spectrum can be determined and a value of $\bar{\nu}_p$ for ^{252}Cf obtained from the ^{252}Cf counting data.

2.4. Experimental verification of efficiency curve

It was necessary to demonstrate experimentally the reliability of the two above assumptions. This was done in a series of high-energy runs in which 16-MeV neutrons from the T(D, n) reaction were scattered by a 0.25-mm-thick hydrogen target (polythene) located at the centre of the scintillator. The background from competing reactions, e.g. (n, p) and (n, α), in the silicon detector was so high that a single surface barrier detector could not be used. To overcome this problem a double detector system was used (Fig. 4). The first detector after the polythene target was a 100-μm-thick, fully depleted wafer, which acted effectively as a dE/dx detector. The second was a 2-mm-thick, fully depleted surface barrier detector [E]. An 800 mg/cm^2 carbon disc was placed in front of the hydrogen target to shield the surface barrier detector from (n, p) and (n, α) reaction in the aluminium counter material, and this ensured that all reaction products entering the surface barrier detection system originated in the hydrogen target. Carbon was chosen because of the high Q for (n, p) and (n, α) reactions. Approximately 4% of the incident neutron beam will be scattered in the carbon shield. However, because of the very high forward peaking of the scattered neutrons, the average energy loss is very small and the end result on the experiment minute.

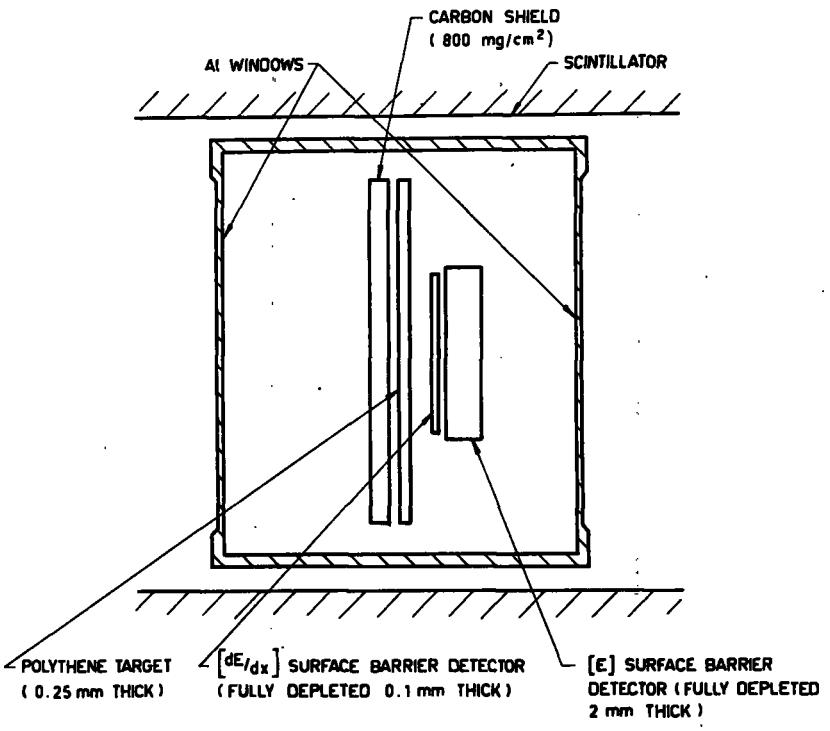

FIG. 4. Proton recoil counter for high-energy run.

The electronics are shown in Fig. 1(c). A fast pulse in the [E] detector gated the analogue-to-digital converters on both the [dE/dx] and [E] lines and also strobed the neutron counting gates on the liquid scintillator. Data were recorded event by event on magnetic tape. The digitized pulse heights from both detectors were stored in eight-bit words, the counting data from the scintillator were stored as before. The resolution of the total energy [E + (dE/dx)] had contributions from the neutron energy spread, the recoil proton energy loss in the hydrogen target, and from the dE/dx and E detectors. The former detector had an energy resolution of approximately 250 keV, which was satisfactory for the experiment, the latter had an energy resolution of approximately 80 keV. For a number of energy ranges spanning the acceptable data (E_p from 16 MeV to 6.67 MeV), the estimated response in both detectors was calculated after allowance for the scattering angle of the recoil proton and the energy loss in the hydrogen target. This allowed restrictions to be set on each detector before considering an event for analysis. Fortunately, the experimental system behaved quite well. Figure 5 shows the dE/dx spectrum for three selected total energy ranges [E + (dE/dx)] for a typical run. All dE/dx curves peak at the appropriate energy, and windows can be set on each side of the peaks.

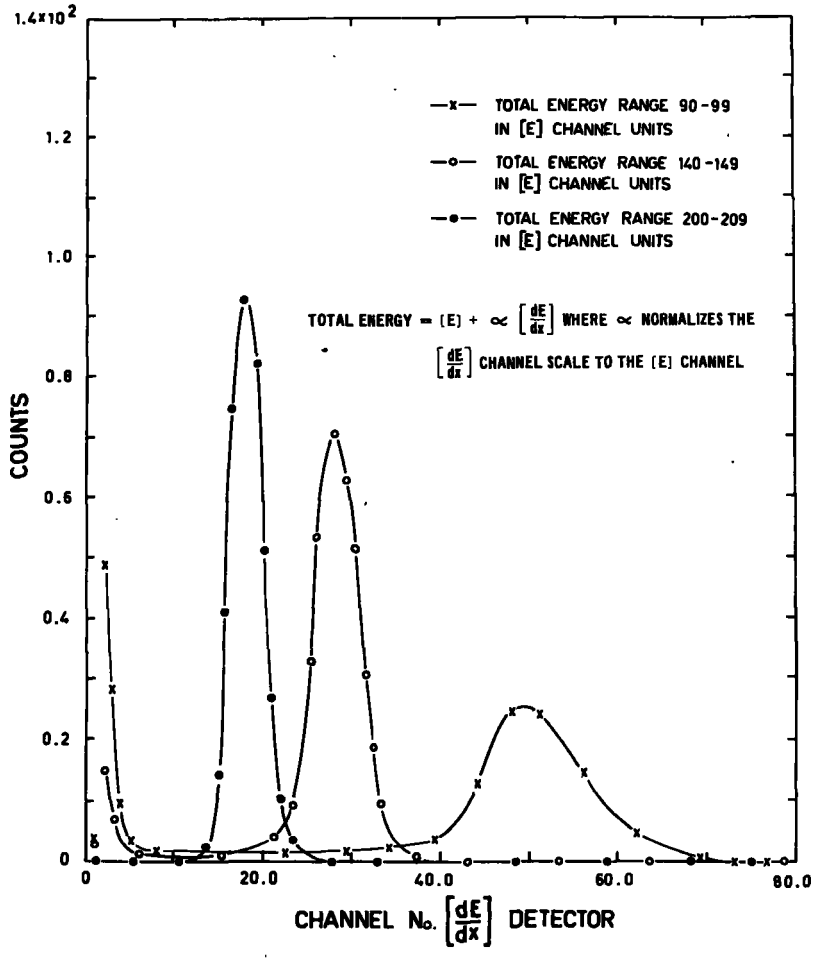

FIG. 5. Typical [dE/dx] detector spectra for three ranges of the total energy.

There was some contribution to the count rate from non-genuine neutron proton scattering events. This contribution was determined experimentally by repeating the measurement with the hydrogen target removed. The correction for background events was 0.1% at high energies, of the order of 2 to 3% for intermediate energies and 15% for the lowest energy. After background events are subtracted, the total energy proton recoil spectra approximate the theoretical shape to at least 6 MeV.

Eight independent measurements were made. During the course of each, frequent ^{252}Cf calibrations were made to detect any drift. A background run was also made for each measurement.

For each energy range shown in Table II the relative efficiency of the liquid scintillator was calculated for neutrons entering the liquid

TABLE II. COMPARISON OF CALCULATED AND EXPERIMENTAL NEUTRON DETECTION EFFICIENCIES

Energy (MeV)	Calculated efficiency (%)[a]	Experimental efficiency (%)
$0.35^{+0.23}_{-0.35}$	89.1	89.3 ± 0.9
$0.99^{+0.35}_{-0.41}$	88.8	89.1 ± 0.8
1.70 ± 0.36	88.4	89.2 ± 0.8
2.42 ± 0.36	87.1	86.7 ± 0.7
3.14 ± 0.36	85.8	84.5 ± 0.7
3.86 ± 0.36	84.4	83.3 ± 0.7
4.59 ± 0.37	82.1	81.5 ± 0.8
5.32 ± 0.37	80.0	79.7 ± 0.7
6.03 ± 0.36	76.8	76.8 ± 0.8
6.73 ± 0.35	73.5	74.2 ± 0.9
7.42 ± 0.35	73.5	73.2 ± 1.0
8.56 ± 0.77	69.4	69.2 ± 0.8

[a] Statistical accuracy ±0.1%.

scintillator at the average angle for that range. The calculated data were then normalized via the ^{252}Cf calibration data using the $\bar{\nu}_p$ value determined in the low-energy measurement. Table II shows an un-normalized comparison of the calculated and experimental values for each energy range. The agreement between the two sets of data is good and confirms the reliability of the calculation.

3. RESULTS AND DISCUSSION OF ACCURACY

The final value obtained for the number of prompt neutrons emitted in the spontaneous fission of ^{252}Cf was 3.735 ± 0.014. Corrections and contributions to the experimental accuracy are considered below and summarized in Table III.

3.1. Statistical accuracy

The statistical accuracy calculated as in Ref. [14] was 0.24%. This was the accuracy from the comparison of the low-energy calibration of

TABLE III. CORRECTION TO EXPERIMENTAL DATA AND SOURCE OF ERROR

Effect	Correction (%)	Contribution to Accuracy (%)
1. Statistical accuracy		0.24
2. Dead-time correction:		
(a) ^{252}Cf	+1.107	
(b) Low-energy proton recoil	+0.279	
(c) Relative	+0.828	0.10
3. Delayed gamma rays	-0.28	0.07
4. Fission neutron spectra		
(a) Accuracy of $\bar{E}$		0.12
(b) Accuracy of energy calibration		0.17
5. French effect	-0.10	0.10
6. Effect of hole through scintillator		0.10
7. Background error in proton recoil counter	0.33	0.10
TOTAL		0.38

the scintillator with the ^{252}Cf count rate. A value for $\bar{\nu}_p$ for ^{252}Cf could be deduced from the high-energy measurements. However, these measurements were regarded purely as establishing the reliability of the Monte-Carlo calculations and have not been included.

3.2. Dead-time correction

The dead-time correction followed the method given in Ref. [14] but was extended to allow for triple pulse overlap and two overlaps per gate. The increased correction was estimated from a test case and amounted to a 2% increase of the normal dead-time correction for a typical ^{252}Cf measurement.

The dead time of the counting system was determined by measuring the average minimum separation of countable pulses [14]. The average minimum separation was found to be 87 ns. However, it was noticed that the time spectrum for the distribution of pulse separations had a small peak slightly above the average separation. This was originally considered to be the result of a small double pulsing probability. Subsequent investigation showed the time spectrum to be independent of the count rate.

TABLE IV. DELAYED GAMMA-RAY DATA

Experiment	Fission process	Half-life (μs)	Cascade energies (keV)	Isotope	Yield per fission (%)
Guy [17]	^{252}Cf	0.162	115.0, 296.9, 1279.8	A=134	1.15±0.10
Ajitanand [18]	^{252}Cf	<0.250	115.2		
Walton & Sund [19]	^{235}U(n,f)				
Walton & Sund [19]	^{239}Pu(n,f)				
Grüter et al. [20]	^{235}U(n,f)	0.20	115,297, 1280	^{134}Te	
Boldeman [21]	^{252}Cf				
Data for correction		0.162	Total energy 1691.7		1.15±0.10
Guy [17]	^{252}Cf	0.62	324.5, 1181.0	A=135	0.31±0.02
Ajitanand [18]	^{252}Cf	0.63	326.7		0.25±0.02
Walton & Sund [19]	^{235}U(n,f)	3.4		^{134,135}Sb ^{134,135}Te	
Walton & Sund [19]	^{239}Pu(n,f)				
Grüter et al. [20]	^{235}U(n,f)	0.57	324.5, 1181.0	^{134,135}Sb ^{134,135}Te	
Boldeman [21]	^{252}Cf				
Data for correction		0.62	Total energy 1505.5		0.28±0.02

TABLE IV. (cont.)

Experiment	Fission process	Half-life (μs)	Cascade energies (keV)	Isotope	Yield per fission (%)
Guy [17]	^{252}Cf	3.1	197.3, 380.7, 1313.3	A=137	0.66±0.05
Ajitanand [18]	^{252}Cf	3.4	383.5		0.39±0.09
Walton & Sund [19]	$^{235}U(n,f)$	3.4	205, 390, 1330		0.63±0.20
Walton & Sund [19]	$^{239}Pu(n,f)$	3.4	205, 390, 1330		1.30±0.30
Grüter et al.[20]	$^{235}U(n,f)$	3.2	197, 381, 1313	^{136}Xe	
Boldeman [21]	^{252}Cf		390, 1310		0.30
Data for correction		3.1	Total energy 1891		0.60±0.05
Guy [17]	^{252}Cf				
Ajitanand [18]	^{252}Cf				
Walton & Sund [19]	$^{235}U(n,f)$	26.7	720, 990		0.45±0.12
Walton & Sund [19]	$^{239}Pu(n,f)$	26.7	720,990		0.73±0.15
Grüter et al. [20]	$^{235}U(n,f)$				
Boldeman [21]	^{252}Cf		990		0.19
Data for correction		26.7	1710		0.38±0.19

TABLE IV. (cont.)

Experiment	Fission process	Half-life (μs)	Cascade energies (keV)	Isotope	Yield per fission (%)
Guy [17]	^{252}Cf				
Ajitanand [18]	^{252}Cf				
Walton & Sund [19]	$^{235}U(n,f)$	54.0	260, 850		0.85±0.07
Walton & Sund [19]	$^{239}Pu(n,f)$	54.0	260, 850		0.72±0.06
Grüter et al. [20]	$^{235}U(n,f)$	57.0	257	^{93}Rb	
Boldeman [21]	^{252}Cf		250, 850		0.25
Data for correction		54.0	1110		0.50±0.25
Guy [17]	^{252}Cf				
Ajitanand [18]	^{252}Cf				
Walton & Sund [19]	$^{235}U(n,f)$	80	460, 1250		0.32±0.05
Walton & Sund [19]	$^{239}Pu(n,f)$	80	460, 1250		0.46±0.03
Grüter et al.[20]	$^{235}U(n,f)$				
Boldeman [21]	^{252}Cf		1250		0.32
Data for correction		80	1710		0.64±0.32

Thus, although the effect is not entirely understood, it is now considered to be a consequence of the coincidence arrangements on the photomultiplier tubes. Then the small peak is produced by pulses occurring earlier than the apparent minimum separation. The value for the dead time has been adjusted accordingly to 78 ns, and an error equal to the correction is assumed.

The accuracy of the calculated dead-time correction is limited by the statistical accuracy of the counting data. Thus, there is a significant distribution in the magnitude of the dead-time correction for the low-energy proton recoil measurements. To bypass any error introduced on this account, a composite dead-time correction was made to the combined data from the 17 different measurements. There is, of course, no problem with the statistically highly accurate ^{252}Cf data. The dead-time correction for the low-energy proton recoil measurements was 0.279% and for the ^{252}Cf data 1.107%.

3.3. Delayed gamma rays

Delayed gamma rays have been observed following fission and have been attributed to the decay of isomeric states. A number of these isomeric states have half-lives in the range 0.15 to 80 μs and emit cascade gamma rays which exceed in total energy the threshold of the liquid scintillator. They will, therefore, make a contribution to the ^{252}Cf neutron count rate. Table IV lists the experimental data for half-lives, cascade gamma-ray energies, an assignment as to their mass and charge and percentage yields per fission of the isomeric states that contribute significantly. Also shown in Table IV are the yield data used for the delayed gamma-ray correction.

For the 0.162-μs isomer, the yield has been taken directly from Guy [17]. From this reference, the yield for the two higher-energy gamma rays has been averaged. Because the difference in yield exceeds the experimental error, a slightly larger error has been assumed.

For the 0.62-μs and the 3.1-μs isomers, weighted averages of Guy [17] and Ajitanand [18] have been used. For the longer half-life isomers there are only some unpublished data from an experiment performed several years ago [21]. The data from this experiment were never satisfactory, and it can be seen in Table IV that they are systematically low by a factor of 2, especially in comparison with the ^{235}U and ^{239}Pu data from Walton and Sund [19]. The data of Ref. [21] have therefore been renormalized to the Guy [17] and Ajitanand [18] average data for the 3.1-μs isomer. Errors of 50% have been given to the yield data.

The efficiency of the scintillator was measured for a number of gamma-ray energies up to 3 MeV using calibrated isotopes. The contribution of the delayed gamma rays to the ^{252}Cf neutron counts was estimated from the yield data in Table IV and the gamma-ray efficiency data. The correction to the ^{252}Cf count rate was -0.28 ± 0.07%. The errors have been added linearly.

3.4. Fission neutron spectra

The efficiency of the liquid scintillator for ^{252}Cf fission neutrons has been calculated assuming that the spectrum is accurately represented by a Maxwellian distribution. The experimental data generally agree with

TABLE V. COMPARISON OF MAXWELLIAN ENERGIES

Experiment	Maxwellian energy
Werle and Bluhm [23], proton recoil	2.155
Werle and Bluhm [23], ^{3}He spectrometer	2.130
Pauw and Aten [24]	Consistent with 2.085
Zamyatnin [25]	2.22
Green [26]	2.085
Meadows [27]	2.348
Condé and During [28]	2.085
Bonner [29]	2.05
Smith et al. [30]	2.35

this view, but there are serious discrepancies between experimental determinations of the temperature describing the Maxwellian shape [22]. The discrepancy between microscopic and macroscopic determinations of the fission neutron spectrum for ^{235}U and ^{239}Pu is to some extent relevant to ^{252}Cf. In addition, there appears to be a discrepancy between microscopic measurements of the temperature for ^{252}Cf. Table V shows a compilation from Werle and Bluhm [23]. Stated simply, one group of measurements cluster around $\bar{E}$ = 2.15 MeV ($\bar{E}$ = 1.5 T) and the other group near 2.35 MeV. The evidence in our judgement is stronger for the lower average. We have accordingly used a value of 2.15 MeV for the average Maxwellian energy and have subjectively given this value an error of ±0.05 MeV. The present value of $\bar{\nu}_p$ for ^{252}Cf can be adjusted in the future when the problem is resolved, using $d\bar{\nu}_p/d\bar{E}$ = 0.091 MeV^{-1}.

A further contribution to the experimental uncertainty arises from the precision with which the neutron energy dependence of the scintillator is known. The value of $\bar{\nu}_p$ obtained in the present experiment is based on the calculated energy dependence of neutron detection. From Table III, the calculated shape has been confirmed experimentally to better than 1.5%. An error (Table IV) has been estimated assuming that the energy dependence is exact between thermal and 1 MeV and is in error from 1 MeV upwards at the rate of 1.5% per 10 MeV.

3.5. French effect

It has been suggested by Soleilhac et al. (quoted in Ref. [31]) that the requirement of a prompt scintillator pulse introduces an error in ^{252}Cf fission neutron counting. The error is considered to occur because the probability of a coincident scintillator pulse is not independent of the number of emitted neutrons. The magnitude of the effect given by Soleilhac et al. was approximately -1.5% for a measurement performed under conditions similar to the present one. A subsequent investigation of the French effect

has shown it to be significantly smaller. Mather et al. (quoted in Ref. [31]) obtained a value of -0.1% for an experimental set-up similar to the present experiment. Signarbieux et al. [32] find a value of -0.1%.

For the present experiment, the neutron detection efficiency of the liquid scintillator with and without a prompt scintillator pulse has been compared at a neutron detection efficiency of 84.5%. The efficiency was apparently 0.10 ± 0.10% higher if a coincident scintillator signal was required. The present experiment could have been performed without a coincident signal being required in the ^{252}Cf neutron counting, but additional care is needed in these circumstances to minimize electrical interference (e.g. from other experiments). We prefer to use the coincidence and accept the accompanying small correction and the slight increase in the experimental error.

3.6. Effect of the hole through the scintillator

The hole through the scintillator has a direct geometrical effect of 0.5% on the neutron detection efficiency of the scintillator. In addition, the liquid scintillator in the vicinity of the hole presents a varying thickness to the neutrons depending on the emission angle with respect to the scintillator axis. The experiments conducted for calibrating and proving the performance of the scintillator have not tested the adequacy of the calculation in assessing this second effect. In these experiments it was not possible to investigate sufficiently low-energy recoil protons. A possible alternative method of testing the calculation is to insert a plug into the hole and measure the efficiency with and without the plug. The effect to be looked for in terms of leakage is of the order of 0.25% (only half of the hole can be filled because of the ^{252}Cf fission counter). Under these conditions, however, there is a further effect that completely dominates any variation in the leakage. Neutron capture in the hydrogen plug is of the order of 1.2%, and since the majority of these hydrogen captures will not be detected by the photomultiplier tubes, the efficiency of the scintillator will fall with the plug inserted rather than rise, by up to 0.9%. An effect of this size has been observed. There does not appear to be any satisfactory method of testing for the influence of the hole.

An error has been assessed as follows. The leakage from the scintillator has been determined with and without the holes filled with scintillator. The total effect is 0.6%. There is a geometrical effect of 0.5% and therefore the calibration estimates an effect of 0.1% because of the influence of the hole on the surrounding volume. We have assumed an error of 100% in this estimate.

3.7. Multiple pulsing

Signarbieux et al. [32] have suggested that multiple pulsing of the photomultiplier tubes on the liquid scintillator may cause an error in $\bar{\nu}_p$ measurement. The multiple pulsing probability could not be measured for the present experiment. The neutron distribution data for the low-energy proton recoil measurements suggest that the probability is less than 0.2%. The multiple pulsing probability would also appear in a measurement of the average pulse separation. The strange effect observed in this measurement (referred to in section 3.2 and, if a multiple pulsing problem,

equal to a probability of less than 0.2% for neutron counting during a ^{252}Cf fission neutron gate) has been attributed to an effect of the triple coincidence.

However, it is not considered that a large multiple pulsing probability would affect the data. The comparison in this experiment is between neutrons emitted in ^{252}Cf fission and neutrons from proton recoil reactions. The multiple pulsing probability should be independent of the neutron source and therefore would cancel. An effect would only be experienced if the probability becomes high enough to influence the dead-time correction.

3.8. Other sources of error

A number of other sources of error were considered:

(a) Physical difference between the ^{252}Cf fission counter and the proton recoil counter. For ^{252}Cf fission neutrons, no variation in efficiency of the scintillator could be measured between the proton recoil counter placed directly beside the fission counter and withdrawn entirely.

(b) Gate starting time. There is a possible difference of ±5 ns in the starting time of the liquid scintillator counting cycle between ^{252}Cf neutron counting and neutron counting from proton recoil reactions. This difference in the efficiency is insignificant.

(c) A number of other sources of error have been considered in Ref. [14]. They have been shown there to be minute.

4. CONCLUSION

The value obtained for $\bar{\nu}_p$ for the spontaneous fission of ^{252}Cf, 3.735 ± 0.014, is in better agreement with results from the boron pile and $MnSO_4$ bath determination than with previous liquid scintillator measurements. Therefore, the present value seems to confirm a lower value of $\bar{\nu}_p$ for ^{252}Cf.

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DISCUSSION

J.W. BOLDEMAN: Another effect I should mention is neutron capture in the steel frame of the scintillator. I do not think this effect is important although it does contribute to the neutron count rate. Neutron capture in iron makes available about 8 MeV of gamma energy so that it does not matter significantly whether the capture takes place in iron or in the gadolinium loading.

B.C. DIVEN: As a result of correspondence with Mr. Axton we included all iron parts of our system in our Monte-Carlo calculations. I also looked up the pulse-height spectrum obtained from capture in an iron sample at the centre of the scintillator and compared this with the pulse-height spectrum obtained from capture in cadmium, with which our scintillator was loaded.

As expected, very little capture occurred in the iron structure, the spectrum for capture in iron was slightly different from capture in cadmium, and the efficiencies were a little different. The resulting correction was negligible. Since the neutron lifetime in Boldeman's scintillator was a little shorter than in ours, the correction would be even smaller for his experiment.

J.W. BOLDEMAN: The thickness of iron might be considered to contribute to the effective volume of the scintillator. I have compared two cases. In the first, it was assumed that scintillator liquid also occupied the space normally occupied by iron. In the second, it was assumed that the scintillator liquid occupied only its own volume and that no iron was present. The difference in $\bar{\nu}$ for the two cases was only something like 0.04%.

H. CONDE: I would like to know more about the normalization of the relative efficiency.

J.W. BOLDEMAN: We calculated the efficiency of the scintillator at a series of neutron energies for a series of angles of entry of neutrons into the scintillator. We thus obtained the calculated efficiency for a matrix of neutron energies and entry angles. From our low-energy proton-recoil

measurements, we determined the observed efficiency for neutron detection corresponding to points along one of the semi-diagonals of the energy-angle matrix. The entire matrix was then renormalized to these data. It was then necessary to demonstrate that the variation of efficiency across the energy-angle matrix had been correctly calculated. Therefore we made more proton-recoil measurements to obtain data corresponding to points on another semi-diagonal which cuts across the entire range of energies and angles in the matrix. If the measurements agreed with our calculations, then we assumed that we have sampled simultaneously, and calculated correctly, the variation in efficiency both with neutron energy and with angle of entry of the neutron into the scintillator. To determine every point in the matrix experimentally would require measurements at all neutron energies up to 16 MeV.

H. CONDE: Is the axial hole through the scintillator included in the Monte-Carlo calculations?

J.W. BOLDEMAN: The exact geometry is specified.

E.J. AXTON: Is it possible for a calibration neutron or a background neutron to find its way into the proton recoil counter and to start an event?

J.W. BOLDEMAN: I have not specifically worked this out, but I think there is sufficient information in the paper. The probability for multiple scattering of each calibration neutron which enters the proton-recoil counter is 5×10^{-5}. Since there is no multiplication of neutrons, I do not see why the probability of producing a proton recoil should be any greater for a neutron scattered back into the detector than for one which originally enters as part of the neutron beam.

A neutron scattered back into the detector would also have very low energy so that it would be even less likely to cause an event. This was one reason for using low-energy neutrons in normalization experiments. Finally, any uncertainties contributing to error in the proton recoil energies are minimized because the variation in efficiency for neutron detection is only 0.3% over the entire recoil-proton energy range.

R.W. PEELLE: I have several interrelated questions.

(1) What does the pulse-height spectrum of the scintillator look like and where is the bias set?

(2) Is it possible to make an estimate of the absolute efficiency without normalizing? This would help in understanding the processes which occur, even though the calculated efficiency might not be so precise as required or as obtainable experimentally.

(3) It appears not to matter significantly that the efficiency for detection of a neutron varies, depending on how close to the shell of the scintillator it is absorbed. Perhaps this is because, even at the highest energies, few neutrons are absorbed near the edge. In this context, what is the effect of the tube which passes through the scintillator?

J.W. BOLDEMAN: We do not claim that the probability for detection of gamma rays from neutron capture is constant over the entire volume or even that the variation is small. Neutrons entering the scintillator undergo a large number of scattering collisions so that it is not long before they have forgotten their original starting energy. It is only the small difference in the average position of neutron capture which affects the relative detection efficiency for gamma rays.

Mr. Diven pointed out earlier that the difference in average position of capture for californium neutrons and for calibration neutrons was small

so that the effect of differences in efficiency for detection of gamma rays as a function of capture position is much reduced.

B.C. DIVEN: As a result of correspondence with Mr. Axton, I have recently reviewed information relevant to Mr. Peelle's questions. The shape of the pulse-height spectrum will, of course, vary from one scintillator to another. Our scintillator was considerably larger than Boldeman's and a greater percentage of its area was covered by a photocathode so our pulse-height spectrum may have had a somewhat more desirable shape than his.

In the pulse-height spectrum (probability versus pulse height) of our scintillator, the probability was near zero at zero pulse height and rose almost linearly to a maximum at the maximum pulse height of about 9 MeV, which is approximately the binding energy for thermal neutrons in cadmium. With a bias of approximately 1 MeV, we could make a small extrapolation to estimate the fraction of pulses missed. We started our gate at about 1 μs and were able to estimate the fraction of events which occurred before and the fraction which occurred afterwards. If these various fractions and the estimated leakage are added up, the loss of efficiency of the scintillator can be accounted for within probably 2%, which is within the accuracy to which the approximations can be made. The estimated efficiency is not nearly so accurate as measuring the overall efficiency by scattering neutrons off protons, but the agreement is very reasonable.

M.S. COATES: Do you get a severe background effect from cosmic rays at low pulse heights corresponding to about 2 MeV?

J.W. BOLDEMAN: Yes. We operate the scintillator at lower efficiency in order to eliminate background which rises very rapidly at low pulse heights. In the californium measurements the background was about 0.26 - 0.28 counts per gate.

If a large amount of data is accumulated, one occasionally finds very high energy peaks which might be attributed to cosmic rays. In some early experiments, scintillators were equipped with a device to ignore all events whenever a cosmic-ray event was detected. We have never used such a device because we think the probability of such an event is the same for fission neutron or background neutron counting.

IV. FISSION AND CAPTURE STANDARDS

D. The 2200-m/s fission and capture cross-sections of the common fissile nuclides

SUMMARY OF THE CONSULTANTS' MEETING ON THE 2200-m/s FISSION AND CAPTURE CROSS-SECTIONS OF THE COMMON FISSILE NUCLIDES

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Abstract

SUMMARY OF THE CONSULTANTS' MEETING ON THE 2200-m/s FISSION AND CAPTURE CROSS-SECTIONS OF THE COMMON FISSILE NUCLIDES.

In 1969, IAEA consultants reviewed the 2200-m/s fission parameters of the common fissile nuclei and recommended values on the basis of least-squares fits of the available data. In November 1972, another group of consultants met at the IAEA to review the input data and the assigned errors which determine the weighting of the various data in least-squares fitting procedure. New recommended values of the 2200-m/s fission parameters of ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu and ^{252}Cf will be published.

In November 1972, a consultants' meeting was held at the IAEA to discuss and reassess input data for the IAEA evaluation of the 2200-m/s fission parameters of ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu and ^{252}Cf. The participants were B.R. Leonard, Jr., J. Story, E. Axton, H. Lemmel and C. Dunford from the IAEA and myself.

It is impossible to go through each single entry to this least-squares fit. This paper refers to a few points where information is still lacking, where serious doubts about some measurement groups exist, or where striking difference in the output is obtained when one measurement or a group of measurements is discarded from the input.

Discrepancies exist among the alpha half-lives used to determine the number of atoms in many fission cross-section measurements at 2200 m/s. In the 1969 IAEA review [1] the value recommended for the half-life of ^{234}U was $(2.488 \pm 0.016) \times 10^5$ a, whereas a recent determination at the Central Bureau for Nuclear Measurements (CBNM) using several counting techniques and several destructive methods (controlled potential coulometry, isotopic dilution, weighing in ultra-high vacuum) produced the value 2.446×10^5 a $\pm$ 0.3% (99.7% confidence limit). This value has been confirmed at Chalk River and at Argonne National Laboratory.

For the ^{233}U alpha half-life the situation is much more uncertain. There is a group of half-life values situated around 1.61×10^5 a, and there are two recent low values, one by Oetting who used calorimetry and one by Keith who used an alpha counting technique, of about 1.553×10^5 a. However, there is a preliminary Chalk River value of 1.583×10^5 a that falls in between the two groups. The value used in the 1969 evaluation was 1.593×10^5 a ($\pm$ 1.5% accuracy). In our last run, we used 1.58×10^5 a $\pm$ 0.03 (average between high and low) with the hope that a more precise and decisive measurement will become available soon from CBNM.

As regards the ^{239}Pu alpha half-life, another problem arose. Until now, all half-life measurements had been very consistent at about 24 395 ± 29 a. The earlier calorimetric measurements of Oetting in 1969 were also in reasonable agreement. However, the recent value of Oetting from his calorimetric experiment is 24 065 ± 50 a, which is discrepant from the value obtained by other experimenters by about 1.3%. This will be directly reflected by the σ_f-values entered in the evaluation. Here also, a precise counting experiment could be decisive. One should not overlook that in a calorimetric experiment the branching ratio as well as the energies of the alpha particles are required to derive the half-life value.

There was an extensive discussion of use of 2200 m/s fission cross-sections of earlier experimenters with full regard to problems such as extrapolation to zero thickness for efficiencies determined in the range 0.1 mg/cm^2 to 0.5 mg/cm^2.

The problem of underestimating fission fragment losses in thin foils was also discussed. Further, the effect of foil absorption in connection with fission neutron detection under 90° and 45° carried out at Aldermaston was brought up. Our work at CBNM concerning foil assay and spectrometer calibration was extensively and critically discussed, and finally a higher fission cross-section for ^{235}U was introduced taking into account the alpha half-life change in ^{234}U and some underestimated corrections in previous measurements. The input value for ^{235}U now is about 587.5 b, whereas in the previous review it was 581.6 b.

Another problem that was discussed at length was the deduction of a 2200-m/s value from an irradiation experiment. It is very hard to estimate the errors on such an experiment because of the assumptions that are made concerning the neutron spectrum in which the irradiation took place. Not only the temperature but also eventual distortions from a Maxwellian distribution have to be accounted for. However, in the 1969 evaluation, some alpha values and especially fission cross-section ratios from the Chalk River (CRC) irradiation experiments were entered with such a high precision that they limit to some extent the variation of other parameters. The value from the 1958 CRC experiment for the σ_f-ratio of $^{239}Pu/^{235}U$ comes down a little when the new alpha half-life of ^{233}U is taken into account, but a recent experiment [2] is half-life independent and gives a value which is only 0.4% lower than the previous one. To illustrate what this means for $^{239}Pu/^{235}U$, some recent values of g-dependent fission cross-section ratios are given as follows:

White et al. [3]:	Direct monokinetic energies	1.253 ± 0.022
	Thermal column irradiation (g-dependent)	1.277 ± 0.025
	Beam extracted from thermal column	1.235 ± 0.022
Keith et al. [4]:	Thermal column irradiation	1.271 ± 0.015
Bigham et al. [5]:	1958 CRC	1.2970 ± 0.0075
Lounsbury et al. [2]:	CRC	1.2926 ± 0.0081 (temperature controlled)
CBNM ratio:		1.263 ± 0.0096

The last two measurements are contradictory and prevent to some extent the ^{235}U fission cross-section from becoming higher as a result of the least-squares fit of all the data. Therefore, trial least-squares fits are planned to evaluate the effect of some highly weighted values, which tend to exclude less highly weighted data with which they disagree.

There were similar problems regarding the weight that the evaluated $\bar{\nu}$-value for ^{252}Cf carries as compared to the η-values entered by Macklin et al. [6] and Smith et al. [7], essentially with an error of 0.3%, based on Monte-Carlo calculations of a rather complex system (3% corrections).

A run made using all data (η, $\bar{\nu}$ ratios, absolute values of $\bar{\nu}_{Cf}$) yielded 3.750 ± 0.007 for $\bar{\nu}_{Cf}$ as output. When the absolute $\bar{\nu}$ (^{252}Cf) values were left out, $\bar{\nu}_{tot}$ became equal to 3.785 ± 0.012. So here there is an inconsistency and probably the η-values should have wider errors assigned. A critical review of the $\bar{\nu}$-ratios has to be carried out.

It was also suggested to allow for differences in the fission neutron spectrum of various isotopes and for the small effects that may result from a poorly defined distinction between $\bar{\nu}_p$ and $\bar{\nu}_{tot}$, but they were considered of minor importance.

Several trial runs have already been performed by Lemmel with the new input data, but some of the problems noted here need further consideration before a final run can be made and before final values for all the parameters can be recommended. Nevertheless, when comparing the very preliminary output from the last trial runs, we see that, compared to the 1969 IAEA evaluation, there will be several changes in the recommended values, i.e. σ_f of ^{235}U will be increased, σ_s^b of ^{235}U will be decreased, $\bar{\nu}_{tot}$ of ^{252}Cf will be decreased, changes will be made in almost all g-factors, and $\alpha(^{235}U)$ will be decreased. There will be relatively little change for ^{241}Pu because the input data are so poor.

It can be concluded that, for the final revision of the 2200-m/s fission parameters, analysis work still remains to be done especially for g-dependent values, perhaps for η and certainly for the alpha half-life values and $\bar{\nu}$ ratios.

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DISCUSSION

R. W. PEELLE: In the two η-measurements, one by Macklin et al.[1] and one by Smith et al.[2], is it possible that there are correlations between the uncertainties because both experiments were done with manganese baths? Are these correlations taken into account in the least-squares programme used to evaluate the 2200-m/s data?

E. J. AXTON: In the η-measurements using manganese baths, there are three important effects for which corrections must be made.

(1) In the centre of the bath is a cavity with the neutron source inside it. It is necessary to estimate the number of neutrons which are captured in the structural material of the cavity after thermalization in the solution. The flux density at the cavity boundary is estimated by calculation and by measurements using foils. The problem is complicated by self-shielding effects of the many materials present.

(2) Fast fission caused by fission neutrons leaving the sample produces additional neutrons which should not be counted.

(3) In addition to the thermal flux striking the cavity, there is a slowing-down flux from the solution which can penetrate the cadmium shields and cause fast fission.

The correction given by most authors is 3.3%, and they estimate its uncertainty to be 6%. Even using Monte-Carlo techniques, I would think that a 20% uncertainty in the correction would be more realistic.

The geometrical arrangement in all these experiments is similar, but I do not know to what extent this implies correlation among errors. Some errors are obviously correlated, but not all.

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IV. FISSION AND CAPTURE STANDARDS

E. Fission neutron spectra

ПРИМЕНЕНИЕ КАЛИФОРНИЯ-252 В КАЧЕСТВЕ СТАНДАРТА ПРИ ИЗМЕРЕНИЯХ $\bar{\nu}$ И СПЕКТРА НЕЙТРОНОВ

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Abstract–Аннотация

USE OF CALIFORNIUM-252 AS A STANDARD FOR $\bar{\nu}$ AND NEUTRON SPECTRUM MEASUREMENTS.

In the paper, the use of spontaneous californium-252 fission events as a standard for the calibration of neutron detectors in large-scale studies is considered and the difficulties of using californium in measurements of $\bar{\nu}$ are pointed out. Measurements of the neutron spectrum *in spontaneous* californium-252 fission require the collection of data in order to derive a recommended spectrum form which could be used as a standard in fast-neutron spectroscopy. The detectors used in the studies are described.

ПРИМЕНЕНИЕ КАЛИФОРНИЯ-252 В КАЧЕСТВЕ СТАНДАРТА ПРИ ИЗМЕРЕНИЯХ $\bar{\nu}$ И СПЕКТРА НЕЙТРОНОВ.

В докладе рассматривается вопрос об использовании спонтанных делений калифорния-252 в качестве стандартного источника нейтронов для градуирования детекторов в массовых нейтронных исследованиях. Отмечаются трудности применения калифорния для измерений $\bar{\nu}$. Измерения спектра нейтронов при спонтанном делении калифорния-252 предусматривают сбор данных для выработки рекомендованной формы спектра, которую можно было бы использовать в качестве стандарта в спектроскопии быстрых нейтронов. Дается описание детекторов, используемых в исследованиях.

Большинство нейтронных измерений сводится к определению количества и спектрального состава нейтронов. Для калибровки детекторов нейтронов при решении таких задач используются сложные и трудоемкие методы, включающие в себя определение абсолютной эффективности и энергетической чувствительности детекторов.

Для измерения количества нейтронов обычно используются такие методы как поглощение нейтронов в марганцевой ванне, метод сопутствующих частиц, измерение интегральной активности, сопровождающей образование нейтронов, например в реакции $^{7}Li(p,n)\,^{7}Be$, счет числа протонов отдачи при рассеянии нейтронов на водороде. Измерения энергетической зависимости эффективности детектора обычно проводятся относительно сечений таких стандартных реакций, как $H(n,p)$; $^{10}B(n,\alpha)$; $^{6}Li(n,\alpha)$; $Au(n,\gamma)$; $^{235}U(n,f)$. Все эти методы достаточно подробно изложены в работах других авторов. Следует лишь подчеркнуть, что все они требуют от экспериментатора большого искусства и больших затрат времени.

В связи с этим представляется весьма полезным иметь источник нейтронов с хорошо известными интенсивностью и спектром нейтронов, который можно было бы использовать для градуирования детекторов в массовых нейтронных исследованиях без применения описанных выше методов. На наш взгляд в качестве такого удобного стандартного источника нейтронов можно использовать спонтанные деления калифорния-252.

Интенсивные исследования среднего числа мгновенных нейтронов при спонтанном делении калифорния-252 позволяют надеяться, что в ближайшее время неопределенность знания этой величины достигнет 0,5%. Это обстоятельство делает калифорний исключительно надежным стандартом для измерений $\bar{\nu}$ изотопов тория, урана, плутония и других делящихся элементов. Однако следует отметить и те трудности, которые возникают при использовании калифорния в качестве стандарта для измерений $\bar{\nu}$:

1. Для прецизионных измерений необходимо знание спектров нейтронов при спонтанном делении калифорния-252 и делении исследуемого изотопа.

2. Процесс деления сопровождается излучением разнообразных по природе γ-квантов в широком интервале энергий и времен испускания. В связи с этим могут возникнуть трудности в измерениях, особенно при использовании детекторов, чувствительных к γ-излучению.

3. Количество и спектральный состав испускаемых осколками нейтронов сильно скоррелированы с такими характеристиками осколков, как их массы, кинетические энергии и взаимные угловые распределения. Это обстоятельство требует глубокого изучения корреляционных свойств продуктов деления и тщательного анализа постановки опытов при измерениях таких интегральных характеристик, как $\bar{\nu}$.

4. Различие величин $\bar{\nu}$ для калифорния-252 и исследуемого изотопа требует внесения соответствующих поправок с учетом счетных качеств детектора нейтронов и электронной аппаратуры.

Вторая сторона проблемы — определение спектрального состава нейтронов при спонтанном делении калифорния-252 — на наш взгляд довольно далека от совершенства. До последнего времени основное внимание при измерениях спектров уделялось определению средней энергии. Спектр, как правило, описывался распределением Максвелла $N(E) \sim E^{1/2} \exp(-E/T)$ и тем самым исключались из рассмотрения локальные отклонения от плавной кривой, а также вариации T по разным интервалам

ТАБЛИЦА I. ЗНАЧЕНИЯ ПАРАМЕТРА I, ПОЛУЧЕННЫЕ В ДИФФЕРЕНЦИАЛЬНЫХ ИЗМЕРЕНИЯХ СПЕКТРОВ НЕЙТРОНОВ ПРИ СПОНТАННОМ ДЕЛЕНИИ КАЛИФОРНИЯ-252

Метод измерений	I	Литература
Фотопластинки	$1{,}402 \pm \begin{matrix}0{,}098\\0{,}085\end{matrix}$	1
Фотопластинки	1,57 ± 0,05	2
Время пролета	1,42 ± 0,05	2
- " -	1,56	3
- " -	1,39 ± 0,04	4
- " -	1,565	5
- " -	1,48 ± 0,03	6
Протоны отдачи		7

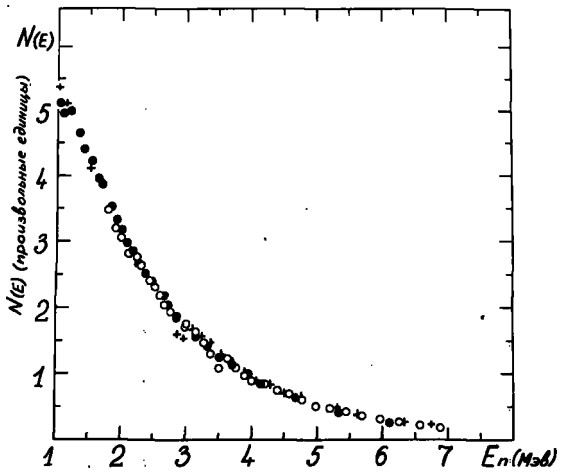

Рис.1. Спектры нейтронов при спонтанном делении калифорния-252, полученные при использовании детектора I (+), II (•), III (○).

энергии нейтронов. Очень трудно судить о степени согласия спектров, измеренных в работах [1-7], поскольку результаты представляются обычно в виде графиков в полулогарифмическом масштабе. Однако тот факт, что значения параметра T имеют значительный разброс (табл.I), говорит о том, что и формы спектров, полученных в работах [1-7], не совпадают. Так различие T на 7% означает различие максвелловских кривых на 8-25% в интервале энергий нейтронов (0,5-7) МэВ.

В нашем институте предпринимаются усилия по измерению спектра нейтронов при спонтанном делении калифорния-252 с тем, чтобы на основе полученных данных, а также числовых данных других авторов выработать рекомендованную форму спектра, которую можно было бы использовать в качестве стандарта в спектроскопии быстрых нейтронов. Для большей достоверности результатов измерения спектра нейтронов деления ведутся с использованием разных детекторов и разных методов их калибровки. На рис.1 приведены результаты измерений спектра нейтронов при спонтанном делении калифорния-252, полученные Г.Н.Ловчиковой, В.Пляскиным и др. Использовались следующие детекторы:

1. Жидкий сцинтиллятор (ϕ = 60 мм и L = 60 мм) просматривался двумя ФЭУ, включенными на совпадения. Детектор окружен свинцом, на пути нейтронов толщина свинца составляла 1 см. Порог детектора 0,17 МэВ; эффективность измерялась от порога до 1,5 МэВ с привязкой к всеволновому счетчику. Выше 1,5 МэВ использовалась расчетная кривая эффективности. Пролетная база составляла 1,0 м (+).

2. Жидкий сцинтиллятор в сосуде овальной формы. Два ФЭУ расположены параллельно и просматривают жидкость со стороны задней торцевой поверхности. ФЭУ включены на совпадения. Детектор был защищен свинцовым экраном. Толщина свинца на пути нейтронов составляла 4 см. Эффективность (•) измерялась по отношению к сечению (n,p)

рассеяния на импульсном тандемном ускорителе ФЭИ в диапазоне энергий 0,5-6,5 МэВ. В интервале от 0,2 до 2,0 МэВ эффективность измерялась на электрическом ускорителе ЭГ-I ФЭИ. Порог по нейтронам был на уровне 200 кэВ. Пролетная база при измерениях спектра нейтронов делений составляла 1,0 м (•).

3. Тот же детектор, что и в п. 2, но пролетная база составляла 1,5 м (○). Как видно, были получены хорошо совпадающие данные. Однако предстоят измерения ниже 1 МэВ, совершенствование методов калибровки детекторов, выяснение причин расхождения с результатами других авторов и и другие процедуры, прежде чем удастся выработать рекомендованную форму спектра при спонтанном делении калифорния-252 с приемлемой надежностью.

В заключение следует отметить необходимость публикования цифрового материала по результатам измерения спектров нейтронов деления.

Автор выражает глубокую признательность коллективу сотрудников спектрометрической лаборатории ФЭИ, выполнившему обсуждавшиеся в докладе измерения спектров нейтронов деления.

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DISCUSSION

W. P. POENITZ: How did you determine the efficiency of your neutron detectors?

B. D. KUZMINOV: In the lower-energy region, the detector was calibrated against a long counter. Above 1 MeV, the detector was calibrated against the n-p scattering cross-section using recoil protons from a thin stilbene crystal.

W. P. POENITZ: How did the lead shielding affect the detection efficiency and time resolution? There must have been considerable scattering in the 4-cm-thick lead shielding.

B. D. KUZMINOV: The detector was calibrated with the lead shielding in place. The time resolution was 4 ns.

L. STEWART: Were you able to measure neutron energies below 1 MeV?

B. D. KUZMINOV: No. We have measurements below 1 MeV, but the agreement among results obtained with different detectors is poor. We must study this region more closely, and I think we shall have to use a different type of detector, probably one based on the $^{6}Li(n,\alpha)$ reaction.

A. T. G. FERGUSON: Mr. Nefedov has reported previously[1] results on the fission spectrum of ^{252}Cf which showed that there might be some line-structure superimposed on the smoothly varying shape of the spectrum. Have you any additional information on such structure?

If Nefedov's ideas are correct, the time spread of the emission of the neutrons contributing to the line spectra can be quite large, perhaps some tens of nanoseconds. With such relatively short flight paths as yours — 1 m and 1.5 m — it is possible that you would not see such structure even if it did exist. Nefedov used flight paths in excess of 3 m.

B. D. KUZMINOV: All of Nefedov's results indicate the presence of this structure. We have not observed it in our own work. This is a question which we plan to investigate, and I think it would be useful if other groups would study it also.

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EXPERIMENTAL STUDY OF THE PROMPT FISSION NEUTRON SPECTRUM INDUCED BY 0.5-MeV INCIDENT NEUTRONS ON ^{235}U

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Presented by H. CONDE

Abstract

EXPERIMENTAL STUDY OF THE PROMPT FISSION NEUTRON SPECTRUM INDUCED BY 0.5-MeV INCIDENT NEUTRONS ON ^{235}U.

The spectrum of fission neutrons in the energy range 0.6 MeV to 15 MeV from fission of ^{235}U induced by 0.53-MeV neutrons has been measured by time-of-flight techniques with a large liquid scintillator. The relative flux of incident neutrons produced by the $^{7}Li(p,n)^{7}Be$ reaction was monitored with a direction-sensitive long counter. The energy scale of the time-of-flight spectrometer was calibrated over the range 0.5 MeV to 21 MeV by observing neutron groups from common source reactions and by neutron scattering from carbon. The relative efficiency of the liquid scintillator was determined in the range 0.9 MeV to 15 MeV by measuring n-p scattering, and below 0.9 MeV with the $T(p,n)^{3}He$ reaction. Pulse-shape discrimination was used to eliminate gamma background. Corrections were applied for flux attenuation and for neutrons scattered from the uranium sample. The Watt form of the fission spectrum fits the experimental data reasonably well over the entire energy range covered, whereas the Maxwellian form does not.

The shapes of prompt fission neutron spectra of the main fissile and fertile isotopes have recently attracted great interest, although it had been assumed previously that these spectra had been measured with satisfactory precision. Despite the many microscopic measurements made of prompt fission neutron spectra, the results of different experimenters disagree by amounts exceeding the given error uncertainty, which indicates the existence of large systematic errors. Furthermore, the macroscopic measurements recently performed indicated appreciably harder spectra than those extracted from microscopic measurements. The general shapes of the spectra are known, but accurate relative intensities are not well known above about 7 MeV and below 2 MeV where the microscopic and macroscopic measurements differ most. One possible reason for the discrepancies between fission spectra observed at different laboratories using a scintillation detector may be uncertainties in the measurements of the detector efficiency curve. The methods of calculating this curve or the use of angular distributions of neutron source reactions, i.e. $T(p,n)^{3}He$, $D(d,n)^{3}He$, etc., for measuring the response function do not give sufficient precision. A more accurate way which has been used in the present experiment is to measure the efficiency curve by observing angular distributions of the well-known n-p scattering process at different energies.

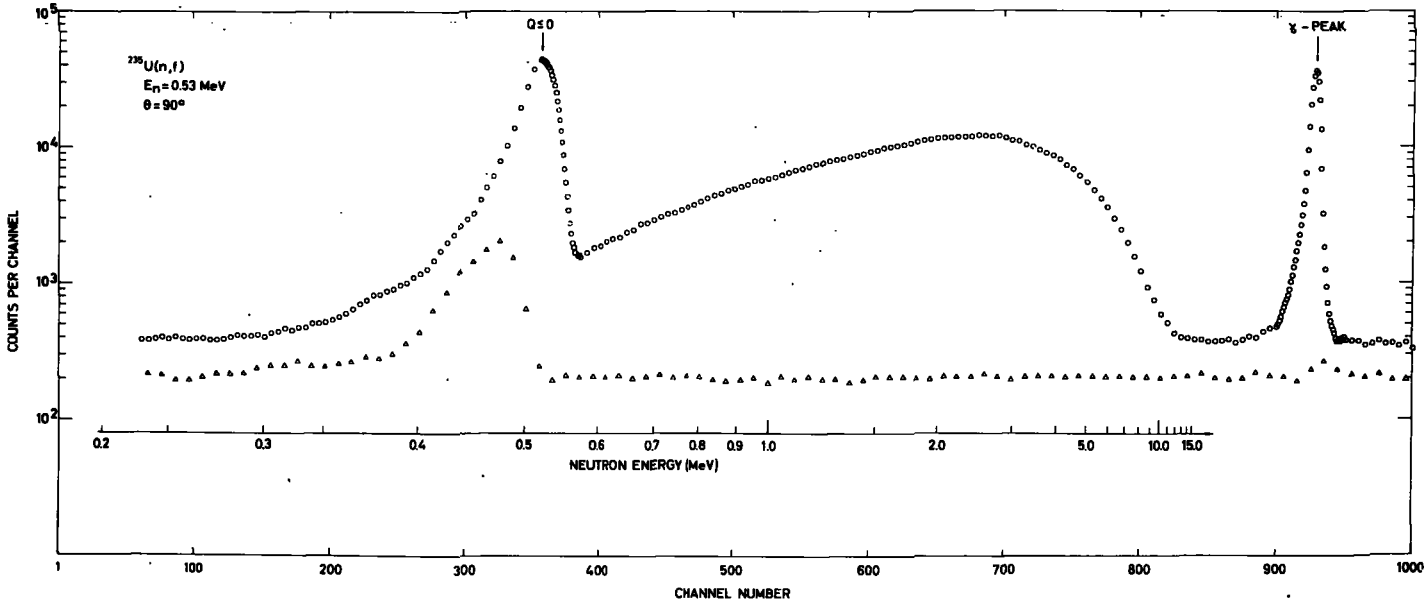

FIG. 1. Time-of-flight spectra from neutron-induced reactions in ^{235}U at 0.53 MeV incident neutron energy (circles), and background observed without any scatterers in position (triangles). The running time was about 40 hours.

This investigation is concerned with the fission neutron spectrum from ^{235}U using time-of-flight techniques at an incident neutron energy of 0.53 MeV. The purpose was to measure the fission spectrum over as large an energy range as possible, i.e. from 0.6 MeV to 15 MeV. The accuracy should allow relative measurements of fission neutron spectra from other fissile isotopes. A comparatively large liquid scintillator was chosen as a detector element in order to get a high efficiency extending over a large energy range, good time resolution and the possibility of using pulse-shape discrimination to suppress the gamma background. This experimental technique has given very positive results in that it has enabled the recording of a fission neutron spectrum with very satisfactory statistics from about 0.5 MeV up to the highest fission neutron energy. The background conditions were extremely good, even up in the high-energy range, thus giving an extraordinary accuracy for an experiment of this type. The detector arrangements were located in a large shielding of lithium-paraffin, iron and lead. The distance between the uranium sample and the detector was 300 cm at a detector angle of 90° relative to the incident neutron beam. Neutrons of 0.53 MeV energy were produced by the $^{7}Li(p,n)^{7}Be$ reaction, and the relative neutron flux was monitored with a direction-sensitive long counter. The time-of-flight spectra from neutron-induced reactions in ^{235}U at 0.53 MeV incident neutron energy are shown in Fig.1.

It is very important in measurements of this type, requiring accurate determinations of intensities in different energy intervals covering a large energy range, that the energy scale as well as the energy dependence of the neutron detector be known with high accuracy. The energy calibration of the time-to-pulse-height converter of the neutron spectrometer was performed by observing neutron groups from the reactions $T(p,n)^{3}He$, $^{9}Be(d,n)^{10}B$ and $T(d,n)^{4}He$ as well as neutron scattering from carbon. The energy covered was 0.5 MeV to 21 MeV.

The relative efficiency of the neutron detector was determined by observing neutron scattering from hydrogen at different primary energies and at different angles. For these measurements, new target facilities have been used, allowing the production of high neutron flux intensities. This equipment, in combination with the pulse-shape discrimination, made it possible to measure the detector response function with high accuracy from 0.9 MeV to 15 MeV with the n-p scattering process. The low-energy part of the efficiency curve has been measured by detecting neutrons from the $T(p,n)^{3}He$ reaction. Thus the energy range from 0.5 MeV to 15 MeV was covered. In this experiment, which required a long running time, the efficiency curve as well as the time calibration of the spectrometer were performed before as well as after the fission experiment.

At fast primary neutron energies, the analysis of fission neutron spectra recorded by time-of-flight techniques becomes somewhat complicated because of the interference between the continuous fission spectrum and neutrons emitted in competing elastic and inelastic scattering processes. The high-energy end of the fission spectrum may also be somewhat influenced by the low-energy tail of the peak from sample gammas incident on the detector. The distortion of the high-energy region of the neutron spectrum by the gamma peak has been eliminated by pulse-shape discrimination. The contributions to the fission spectrum caused by the high-energy tail of the elastic peak were taken care of by subtracting neutron scattering spectra recorded with a ^{181}Ta sample.

The corrections for flux attenuation in the uranium sample have been performed on the basis of the Monte-Carlo technique. The relative correction factor was found to vary little with the fission neutron energy, i.e. between 1.03 and 0.99. Up to now, the effects of neutron multiple scattering on the shape of the fission neutron spectrum have been considered to be small and have accordingly been neglected. However, a more careful investigation of the importance of the multiple scattering effects is highly desirable.

The standard procedure for interpreting the fission neutron spectrum from microscopic measurements has been to fit a semi-empirical function derived from nuclear evaporation theory to the experimental data. One function chosen is that proposed by Watt:

$$N_W(E) \sim \exp(-E/A) \sinh (BE)^{1/2}$$

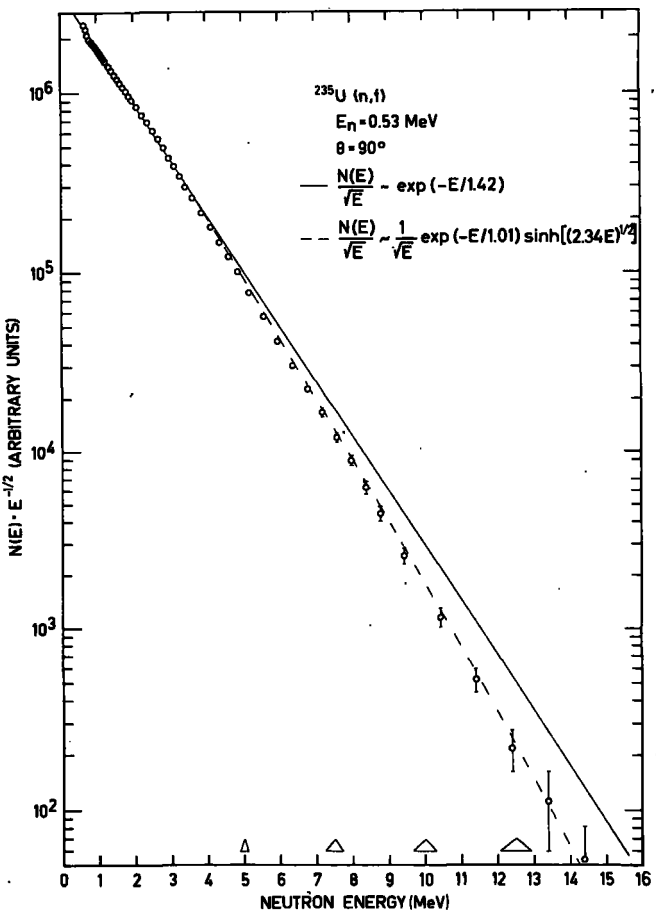

FIG. 2. Fission neutron spectrum from ^{235}U obtained at 0.53 MeV incident neutron energy. The solid line and the broken line are least-squares fits to the experimental points, assuming Maxwellian distribution and Watt distribution respectively.

Here E is the neutron energy, and A and B are constants chosen to fit the data points. Another function used is the Maxwellian distribution proposed by Terrell:

$$N_M(E) \sim E^{1/2} \exp(-E/T)$$

Here E is the neutron energy and T is the so-called Maxwellian temperature. Since there is no theoretical reason for using one or the other form, the Maxwellian distribution has hitherto been most commonly used. However, the use of a one-parameter expression leads to a lack of flexibility in making a very close fit to experimental data, in particular at high and low neutron energies.

In the present case, the experimental fission neutron distribution given as the number of neutrons per unit interval was fitted by the method of least squares with the Watt distribution and the Maxwellian distribution in the form given above. The quantity χ^2, defined as

$$\chi^2 = \frac{1}{N} \sum_{i}^{N} \left[\frac{N_i(E)_{cal} - N_i(E)_{exp}}{w_i} \right]^2$$

was chosen to compare the results of the fitting with the two functions. Here $N(E)_{cal}$ is the calculated value, $N(E)_{exp}$ is the measured one and w_i is

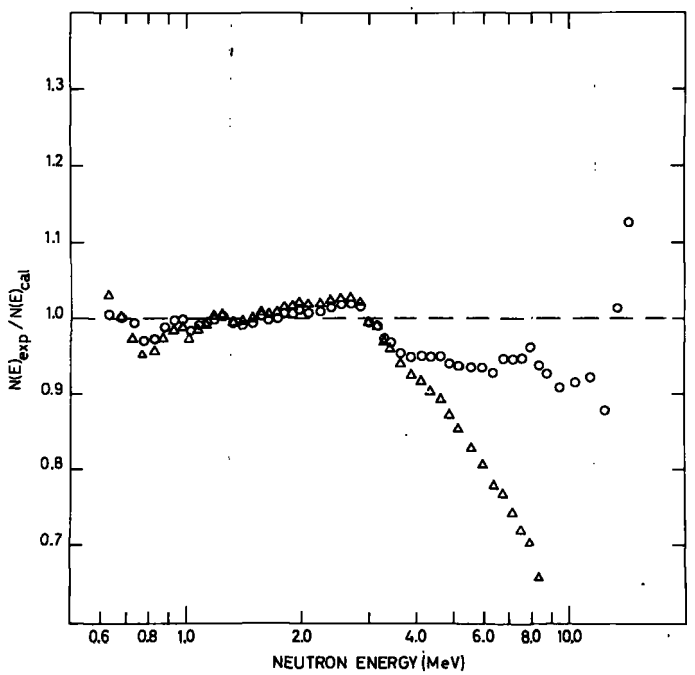

FIG. 3. Comparison between the experimental and the least-squares-fitted Watt and Maxwellian neutron distribution from ^{235}U at 0.53 MeV incident neutron energy. The circles and triangles are the quotients between the experimental values, $N(E)_{exp}$, and values calculated from the Watt distribution and the Maxwellian distribution respectively.

the experimental error. The experimental data are shown in Fig. 2 as the number of fission neutrons per unit energy interval, $N(E)_{exp}$, divided by $E^{1/2}$, with arbitrary normalization. It is shown that the Watt distribution (broken line) fits the data over the entire energy range. The best-fit parameters are A = (1.01 ± 0.03) MeV and B = (2.34 ± 0.30) MeV^{-1}. The χ^2-value is 4.3×10^{10}. The least-squares fit with the Maxwellian distribution (solid line) gave the parameter T = (1.42 ± 0.01) MeV and the χ^2-value 13.3×10^{10}. It is evident that the Maxwellian distribution does not fit the data points so well as the Watt distribution. This is also shown in Fig. 3, where the quotients between the experimental values, $N(E)_{exp}$, and the corresponding values calculated from the Watt and Maxwell distributions have been plotted versus the neutron energy.

DISCUSSION

A. T. G. FERGUSON: For the fission neutron spectrum of ^{235}U there seems to be good consensus about the shape in the energy range 0.5 MeV to about 5-6 MeV. The differences among various measurements come at lower energies and at higher energies; some are quite substantial and should be resolved.

I think it is becoming apparent that experimenters have devoted much attention to the efficiency calibrations of their detectors and to the energy calibration of the time scale. The remaining difficulties probably lie in the area of background subtraction.

L. STEWART: In studies of the ^{235}U fission neutron spectrum, I wonder whether the geometries have always been such that anisotropy of the fission neutrons does not influence the measurements as the energy of the fission-inducing neutrons is increased.

STANDARD FISSION NEUTRON SPECTRA

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Abstract

STANDARD FISSION NEUTRON SPECTRA.

This paper reviews the advantages to be derived from the availability of a standard fission neutron spectrum. The most appropriate choice of standard is considered and its status reviewed. It is concluded that the fission neutron spectrum from low-energy neutron-induced fission of ^{235}U and that from spontaneous fission of ^{252}Cf would both be valuable as standards. In the case of ^{252}Cf it is not yet possible to give a satisfactory evaluation. The situation for ^{235}U is much more satisfactory and this body of data is ready for re-evaluation.

1. INTRODUCTION

The importance of neutron standard cross-sections derives from the fact that absolute measurements from first principles are difficult and time-consuming. If appropriate standard cross-sections were available, only ratio mesurements of the required to the standard cross-section would be needed. Such ratio measurements are in almost all cases simpler and more precise. This paper is concerned with a standard fission neutron spectrum, and deals with the following questions:

(a) For what purposes would a standard fission neutron spectrum be used?
(b) What is the most suitable standard or standards?
(c) What is the current status of such standards and how well are they known?

2. USES OF A STANDARD FISSION NEUTRON SPECTRUM

2.1. Determination of fission neutron spectra

For reactor physics calculations, a substantial body of fission spectrum data is required, especially for ^{235}U, ^{239}Pu and ^{238}U. The data for ^{240}Pu and ^{241}Pu are becoming of increasing significance. There is some evidence that the shape of the fission spectrum and its average energy depend also on the energy of the primary neutron [1] and that the angular distribution is non-isotropic. The measurement of the fission neutron spectrum for five elements at several primary energies and several angles is clearly a substantial task.

Measurements of fission neutron spectra have certain components in common, whether they use time-of-flight techniques or a proton recoil spectrometer or other nuclear reactions, for instance:

(a) Determination of an observed spectrum including background
(b) Determination and subtraction of background
(c) Calibration of the time or pulse height versus energy scale
(d) Initial determination of the detector efficiency as a function of energy and its repeated verification
(e) Corrections of the reduced results for multiple scattering, etc.

Component (d) is by far the most time-consuming and makes a major contribution to the uncertainty of the final value. If all other experimental conditions can be kept constant and a standard fission source of known spectrum can replace that under study, there is no need for such careful and repeated calibration and the experiment is greatly simplified. This is a significant gain although the other measurement components remain.

2.2. Detector calibration

The efficiency variation as a function of energy of a neutron spectrometer can be determined by observing a known spectrum covering the desired energy range. A standard fission spectrum could be valuable for this. A well-known spectrum with a marked energy structure would be ideal, i.e. a spectrum with easily recognizable peaks. The fission spectrum covers the required energy range but is itself featureless. Artificial structure can be introduced by filtering through iron, carbon or boron, whose well-known resonances are clearly imprinted and give an energy calibration with numerous well-known points up to 6.5 MeV.

2.3. Semi-integral experiments

There are a number of classic semi-integral experiments such as the measurement of the fission cross-section of ^{238}U [2-4] in the fission neutron spectrum of ^{235}U. If the latter were a very precisely known spectrum, a valuable cross-check on an extremely important set of data would be obtained. Similar checks on other fission and threshold reaction cross-sections would be of great value.

2.4. Importance for determination of ν

The quantity ν is one of the few nuclear quantities known to a comparatively high precision. Axton [5] and Boldeman [6] point out that the uncertainties in the fission neutron spectra, particularly those of ^{235}U and ^{252}Cf, represent a significant component of the error in ν. The spectrum is of particular importance in reconciling results obtained with very large and comparatively small detectors.

From the above, it is concluded that a standard fission cross-section should have its place in the list of standard neutron data.

3. CHOICE OF A STANDARD

There are two main candidates for the role of fission neutron spectrum standard, i.e. that from the spontaneous fission of ^{252}Cf and that following fission of ^{235}U induced by neutrons of low energy.

3.1. Standard for fission spectrum measurement

For this role, a standard is needed whose spectrum measurement is easier than that of any of the other materials with which it will be compared. This condition is largely satisfied by ^{235}U, which has the following advantages:

(a) For time-of-flight measurements it can be used either as a solid sample or in a counter, e.g. a gas scintillator.
(b) Because of its comparatively low specific activity, amounts in excess of 1 g can be accommodated in a fission chamber.
(c) As its biological hazard is comparatively low, it can be used in the form of an uncanned solid sample, thus simplifying multiple scattering corrections.
(d) It is widely available in quantities adequate for such experiments in high isotopic purity. At incident neutron energies at which it is proposed as a standard, i.e. less than a few hundred keV, the principal impurity, ^{238}U, is largely inert.
(e) Because thermally fissile, it can be stimulated either by neutrons from a reactor or by low-energy neutrons from an accelerator.

The ^{235}U fission neutron spectrum can equally well be observed with a proton recoil or nuclear reaction spectrometer [7].

A strong case can also be made for ^{252}Cf on the following grounds:

(a) Although for time-of-flight experiments it must always be used in some form of a detector, it is expected that the majority of comparable measurements will in future be made under similar conditions.
(b) The ratio of spontaneous fission to α-particle emission is such that sources giving adequate neutron intensity can be incorporated in counters.
(c) It is widely available either as a thin exposed deposit or, usually in larger quantities, as an encapsulated source.

3.2. Standard spectrum for other purposes

For detector calibration and semi-integral experiments, ^{252}Cf has the following advantages:

(a) It requires neither reactor nor accelerator.
(b) It can be used in idealized environments where there are no problems such as, for example, in-scattering.
(c) It presents a 'point source' in either a fission chamber or in a tiny capsule.
(d) Sources of sufficient strength are now available to make such measurements quickly and with good statistical accuracy.

From the above, it can be concluded that no preference can be given either to ^{235}U or to ^{252}Cf in all fields in which their use as standards is envisaged. Both are usable in an adequate fashion in all fields. It is clear that the balance of argument which, at the Consultants' Meeting on Prompt Fission Neutron Spectra, led to their being adopted as joint standards has been maintained.

4. STATE OF KNOWLEDGE OF THE STANDARD SPECTRA

This was extensively reviewed at the Consultants' Meeting on Prompt Fission Neutron Spectra in 1971.[1] In the following, the evidence on the two standard spectra as then presented is summarized and brought up to date.

4.1. Current status of ^{252}Cf

The known measurements of this spectrum were summarized by Smith [1] and by Koster [8] in 1971. The average neutron energy varies between 2.085 MeV and 2.35 MeV with quoted errors which in general are small compared with the difference between these two figures. In his review, Smith [1] pointed out that many of the early measurements were made with weak sources but that later ones, with adequate source strength, still gave discrepant results. Jeki et al. [9] have suggested that backgrounds due to delayed γ-rays have perturbed the average energies, reducing them by as much as 0.1 - 0.2 MeV, and they favour the higher values. On the other hand, Rose [10] used long flight paths and pulse-shape discrimination, in this way avoiding such corrections, and he found a preliminary value of only 2.13 ± 0.064 MeV.

There are also discrepancies in the shape of the ^{252}Cf fission spectrum. A number of authors report an excess of neutrons above a Maxwellian at energies below 1 MeV, while others fail to observe this feature. Nefedov [11] reports some line structure in his observed ^{252}Cf spectrum which he attributes to delayed neutron groups with half-lives in the nanosecond region. Their intensity is weak, however, and would have no significant effect on the average energy deduced. I would support the comment of Koster [8] that, until further good measurements are available, it is a waste of time to try to arrive at an evaluated representation of the ^{252}Cf fission spectrum.

4.2. Current status of ^{235}U

There is a vast body of data on the fission neutron spectrum of ^{235}U; more than sixty measurements have been reported. This was reviewed by both Smith [1] and Koster [8] in 1971. Smith deduced an average value of $\bar{E} = 1.98$ MeV with an rms deviation of 0.09 MeV. The latter is rather longer than the typical values of undivided authors, i.e. less than 0.050 MeV. During the last few years, the values have systematically increased by 50 - 100 keV. Thus Johansson et al. [12] report a value of $\bar{E} = 2.07$ MeV, Rose [10] finds $\bar{E} = 2.11 \pm 0.05$ MeV, and Knitter et al. [13] find $\bar{E} = 2.02 \pm 0.08$ MeV. Although the results are consistent as to average energy, there is some divergence regarding the detailed shape of the spectra. Over a restricted energy range, say up to 6 - 7 MeV, a Maxwellian gives a good representation, but in general, when the upper limit is raised to 14 MeV, deviations are observed and the greater freedom of the Watt expression is found valuable. There are some discrepancies at these higher energies between recent measurements [10, 12]. Active programmes are in progress in a number of laboratories to attempt to eliminate these discrepancies which are significant mainly where the neutron intensity is very low.

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5. CONCLUSION

It is concluded that the fission neutron spectrum from low-energy neutron-induced fission of ^{235}U and that from spontaneous fission of ^{252}Cf would both be valuable as standards. Neither of them is yet known to adequate accuracy but, whereas a satisfactory state for ^{235}U is in sight, considerable further work is required in the case of ^{252}Cf.

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DISCUSSION

C.D. BOWMAN: To what accuracy do you think the ^{252}Cf fission neutron spectrum must be known in order to be useful as a standard?

A.T.G. FERGUSON: If it were as well known as the ^{235}U fission neutron spectrum, it would begin to be useful as a standard. If such accuracy were achieved, it might be useful to agree somewhat arbitrarily on a 'standard' shape which could be used in the interpretation of integral experiments in particular and also to tie together many diverse experiments. I think it will be at least a year before it becomes worth while to postulate standard values for the ^{252}Cf spectrum. The ^{252}Cf spectrum has such great potential utility that its precise determination is worth significant effort.

C.D. BOWMAN: By tying together diverse experiments, do you mean ratios of cross-sections?

A.T.G. FERGUSON: Yes. I mean integral ratios of fission and other reaction cross-sections in the californium spectrum.

I think the californium spectrum would be useful as a general calibration source because it is an extremely tedious job to make a point-by-point calibration of detectors of the type used in time-of-flight measurements.

B.D. KUZMINOV: I support Mr. Ferguson's opinion regarding the usefulness of the ^{252}Cf spectrum as a standard. I think it is worth while to invest the effort necessary to resolve discrepancies apparent in the currently available data because californium would then become a convenient instrument for a whole series of measurements.

If the ^{235}U spectrum is the only standard, one must always have an accelerator or reactor in order to make use of it. When ^{235}U is used with an accelerator, the fission neutron spectrum obtained cannot extend to energies lower than the energy of the neutrons used to induce fission. To use ^{235}U in a thermal reactor with the time-of-flight method it is necessary to have some kind of fission chamber or a very powerful reactor in order to obtain sufficient intensity. When a large quantity of ^{235}U is used, there is an uncertainty associated with the angular distribution of neutrons with respect to the fission fragments. Because of these difficulties with ^{235}U it seems to me that ^{252}Cf would be a very convenient standard.

E.J. AXTON: I also would like to support the use of ^{252}Cf as a standard. It would be extremely easy to make fission-spectrum-averaged cross-section-ratio measurements using a ^{252}Cf neutron source. The neutron source could be installed in a large room, surrounded by a ring of fission counters, and allowed to operate for weeks. This type of experiment could even be done in a laboratory which has no accelerator or reactor. Very thin sources, which would imply very slow count rates, could be used because the time-duration of the experiment would be unimportant. Such experiments could be usefully carried out even now, while the ^{252}Cf spectrum is not very well known, because later when the ^{252}Cf spectrum becomes better known, the measurements could be correctly related to cross-section-ratio measurements in other spectra.

C.D. BOWMAN: Mr. Grundl at the National Bureau of Standards (NBS) has carried out measurements using a ^{252}Cf fission neutron source located at the centre of a room, which is approximately a cube with about 30-ft sides. He has obtained absolute cross-sections, averaged over the ^{252}Cf fission neutron spectrum, for ^{238}U, ^{235}U and their ratio. He considers this technique to be an important tool in the integral measurement programme at NBS.

R.W. PEELLE: In the time-of-flight measurements of the ^{252}Cf neutron spectrum it is possible that the observed structure might be due to an instrumental effect such as 'ringing' if the observed peaks occur at equal time intervals. I have experienced such problems in time-of-flight measurements even using the best commercially available equipment.

A.T.G. FERGUSON: Our ^{235}U spectra taken at long flight paths show no comparable structure; they are absolutely smooth. I agree with Mr. Peelle, however, that there is not yet sufficient evidence to show that structure in the ^{252}Cf spectrum is real.

B.D. KUZMINOV: The structure observed by Nefedov changed position in the time scale when the flight path was changed, so the effect is apparently not instrumental.

V. CRITERIA FOR STANDARDS FOR NEUTRON DATA MEASUREMENTS: GENERAL COMMENTS

The last paper of the previous section by A.T.G. Ferguson contains remarks relevant to this section.

CONNECTIONS BETWEEN AND COMMON PROBLEMS FOR LOW-ENERGY (eV REGION) AND HIGH-ENERGY (keV TO MeV REGION) PRECISION MEASUREMENTS

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Abstract

CONNECTIONS BETWEEN AND COMMON PROBLEMS FOR LOW-ENERGY (eV REGION) AND HIGH-ENERGY (keV TO MeV REGION) PRECISION MEASUREMENTS.

Experience from low-energy precision measurements should be helpful when moving to higher energy: (1) The definition of the samples (number of atoms, isotopic composition, chemical purity, weight, choice of preparation technique, etc.) should be carefully considered. (2) Measurements should be performed on several samples to permit cross-checks, possibly with other laboratories. (3) Where possible, overlapping energy regions between different machines and (or) detection techniques should be available to take advantage of the 2200-m/s reference values. (4) Analogous detection problems exist in low-energy and higher-energy experiments, e.g. 2π-geometry fission fragment counting. Problems connected with these measurements can sometimes better be studied at low neutron energy because of the available high-intensity neutron beams.

INTRODUCTION

The paper presented at the Helsinki conference on the situation in 1970 concerning standard quantities [1], and especially the EANDC standards symposium held at Argonne National Laboratory [2], are taken as starting points for the following comments.

In the low-energy region, we reach now a 0.5% accuracy for fission cross-section measurements and even somewhat better for a few standard total (and absorption) cross-sections. To achieve this, it was imperative to develop new measurement techniques, but also the target preparation and definition had to be considerably improved. As was proved during this Panel, we are now moving in the direction of higher neutron energies for these standard cross-sections. At this moment we should not make the same mistakes that were made in the earlier low-energy precision measurements, nor forget to use the precise low-energy cross-section values where possible.

From experience at the Central Bureau for Nuclear Measurements (CBNM), I would like to make a few comments on connections between low-energy (eV region) and higher-energy (keV to MeV region) precision measurements.

TABLE I. ISOTOPIC COMPOSITION OF EVAPORATED ELEMENTAL BORON LAYERS

Target number	Witness layer number	$\frac{\text{at.\% } ^{10}B \text{ (witness)}}{\text{at.\% } ^{10}B \text{ (standard)}}$	^{10}B atomic concentration (at.%)
B 92	32	1.0095 ± 0.0010	20.012 ± 0.030
	33	1.0096 ± 0.0010	20.014 ± 0.030
	34	1.0092 ± 0.0010	20.006 ± 0.030
	35	1.0088 ± 0.0010	19.998 ± 0.030
	36	1.0085 ± 0.0010	19.993 ± 0.030
		Average value	20.005 ± 0.030
B 86	37	1.0104 ± 0.0010	20.030 ± 0.030
	38	1.0114 ± 0.0010	20.050 ± 0.030
	39	1.0105 ± 0.0010	20.032 ± 0.030
	40	1.0109 ± 0.0010	20.040 ± 0.030
	41	1.0109 ± 0.0010	20.040 ± 0.030
		Average value	20.038 ± 0.030
B 75	47	1.0193 ± 0.0010	20.206 ± 0.030
	48	1.0198 ± 0.0010	20.217 ± 0.030
	49	1.0193 ± 0.0010	20.206 ± 0.030
	50	1.0193 ± 0.0010	20.206 ± 0.030
	51	1.0200 ± 0.0010	20.220 ± 0.030
		Average value	20.211 ± 0.030
B 98 (not accepted)	42	1.0183 ± 0.0010	20.187 ± 0.030
	43	1.0189 ± 0.0010	20.199 ± 0.030
	44	1.0208 ± 0.0010	20.236 ± 0.030
	45	1.0201 ± 0.0010	20.222 ± 0.030
	46	1.0204 ± 0.0010	20.228 ± 0.030
		Average value	20.214 ± 0.030

DEFINITION OF SAMPLES

Only measurements on well-defined samples are valuable. This means that the number of atoms (e.g. in a layer) has to be determined by several really independent methods. Often the physicist relies on the number of atoms given by the target constructor without much verification.

Out of CBNM experience, I would like to give an example of the possible errors in cross-section values from ill-defined samples. For several measurements we used evaporated layers of elemental boron. Natural

boron metal was used as we did not have a sufficient quantity of highly purified ^{10}B metal necessary for the evaporation in the rather reduced geometry needed to obtain the required layer homogeneity. As a control, the isotopic composition of the starting material and the isotopic composition of each evaporated elemental boron layer was checked by our mass-spectrometric laboratory [3], and we noticed an increase in the ^{10}B to ^{11}B ratio. The atomic ^{10}B concentration of the starting material, the CBNM isotopic boron standard, was (19.824 ± 0.020) at.%. It was established that for each evaporated target this atomic ^{10}B concentration was slightly but significantly different. So for each layer to be used, five small witness layers were evaporated during the evaporation of the target, and afterwards they were carefully analysed by the mass-spectrometric laboratory.

Some typical results in Table I are very instructive. It can be seen that the ^{10}B atomic concentration for all evaporated layers is higher than 20% or at least 1% higher (with a range from 1% to 2.5%) than the concentration of the starting material. A second point is that for each layer the isotopic concentration has to be measured because the enrichment varies from evaporation to evaporation although the evaporation conditions are kept similar. The variations from target to target reach regularly 1% (compare, e.g., B 92 and B 75) and occasionally even more than 2%. The normal spread in the measured atomic ^{10}B concentrations among the five small witness layers evaporated on gold backings and situated around the target is 0.1% (see all witness layers of targets B 92, B 86 and B 75 in Table I). A target like B 98, which is shown in Table I only for comparison, is not accepted as a 'good foil' because the deviation among the witness foils reaches 0.2% and there might be a variation of the ^{10}B atomic concentration across the foil. The errors indicated here consist of the statistical scattering of the measurements as well as of an estimate of the maximum possible systematic errors. The measurements were repeated until the systematic errors were predominant, so the error on the average is equal to the error on the measurements of a single witness layer. The possible error on a measured cross-section, only from the isotopic composition of the layer, could be as high as 2% if such a systematic and thorough mass-spectrometric study of the layers had not been performed.

Now we only have the relative amount of ^{10}B in the evaporated deposits. The total weights of the layers were determined by weighing the total deposit in ultra-high vacuum. Here we need the chemical analysis of the layers. The contaminants were essentially metallic impurities, and the carbon content of the layers was always checked. These measurements showed again the importance of taking witness layers for each of the boron foils. Not only was there a correction to be made but also the correction varied substantially from layer to layer.

In Table II, the characteristics of a few selected boron targets are listed. Variations from 994 ppm (± 20%) for B 69 up to 3883 ppm (± 20%) for B 64 are apparent for the metallic impurities. This means that a correction on the total mass of 0.38% has to be applied for B 64 and a correction of 0.10% for B 69. The correction is meaningful and shows again the importance of witness layers. Also for the carbon content, fluctuations of over a factor of two are noticed, i.e. for B 73 1080 ppm (± 20%) and for B 69 2500 ppm (± 20%), resulting in mass corrections of 0.25% and 0.11% respectively. The total correction for impurities can reach more than 0.5% and has to be checked for each layer independently.

TABLE II. CHARACTERISTICS OF ELEMENTAL BORON LAYERS

Quantity	Unit \ Target number	B 73	B 74	B 64	B 69
Total mass deposited and determined under vacuum: M	μg	412.6	525.0	787.2	493.8
Estimated error on M	μg	2	2	2	2
Metallic impurities	ppm	2215	2171	3883	994
Error on metallic impurities	%	15	15	20	20
Carbon content	ppm	1080	1450	1100	2500
Error on carbon content	%	20	20	20	20
Total mass of boron deposited: M_B	μg	411.2	523.1	783.3	492.1
Error on M_B	%	0.45	0.45	0.35	0.48
Calculated deposit area 0	cm^2	11.357	11.360	11.454	11.453
Error on area	%	0.05	0.05	0.05	0.05
Surface density of boron, $m_B = M_B/0$	$\mu g/cm^2$	36.21	46.05	68.39	42.97
Error on surface density	% $\mu g/cm^2$	0.50 0.18	0.50 0.23	0.40 0.27	0.53 0.23
Isotopic content of ^{10}B	at. % wt%	20.450 18.950	20.330 18.837	20.127 18.647	20.228 18.740
Error on isotopic content	%	0.32	0.32	0.23	0.23
Mass of ^{10}B	μg	77.922	98.536	146.06	92.220
Error on ^{10}B mass	%	0.55	0.55	0.41	0.53
Surface density of ^{10}B	$\mu g/cm^2$	6.861	8.674	12.752	8.052
Error on surface density of ^{10}B	% $\mu g/cm^2$	0.55 0.038	0.55 0.048	0.42 0.054	0.53 0.043

Another problem which turned up was the effect of the temperature of the diaphragm used in the evaporation. For instance, the evaporation of the targets B 73 and B 74 on the one hand and that of B 64 and B 69 on the other hand was performed at different diaphragm temperatures, which resulted in about 1% difference of the calculated deposit areas and could give a 1% difference of the calculated layer thickness. Of course, during the evaporation the diaphragm temperature is constantly controlled. Now we can deduce the total ^{10}B weight and also the surface density of the layers. The errors are in the vicinity of 0.5%. But without all these controls and systematic error analysis, the errors could easily reach several per cent on the number of atoms of ^{10}B.

We had similar problems with solutions of $^{6}Li_2SO_4$ in D_2O which yielded a σ_a^0 cross-section for ^{6}Li that differed from the 'now accepted' value by a few per cent. We are checking at present the origin of the discrepancy and also if the same discrepant results would be obtained with transmission measurements on ^{6}LiF.

NECESSITY OF USING SEVERAL TARGETS

Even now, single targets are often used for the measurements. Under no circumstances should this be permitted. A set of targets should be prepared and they should be compared with each other by non-destructive means, e.g. verification of their relative number of atoms, based on alpha counting (fission foils), neutron-induced alpha counting (^{10}B, ^{6}Li) or neutron-induced fission counting (^{235}U, etc.), preferably at low energy where high-intensity neutron beams are available, the cross-sections well known and the detection geometry can be adapted to the count rate.

As an example, it is assumed that a laboratory wants to measure σ_f of ^{235}U in the keV-region. CBNM would advise it to order five foils, preferably from an institute with good knowledge in target preparation. The experimentalist in charge should consult the laboratory which prepared the samples about the choice of the isotopic composition in view of the present state of knowledge of alpha half-lives and their relation to the determination of the isotopic composition of the samples. In some cases, a small amount of highly alpha-active material (such as ^{234}U) is not cumbersome for the fission measurements as long as one stays below the fission threshold of ^{234}U, but it helps considerably in precise low-geometry alpha counting to determine the amount of material by this method as it increases substantially the specific activity of the material. For instance, in order to have as high an enrichment as possible, a ^{235}U sample with 99.9% ^{235}U is ordered. However, the sample received contains 0.1% ^{234}U. With such a target the definition from alpha-activity measurements will be rather inaccurate as it will not be possible to determine the amount of ^{234}U with sufficient accuracy although it still contributes to a great extent to the alpha activity. However, when one can tolerate 1% of ^{234}U in the fission measurement, the accuracy achieved with alpha low-geometry counting will be even better than that achieved with most destructive methods applied on one sample only.

The choice of the material having been made and the target having been prepared by the best available method, the five foils can be intercompared by low-geometry alpha counting and fission counting in the institute preparing

the foils. This institute keeps two foils which are checked at regular intervals. On receipt of the three other foils, the experimentalist can compare them with his own alpha equipment (preferably in low geometry, perhaps using 2π-detection, although in this case the corrections may depend strongly on the layer thicknesses), and perhaps by fission counting when available. If there is disagreement between the measurements, he can immediately contact the laboratory that delivered the foils. If there is agreement, he can make a proper choice of one layer (or use all three layers), depending on the time available and the count rate used in his experiment. Once the measurements are performed, the foils are again checked for their relative activities. When only one foil has been used, it should nevertheless be compared with the two others. Then the foils are returned to the preparation laboratory where they are now re-measured, together with the two witness foils previously kept there. The intercomparison is very useful for establishing the error limits on eventual loss of material during shipment or while using the layer(s) in the experiment.

OVERLAPPING REGIONS

It would be extremely helpful to have data for overlapping neutron energy regions of reactor neutron measurements (chopper, crystal spectrometer) and linac experiments which on the high-energy side would overlap with Van de Graaff neutron experiments. In this way, measurements of the low-energy end (i.e. the 2200-m/s values) and other 'spot'-point accurate measurements would be better implemented.

At CBNM, we run at present a $^{235}U(n, f)$ experiment relative to $^{10}B(n, \alpha)$ where the time-of-flight decoder is programmed in such a way that a region of the channels covers an energy range from 7 to 15 eV, which is linked to our chopper experiments [4], and is further programmed to start the time-of-flight analysis from 100 keV down to a few hundred eV. Of course, this condition of detecting a part of the low-energy neutrons limits the repetition rate of the linac (or the distance between target and detector) and in this way the intensity, but the cross-sections obtained can be connected finally to the 2200-m/s values.

2π-GEOMETRY FISSION EXPERIMENTS

In the higher-energy region, fission measurements are usually performed with a 2π-geometry ionization chamber or with proportional or gaseous scintillation chambers, depending on the specific alpha activity of the fissile material used. For reasons of intensity, low-geometry measurements are very difficult or even excluded. In such a 2π-geometry, the correction for self-absorption depends strongly on the homogeneity of the foil (preparation method). Generally, a very approximate method is used to determine the number of fission events where the fragments are completely stopped in the foil. The fraction of such events where the fragments are completely stopped is calculated as follows:

$$t^2 + \sigma^2 / 2t\,R \qquad (1)$$

t being the average thickness of the layer and σ the thickness spread over the area, which depends very strongly on the preparation technique; R is the range of the fission fragments in the source material. From CBNM experience, evaporated sources have the best homogeneity. When not sufficient material is available, the electrospraying technique applied with precautions also yields good results. However, often the correction remains of the order of several per cent. Also the extrapolation to zero bias below the discriminator setting is liable to rather subjective criteria and the error assigned can be open to doubt. The differential relation giving the fraction of pulses lost, ΔP, for a bias change ΔB (or ΔE_B in the energy scale) is given as follows:

$$\Delta P = \frac{t}{2(R_0 - R_B)^2} \frac{\partial R_B}{\partial E_B} \Delta E_B \qquad (2)$$

where R_B is the range corresponding to the bias B, and R_0 is the full range. To evaluate Eq. (2), the density of the energy loss along the path has to be known. This long-standing problem has not been solved satisfactorily, especially at the end of the fission fragment range where atomic collisions become important and make extrapolations of this type questionable.

Backscattering of a fission fragment into the counter increases the pulse height of the other fragment pulse but does not result in an extra recorded pulse. Thus, contrary to the 2π-measurements of alpha particles, there is no backscattering correction for fission fragments from foils of zero thickness. For thicker foils, the backscattering of the partner fragment of a totally absorbed fragment may cause a pulse for a fission event that was supposed to be stopped in the layer, and in this way backscattering slightly reduces the correction for total absorption in the foil but increases the uncertainty of the correction.

From this short discussion it follows that the incomplete information available on fission fragment scattering is insufficient, and for this reason the CBNM is starting an experiment on the scattering of a collimated beam of fission fragments on materials of different (A, Z)-values. Again, this problem can be better studied at low energy because of the available high-intensity neutron beams. In this way, valuable information will be obtained for experiments with lower intensity at higher neutron energy.

CONCLUSIONS

Both low-energy and high-energy measurements have certain problems in common such as sample definition and detector efficiency, and it is sometimes easier to resolve them in the low-energy region because high-intensity neutron beams are available there. Overlapping measurements with different types of machines are needed to take advantage of data available at 2200 m/s neutron velocity and of data available at isolated energies such as 14 MeV.

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VI. SUMMARIES, CONCLUSIONS AND RECOMMENDATIONS

SUMMARIES, CONCLUSIONS AND RECOMMENDATIONS

1. METHODS AND TECHNIQUES OF NEUTRON FLUX MEASUREMENTS

Neutron flux measurements are divided into two groups: those methods that may be called absolute because they do not depend on the knowledge of any cross-section and those that are relative in that the flux determination depends upon a known cross-section.

1.1. Absolute methods

The absolute methods can be subdivided into three groups: associated-particle, associated-activity, and total-absorption methods.

1.1.1. Associated-particle method

This method makes use of neutron-producing reactions in which the neutron is produced in conjunction with an associated charged particle of unique type and energy. In the frequently used reactions $D(d,n)^3He$, $T(p,n)^3He$ and $T(d,n)^4He$, the neutron production is associated with the production of a uniquely characterized ^3He, ^3He or ^4He charged particle, respectively. With these reactions, the associated-particle method can be used in either a 'tagged' mode, where the neutron and charged particle are detected in coincidence, or in an 'untagged' mode, where the neutron and charged-particle fluxes are measured independently.

The $T(p,n)^3He$ reaction has been extensively used recently at Cadarache and at the Central Bureau for Nuclear Measurements (CBNM), Geel, for absolute calibration of detectors for use in neutron cross-section measurements. The method is successful from 100 or 200 keV up to about 6 MeV. Much effort has been expended on the development of the method, and about 2% accuracy can be achieved in flux determinations. No single error dominates, and improvements must be along several lines. Among the limitations are charge exchange and scattering in the target at low incident particle energies, solid-angle determinations and backgrounds. Conceivably, 1.3% accuracy could be reached.

The $D(d,n)^3He$ reaction can be used with the same apparatus and similar accuracy as the $T(p,n)^3He$ reaction. Below about 100 keV deuteron energy, a simpler apparatus can be used successfully.

The $T(d,n)^4He$ reaction has been used with the associated-particle method for accurate flux determination at 14 MeV neutron energy, but no measurements were reported at this meeting. One or two per cent accuracy is possible.

1.1.2. Associated-activity method

In this method the residual radioactivity left in the neutron-producing target by the source reaction is used to determine the total neutron production in the target. It is capable of good accuracy, 1% or 2%, in determining neutron production but it is limited to a small number of suitable reactions.

Generally, it is used for spot checks at a few energies and in cases in which a considerable neutron energy spread is acceptable. Examples are the reactions $^{7}Li(p,n)^{7}Be$ at threshold which produces 30-keV neutrons, $^{51}V(p,n)^{51}Cr$ which produces neutrons up to 600-700 keV, $^{65}Cu(p,n)^{65}Zn$ and $^{57}Fe(p,n)^{57}Co$. This method has been used recently in neutron standard determination by several groups for detector calibrations and cross-section measurements.

1.1.3. Total-absorption method

The total-absorption method depends upon absorption of essentially all neutrons incident upon a detector, and upon detection of those neutrons with an efficiency which is independent of neutron energy. While some detectors of this type, such as long counters, are often used only as relative flux monitors and are calibrated by other means, many total-absorption detectors are absolute flux monitors. Those in use at present include the manganese and vanadium baths and various detectors described as 'black' or 'gray' detectors.

An ancient example of the use of the total-absorption method as an absolute flux monitor is the manganese bath method in which incident neutrons are thermalized by scattering in the bath, captured by ^{55}Mn, and detected by the induced ^{56}Mn activity. In an alternative application, neutron sources which have been calibrated absolutely by the manganese bath technique are used to establish detector calibrations.

A recent version of the total-absorption method is the black detector of Poenitz in which a collimated neutron beam falls into a well in a large liquid or plastic scintillator where the neutron loses essentially all of its energy by scattering before it can escape. The proton and carbon recoil atoms produce a prompt scintillation for nearly 100% of the incident neutrons.

The best accuracy with a total-absorption technique has probably been achieved with the manganese bath method. However, it should be noted that while the experts claim 0.3% accuracy, cross-section measurements usually give other errors that are much larger. The various total-absorption detectors which respond promptly, such as the so-called black and gray detectors, often have 2% accuracy.

1.2. Relative methods

Many neutron flux determinations rely on well known cross-sections. Commonly used for reference are the light-element reactions $^{1}H(n,p)$, $^{6}Li(n,\alpha)T$, $^{10}B(n,\alpha)^{7}Li$ and $^{3}He(n,p)T$.

1.2.1. $^{1}H(n,p)$

Observation of recoil protons following neutron scattering from hydrogen is commonly used for measurement of fast neutron flux. At the low-energy end, counters filled with hydrogenous gas are used, and protons recoiling at all angles are detected. The lower-energy limit of a few keV is determined by the small amount of energy available for observation of the recoiling proton. Above 50 keV, 2% or 3% accuracy can be achieved if considerable care is used. A similar proton-recoil arrangement is often used with solid or liquid scintillation counters, but the non-linearity of pulse height with particle energy complicates their application. An accuracy of 4% to 10% is commonly obtained.

For neutron energies above 1-2 MeV the counter telescope is frequently used. In this system a small fraction of the recoil protons from a solid proton radiator of hydrogenous material is selected by means of an aperture located at some distance from the radiator and usually positioned to select protons near zero degrees to the neutron beam. A suitable detector behind the aperture counts with 100% efficiency those protons which pass the aperture. The fraction of protons counted is determined by the easily measured geometry factor of the aperture relative to the radiator. The accuracy of flux measurements with proton telescopes is about 2%. At low energies, background problems and determination of hydrogen masses limit the accuracy, and at high energy the error in the cross-section for production of forward-scattered protons dominates. At 14 MeV this latter error is about 2%.

1.2.2. $^{6}Li(n,\alpha)T$

For neutrons of energy below 100 keV the $^{6}Li(n,\alpha)T$ reaction is one of the most commonly used. Although solid lithium compounds can be introduced into counters for measurement of fluxes with this reaction, this method is not in common use because of the convenience of lithium glass scintillators, which are nearly 100% efficient for the detection of the charged reaction products. Care must be taken that corrections for neutron scattering in the glass and its surroundings are accurate and that the lithium content of the glass is known. The lithium content is commonly determined by measuring the transmission of the glass at low neutron energy where $^{6}Li(n,\alpha)$ absorption dominates. Aside from errors in the $^{6}Li(n,\alpha)T$ cross-section, about 2% accuracy can be achieved below 100 keV with these detectors.

1.2.3. $^{10}B(n,\alpha)$

The $^{10}B(n,\alpha)$ cross-section is well known below about 50 keV. Below about 10 keV, neutron flux is monitored using this reaction by detection of alpha particles; above 10 keV, the detection of the gamma rays from one branch of the reaction seems to be more popular. The detection efficiency for the gamma rays can be determined by a coincidence measurement, but this is seldom done. The reaction usually is used for shape determination and is sometimes normalized at thermal energy. Detectors based on the $^{10}B(n,\alpha)$ reaction can be calibrated to about 2% excluding error in the cross-section.

1.2.4. $^{3}He(n,p)$

This reaction is seldom used as a standard. Probably the unavailability of pure ^{3}He was initially a problem. Recent developments suggest that this reaction will be used more frequently in the future.

Recommendation

Absolute measurements of neutron flux by the associated-activity method are dependent upon absolute measurements of activities from radionuclides such as ^{7}Be, ^{51}Cr, ^{65}Zn and ^{57}Co. Standards laboratories should undertake an international comparison of the absolute calibration of such activities in order to evaluate the accuracy achievable with the available techniques.

TABLE I. SUMMARY OF RECENT RESULTS ON THE $^{6}Li(n,\alpha)$ CROSS-SECTION

Measurement	Energy range	Flux measurement	Normalization
Fort and Marquette [1] ^{6}Li glasses	15 keV - 1.7 MeV	Flat response detector and associated particle	Absolute
Coates et al. [2] ^{6}Li glasses	1.5 keV - 400 keV	Harwell black detector	Between 2 keV and 10 keV to $149.5/\sqrt{E}$ b
Poenitz and Meadows [3] ^{6}Li glasses	90 keV - 600 keV	Gray detector	Absolute
Clements and Rickard [4] ^{6}Li sandwich	160 keV - 3.9 MeV	Harwell long counter and ^{238}U cross-section	Uttley and Diment value between 300 keV and 500 keV

2. LIGHT-ELEMENT STANDARDS

2.1. The $^{6}Li(n,\alpha)$ cross-section

Four recent determinations of the $^{6}Li(n,\alpha)$ cross-section have been reported at this meeting. These are summarized in Table I. Considering first the common energy region across the 250-keV resonance, the ^{6}Li glass results [1] agree to an accuracy of ± 4% between about 150 keV and about 400 keV if systematic energy shifts of up to about 5 keV are accepted and if the data of Poenitz and Meadows [3] are renormalized down by approximately 5%. (This latter shift is acceptable since Poenitz's ^{6}Li-mass assay is preliminary.) The ^{6}Li sandwich detector results [4] in this energy region are known to be inaccurate because of resolution effects and may be ignored. Further work is needed to establish more accurately the ^{6}Li content of the glasses used in the Van de Graaff experiments, and there are some differences of detail in the multiple scattering corrections used by different authors. The correct energy scale in this region must be established by further experiments.

As far as the experimental (n,α) data are concerned, these represent a considerable improvement since the 1970 Argonne Symposium when, due to lack of agreement among the (n,α) data coupled with likely experimental deficiencies, it was recommended that the value of the $^{6}Li(n,\alpha)$ cross-section calculated by Uttley and Diment [5] from accurate total cross-section measurements represented the best available value. Uttley and Diment's cross-section was given some support by a preliminary measurement by Coates et al. The revised data of these authors, however, together with the other data referred to above, are approximately 12% below the Uttley and Diment value at the peak of the cross-section. These results cast doubt on the validity of deriving the (n,α) cross-section from total cross-section measurements with a theory which treats the resonance around 250 keV as a pure single level superimposed on an S-wave background. A calculated

total cross-section which correctly represents the experimental total cross-section always results in a calculated (n,α) value which is too high compared with experiment. This is illustrated in Figs 2 and 3 of the paper of Fort and Marquette in these Proceedings [1] and is also confirmed by some calculations of Meadows and Whalen [6]. It appears likely that a more sophisticated attack is needed before the theoretical prediction of the (n,α) cross-section over the resonance peak is capable of being consistent with the most accurate experimental values. A possible refinement is to consider the likelihood of interference from the just-bound level at 6.64 MeV in ^{7}Li.

Turning now to the energy region below 100 keV, it is considered that the reservations on the reliability of the prediction of the (n,α) cross-section using the simple theory do not significantly alter the value of the (n,α) cross-section recommended at the 1970 Argonne Symposium for energies below approximately 50 keV. At 100 keV, the uncertainty in the P-wave contribution is not likely to be greater than 2%. Unfortunately, the spread in the experimental data is about ± 8% near this energy.

Above 300 keV, the accuracy of the cross-section is difficult to evaluate. The Fort and Marquette [1] data are substantially higher than those of Clements and Rickard [4] which are normalized to the Uttley and Diment [5] value between 300 and 500 keV. No reasonable renormalization helps the situation significantly. It should be noted that the values of σ(n,α) obtained by subtracting the σ(n,n) data from the total cross-section data in this energy region support the Fort and Marquette data [1] although the values obtained are not accurate to better than about 15%. It must be concluded that in this energy region more measurements are urgently needed.

Recommendations

(a) Work should be continued to establish the ^{6}Li(n,α) cross-section to the accuracy requested for nuclear energy programmes (see WRENDA). A further incentive lies in the need for measurements of the lower-energy (less than 500 keV) portion of the fission neutron spectrum where ^{6}Li glass detectors may have to be used.

(b) Effort should be directed towards obtaining a more sophisticated theoretical treatment with close collaboration among the laboratories chiefly concerned (Argonne, Cadarache and Harwell).

(c) The present efforts to establish the correct energy scale over the approximately 250-keV resonance should be continued.

(d) Another white-spectrum measurement of the total cross-section should be made to cover the energy range from about 100 eV to about 5 MeV. At present, the only white-spectrum measurement is that of Uttley and Diment [5] which should be confirmed because it has been heavily relied upon. It is important that the proposed measurement covers the energy range which is accessible to Van de Graaff accelerators using monoenergetic neutron sources and, in addition, extends to lower energies which are inaccessible with Van de Graaffs.

(e) Further total cross-sections should be measured by suitable methods to resolve the discrepancy at higher energies (above 500 keV).

(f) The accuracy to which the ^{6}Li content of glasses is known should be improved.

(g) More angular distribution measurements for the (n,α) reaction are needed above 20 keV to provide a correct basis for interpretation of measurements performed with ^{6}Li sandwiches.

2.2. The $^{10}B(n,\alpha)$ cross-section

Since absolute proton recoil counting with good time resolution has not been demonstrated for neutron energies below 100 keV, other light-element standards are required to extend the energy range up to 1 MeV. Because of the resonance structure near 250 keV in the $^{6}Li(n,\alpha)T$ reaction, a boron standard would also receive useful application up to 1 MeV.

The $^{10}B(n,\alpha)$ reaction (ground-state plus excited state) seems potentially useful to about 1 MeV but is not well established above 80-100 keV. Recent data do not indicate changes since the 1970 Argonne Symposium in the status of the cross-section below 40 keV, but the new measurements of Friesenhahn et al. [7] for the (n,α) reaction are approximately 5% higher at 10 keV and up to 50% higher around 420 keV.

The $(n,\alpha_1\gamma)$ reaction seems potentially more useful at the higher energies (10 keV - 1 MeV), provided the 478-keV gamma ray can be resolved. Above 100 keV the new data of Friesenhahn et al. for the $(n,\alpha_1\gamma)$ reaction and the preliminary results of Coates et al. [8] agree with each other but show systematic disagreement with current evaluated data files and with earlier measurements. This disagreement is approximately 7% at 100 keV and increases with increasing energy.

Recommendation

Further experimental work is recommended in order to establish the total (n,α) and the $(n,\alpha_1\gamma)$ cross-sections to the necessary precision up to approximately 1 MeV. In consideration of the recent experiments of Coates et al. and of Friesenhahn et al., recommended values of the ^{10}B cross-sections above 100-200 keV are difficult to determine.

2.3. The $^{3}He(n,p)T$ cross-section

The cross-section for this reaction is known to 2% below 100 eV, to about 5% below 10 keV and less accurately above 10 keV.

Recommendation

Further measurements at energies above 100 keV are recommended to achieve the accuracy requested for this standard cross-section.

2.4. The $^{1}H(n,p)$ cross-section

This cross-section is reasonably well known from 1 to 14 MeV, but since forward-scattered protons only are detected in the upper energy range, the angular distribution of the reaction is needed.

Recommendation

It is recommended that the absolute cross-section as a function of angle be measured at several energies above 8 MeV with the object of allowing the determination of the cross-section for forward-scattered protons to an accuracy of 0.5 to 1% at 14 MeV.

3. FISSION AND CAPTURE STANDARDS

3.1. The fast-fission cross-section of ^{235}U

Because of considerable structure as a function of energy, the usefulness of the ^{235}U fission cross-section as a standard below 100 keV is questionable. However, if this cross-section is to be used as a standard above 100 keV, the available data sets should be examined below this energy as well as above. Furthermore, cases of usefulness as a standard below 100 keV may occur. The data base in this region continues to improve, but between absolute values at thermal energies and absolute values above 20 keV, differences in the shape of the cross-section among various experiments leave uncertainties of the order of ± 5%.

Since the 1970 Argonne Symposium [9], new measurements and modifications of earlier results have contributed significantly to an improved knowledge of the fission cross-section. These new results include the following:

(a) New relative measurements have been made with ORELA [10] from 100 eV to 100 keV by Perez et al. [11] and from thermal energies to 100 keV by Gwin et al. [12]. Both measurements are normalized at low energies.

(b) Szabo et al. [13] have modified previous absolute measurements made with a Van de Graaff and have obtained new data above 1 MeV. Data based on White's fission chamber and reported previously at the 1970 Argonne Symposium [14] remain unchanged. Data reported at the 1971 Knoxville Conference [15] are currently being reassessed as a result of a foil recalibration.

(c) Relative measurements in the 1-keV to 1-MeV region have been made by Gayther et al. [16] using the Harwell linac. These data are normalized to the 1972 evaluation by Sowerby et al. [17] in the 10- to 30-keV range and have been reported previously only with preliminary flux measurements.

(d) New absolute data of Poenitz [18], obtained with a Van de Graaff, replace the low preliminary values reported previously [19]. The new data cover the range from about 35 keV to 3.5 MeV and are subject to final confirmation of the foil assay.

(e) Both absolute and relative data in the energy range 0.5 - 1.2 MeV have been obtained by Käppeler [20] using the Karlsruhe Van de Graaff.

(f) New but preliminary absolute data in the range 1 - 6 MeV obtained by Hansen et al. with the Los Alamos Van de Graaff were reported by Diven [21].

The new data presented at the Panel are plotted in Fig. 1. A comparison of the data reveals the following features:

(a) From 35 keV to 1 MeV where measurements partially overlap, the ORELA data, the data of Szabo et al. based on White's fission chamber, and the data of Poenitz and of Gayther et al. are in reasonable agreement when the errors of approximately 3 - 4% in the individual data sets are considered.

(b) Of the 19 data points of Käppeler, the five between 500 and 700 keV lie consistently about 6% higher than the data of Poenitz and of Gayther while the data of Szabo et al. have intermediate values.

(c) The data of Käppeler also appear to differ in shape from the other measurements in the 500-keV to 1-MeV region although both he and Poenitz have evidence for a 'step' in the cross-section at approximately 1 MeV.

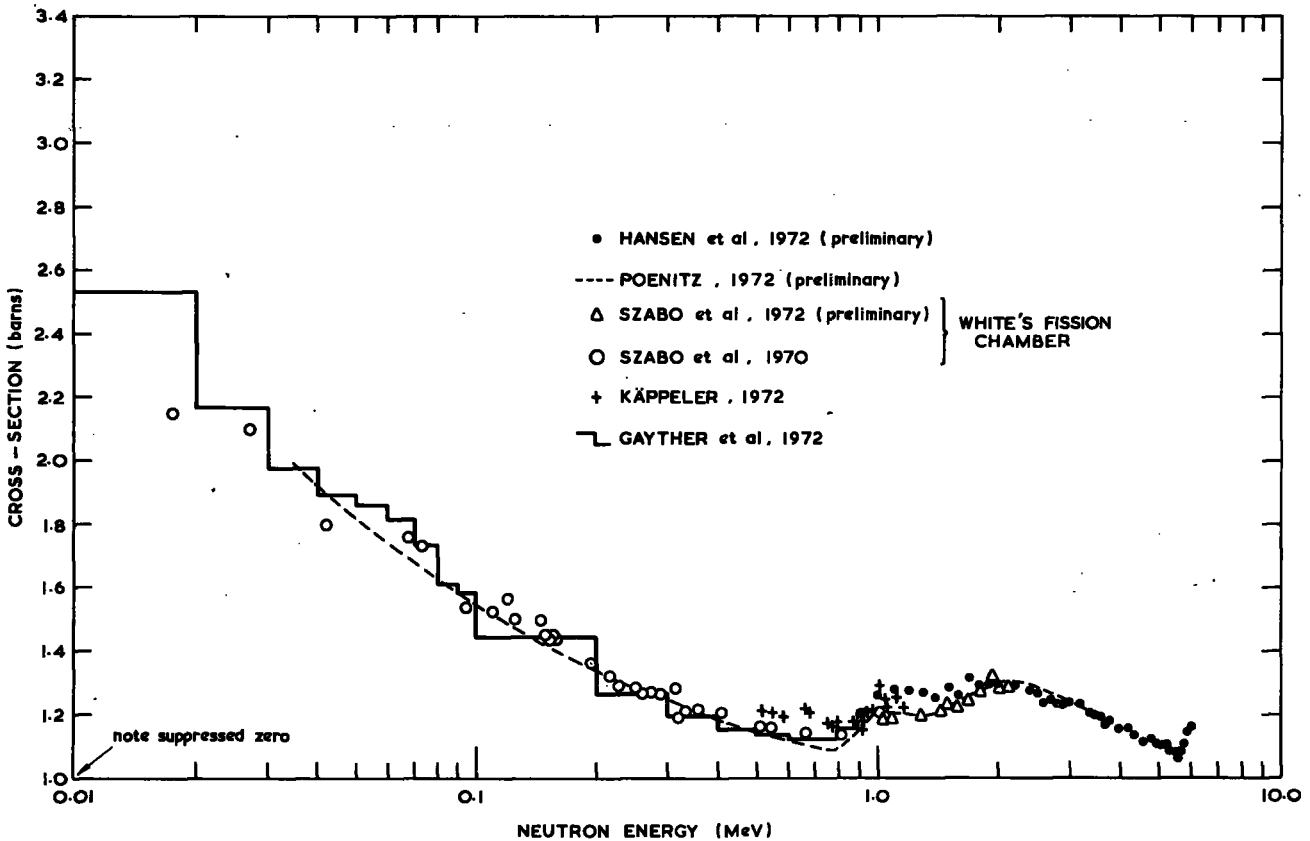

FIG. 1. Recent measurements of the ^{235}U fission cross-section.
(Figure prepared by M.S. COATES and D.B. GAYTHER.)

(d) From 1 to 2 MeV the data of Szabo and Poenitz agree while the data of Käppeler are somewhat higher, and the new Los Alamos data are approximately 6% higher.

(e) From 2 to 3.5 MeV the Los Alamos data and those of Poenitz agree.

Recommendations and observations

(a) In the last two years, overall knowledge of the $^{235}U(n,f)$ cross-section has improved considerably. Below 500 keV the new data appear to agree sufficiently well so that an evaluation would be fruitful.

Above 500 keV there appear to be differences of up to 6% among the new data. In the important energy range 0.5 to 2 MeV it remains somewhat difficult to determine the shape of the cross-section, and a more detailed examination of this range is suggested. Although an evaluation would best await a final assay of the foils used by Poenitz and the Los Alamos group, even now an evaluation might achieve an estimated accuracy of about ± 3%.

(b) Turning attention to lower energies, standard fission cross-section integrals should be established and evaluated periodically as suggested by Deruytter in 1971 [22] in order to supplement the standard thermal fission cross-section of ^{235}U (as well as the thermal cross-sections of all other major fissile isotopes).

(c) For those cases in which the $^{235}U(n,f)$ cross-section must be used as a standard below 50 - 100 keV, it will be important for the experimenter to determine what additional uncertainties must be assigned to his experiment because of fluctuations in the standard cross-section.

(d) For accurate measurement of the standard fission cross-section of ^{235}U it is necessary to have detailed information concerning the angular distribution of fission fragments over the whole range of energies and angles of interest.

3.2. Fast-neutron capture cross-section of ^{197}Au and other appropriate capture standards

The most important advantage in the use of gold as a capture standard is the high accuracy with which the capture rate can be determined from the induced activity of ^{198}Au. This outweighs the disadvantage of the large proportion of gamma transitions to low-lying levels, which may disturb the detection of prompt capture gamma rays (from other materials) with split-tank scintillators. However, such detectors are used at present in only a small number of laboratories.

New absolute measurements of the capture cross-section of ^{197}Au by Fort et al. [23] support previous values reviewed at the 1970 Argonne Symposium and also agree well with the evaluation presented by Poenitz [24]. Preliminary measurements by Fort et al. [23] using the activation technique reveal some discrepancies of the order of 15% which should be resolved.

Based on the independent absolute measurements, the present uncertainty of the gold capture cross-section below 500 keV is less than ± 5%, probably ± 3%. Discrepancy still exists between the absolute cross-section data and values obtained relative to the $^{235}U(n,f)$ cross-section. Similar discrepancy with absolute measurements is also apparent for gold capture data based on the ^{238}U capture cross-section. Consequences of fluctuations, which are present in the lower keV energy range, must be considered in individual experiments when gold is used as a standard.

Recommendations

(a) Gold should be retained as the capture cross-section standard. It would be a useful check to include gold in all measurements of capture cross-sections even if a different standard is used for flux measurements.

(b) The assumption of spectral independence of prompt gamma-ray detectors is a possible source of uncertainty in capture cross-section measurements. It is proposed that, in addition to the capture cross-section of gold, the ratio of gold-to-indium capture cross-sections be reliably established since the capture gamma-ray spectra from these two elements are significantly different.[1]

3.3. The value of $\bar{\nu}$ for spontaneous fission of ^{252}Cf[2]

Recommendations

(a) Based on a least-squares fit of all known absolute measurements of $\bar{\nu}$ for spontaneous fission of ^{252}Cf, the following value was derived:

$$\bar{\nu}_{total} = 3.733 \pm 0.0083 \text{ internal error}$$

$$\pm 0.0078 \text{ external error}$$

It is recommended that this value be used for normalization of future $\bar{\nu}$-measurements relative to ^{252}Cf.

Input data for the least-squares programme included the recent measurements of Boldeman [26] and all absolute measurements discussed in the reviews by Axton [27] and by Condé [28]. Weighting of the various data according to estimates of experimental error was based upon published literature cited in the review papers, upon discussions during the Panel meeting, upon preliminary correspondence with many other authors and upon

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[1] Editor's note: In a review of the Summaries, Conclusions and Recommendations, some participants challenged the technical basis for proposing to measure or establish the ratio of the gold-to-indium cross-sections. It is the opinion of the secretary that the panel participants realized that measurements of the ratio would not resolve the primary problem of insufficient understanding of the spectral response of gamma-ray detectors but thought that reliably established ratios might help to illuminate detector problems in a user's laboratory.

In a preliminary version of this paper, owing to a typographical error, 'iodine' was written instead of 'indium'. Several interesting points arose in the ensuing correspondence.

(a) Iodine would also be useful as a secondary standard under certain circumstances. For example, iodine has a smaller thermal capture cross-section than gold and would be useful when there is a large thermal background.

(b) There are practical difficulties [25] in the use of gold for activation measurements (as opposed to capture gamma-ray measurements). If indium were to be used as a standard in place of gold in order to avoid these difficulties, then absolute values of the indium cross-section would be required — not ratios to gold.

[2] Editor's note: Because of limited time, the recommendations in sections 3.3, 3.4 and 3.5 could not be discussed in complete detail by the full panel so that the contents of these sections may represent the consensus of the working group rather than of the full panel. Some of the recommended numerical values are controversial and might have been somewhat different if they had been discussed by the full panel or by a different group of scientists. Hopefully, the implementation of the recommendations will contribute to the resolution of the controversies.

the proceedings of the Consultants' Meeting on the Third Evaluation of the Thermal Fission Constants (Vienna, 15 - 17 November 1972) [29], in which several members of the present Panel had participated. Values of $\bar{\nu}$ derived from measurements of the parameter η were excluded from the present analysis.

(b) From previous discussions at the Consultants' Meeting [29], it appeared that inclusion of all data, both absolute and indirect, in the least-squares fit produced a value 0.5% higher than that recommended above and that exclusion of all absolute measurements (leaving essentially only values derived from η-measurements) produced a value approximately 1.5% higher than that recommended above.

The discrepancies among the results of the various fitting procedures can apparently be traced to the assignment of high weights to the η-experiments. Therefore, the following is recommended:

(i) either the corrections and author-estimated errors of the η-measurements should be reassessed
(ii) or η should be re-measured.

The Panel was not convinced that either of these recommendations would yield values of $\bar{\nu}$ with errors as small as those of the direct measurements.

The Panel noted that the errors assigned to the value recommended above describe only the uncertainties in the direct values and do not reflect the discrepancy of approximately 1.5% between the directly and indirectly determined values. The problem of the value of $\bar{\nu}$ for ^{252}Cf is therefore not yet satisfactorily resolved in spite of the improved agreement among the direct measurements.

3.4. The 2200-m/s fission and capture cross-sections of the fissile nuclides[3]

Recommendations

(a) Because of difficulties involved in deducing 2200-m/s values from effective cross-section measurements, it is recommended that the quantities required be measured at 2200 m/s neutron velocity.

(b) Extensive work on the alpha half-life of ^{234}U has been performed at CBNM using several counting techniques (low geometry, medium geometry and 4π alpha counting; liquid scintillation counting) and several different mass determinations (controlled-potential coulometry, isotopic dilution, weighing in ultra-high vacuum). While this work is being finalized and confirmed at Argonne National Laboratory and at Chalk River, the Panel received the working group's recommendation of the value

$$T_{1/2}(^{234}U) = 2.446 \times 10^5 \text{ a} \pm 0.3\%$$

for calculation of the amounts of ^{235}U in targets used in fission cross-section measurements. The quoted error represents three standard deviations.

(c) The difference among measured values of the alpha half-life of ^{233}U is striking. Several values from high-precision alpha counting are grouped around $T_{1/2}(^{233}U) = 1.61 \times 10^5$ a. However, recent measurements by

[3] See Footnote 2.

Keith [30] using alpha counting and by Oetting [31] using a calorimetric method both yield the value 1.55×10^5 a. The difference between the two groups of measurements is about 4%. A recent but unpublished measurement performed at Chalk River by Durham et al. using the alpha-counting method yields a value of 1.583×10^5 a.

High-accuracy measurements of the half-life of ^{233}U are recommended to resolve the discrepancy.

(d) For the half-life of ^{239}Pu the counting method yields with high consistency a value of 24 395 ± 29 a (error = 3 standard deviations) [32] while a recent calorimetric measurement by Oetting yields a value of 24 065 ± 50 a [33]. This difference of 1.3% is directly reflected in the 2200-m/s value of the fission cross-section of ^{239}Pu. Measurements of high accuracy are needed to resolve the discrepancy.

(e) There is a persistent difference among values of 2200-m/s fission cross-section ratios deduced from g-dependent experiments. For example, values of the $\sigma_f(^{239}Pu)/\sigma_f(^{235}U)$ ratio measured at Aldermaston in a thermal column are 1.277 ± 0.025 (White et al. [34]), 1.271 ± 0.015 (Keith et al. [35]); the ratio obtained with a beam extracted from the column is 1.235 ± 0.022 (White et al. [34]). Values measured at Chalk River are: 1.2970 ± 0.0075 (Bigham et al. [36]), 1.2926 ± 0.0081 (Lounsbury et al. [37]). The ratio is rather sensitive to the assumed g-factors, and results are different for different irradiation facilities. Ratio values obtained with monokinetic neutrons of 2200-m/s velocity are much more consistent.

It is therefore recommended that evaluators reassess the g-factors for the fissile isotopes to take into account recent low-energy data. In view of the inconsistency of ratio values obtained with different neutron spectra, it is recommended that the errors assigned to the Maxwellian ratios be increased to a realistic value when those ratios are used in an evaluation of all parameters at 2200 m/s.

(f) For ^{241}Pu the available information is very scarce. Further measurements of the 2200-m/s fission and absorption cross-sections are needed in connection with the least-squares evaluation by the IAEA [29].

(g) When 2200-m/s fission cross-section measurements are performed, they should be carried out in low geometry in order to avoid significant corrections for scattering and self-absorption of fission fragments in the fissile layers.

(h) It is recommended that the output parameters from the IAEA revision [29] of the 2200-m/s parameters should be used for future normalizations of relative measurements.

3.5. Fission neutron spectra[4]

Recommendations

(a) It is recommended that a standard fission neutron spectrum be included among the neutron standard reference data. The spectrum of neutrons from spontaneous fission of ^{252}Cf should be a primary standard but the fission spectrum of ^{235}U induced by neutrons below 150 keV should be regarded as an associated standard.

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[4] See Footnote 2.

The choice of ^{252}Cf is derived from its wide potential utility, not only for comparison with other fission neutron spectra but also in simple integral experiments, detector calibration, etc. [38]. ^{235}U is included because of its convenience as a standard for comparison in many experimental applications involving accelerators or reactors and because at present it is comparatively well known. Over the energy range 0.5 - 7 MeV there is good consensus, and there is already at least one set of very careful measurements [39] extending to 15 MeV.

(b) While appreciating the value for discussions and scientific analysis of representations of the fission spectrum in terms of parametrized analytical formulae, the Panel insist that the definitive representation is in numerical terms, i.e. a table of intensity as a function of energy. This table should include a statement of the statistical and systematic errors.

(c) The correction of standard measurements for the effects of multiple reactions requires data for the elastic and inelastic cross-sections of all materials of importance in the experiment, e.g. ^{235}U, platinum and the components of stainless steel. It is suggested that the effect of present uncertainties in these cross-sections on the accuracy of the fission neutron spectra should be assessed.

(d) The possible existence of delayed neutron groups with half-lives less than those currently accepted makes difficult a clear definition of prompt and delayed fission neutron spectra and hence could in principle result in observation of different spectra depending on the technique or parameters used. It is recommended that attention be given to establishing the existence and intensity of such neutron groups and their associated spectra.

(e) The present state of our knowledge of the fission neutron spectrum of ^{252}Cf is regarded as far from satisfactory. The experiments are mostly rather old, and many of them require substantial corrections, as was pointed out at the Consultants' Meeting on Prompt Fission Neutron Spectra [40]. Less attention was paid to detector and energy calibration than now seems desirable. It is recommended that the results of experiments currently in progress be awaited before an evaluation of the ^{252}Cf fission neutron spectrum is attempted.

(f) It is felt that an evaluation of the ^{235}U fission spectrum could profitably be carried out as soon as all the results currently known in preliminary form have been finalized.

4. GENERAL RECOMMENDATIONS

(1) Particularly in the field of neutron standard reference data, experimenters are strongly urged to make special effort to include in the publication of their work all details required for evaluation and permanent documentation.

The reporting of sources of error and estimates of uncertainty is frequently in need of improvement. Although similar recommendations have appeared in the proceedings of numerous meetings, further improvement is necessary, and therefore the following recommendations on reporting of errors are made. Recommendations (2a) through (2c) are based on similar recommendations of the Consultants' Meeting on the Third Evaluation of the Thermal Fission Constants [29] which the Panel strongly affirm.

(2a) In general, greater care should be devoted to the identification of sources of uncertainties.

(2b) Overall random and systematic errors should be reported separately, whether or not the combined uncertainty is also reported.

(2c) Component uncertainties in each of the above categories should be itemized, especially the systematic errors.

(2d) Experimenters should report precisely how components of the uncertainty correlate among various subsets of the data. For example, the full estimated variance (or relative variance) on a value might be given as a sum of terms, one of which is not shared by any other value, one of which might depend on a foil assay and be shared equally among all values given, and perhaps one of which is shared among a subset of points with correlated background or flux determination uncertainties. Such an organization of uncertainties would allow the construction, if necessary, of the full covariance matrix of the data given.

This information is needed even to determine correctly whether various sets of experimental values are discrepant, and it is further needed to allow a rational combination of results from various investigators. As pointed out by Usachev et al. [41], such detailed estimates of uncertainties are needed for final evaluated cross-sections to allow the assessment of the effects of uncertainties upon applications such as reactor design.

Evaluators cannot supply the required details about uncertainties unless measurers provide the basic information in a clear way. Of course, much relevant information is often supplied by experimentalists, but the manner of presentation is sometimes ambiguous.

When the published account of research lacks sufficiently detailed information about uncertainties, often an evaluator's only option is to revise the uncertainties to a 'realistic' value. Regrettably, such revision may in effect exclude the measurement from the evaluation.

(2e) Journals are urged to accept for publication the detailed information required for subsequent evaluation and documentation, especially in the case of neutron standard reference data. Many Panel members had personally encountered rejection of important information by journals on the basis of excessive detail.

(3) The Panel recognized the important role of the Central Bureau for Nuclear Measurements (CBNM) in providing targets and samples relevant to neutron standard reference data measurements, both inside and outside the European communities. They expressed hope that the same service will continue to be available in the future.

(4) The comparison of neutron flux measurements which has been undertaken by the International Bureau of Weights and Measures (BIPM) is a valuable approach which should be supported.

(5) The Euratom Working Group on Reactor Dosimetry (EWGRD) has indicated the need for rapid improvement of certain capture and threshold reaction cross-sections relevant to reactor dosimetry. In view of present concentration on only six primary standard reactions, three of which also belong to the reactor-dosimetry class, EWGRD should consider whether the adoption of the neptunium fission cross-section as a primary standard would improve the situation. This suggestion should also be brought to the attention of the International Working Group on Reactor Radiation Measurements (IWGRRM) of the IAEA.

(6) The Panel strongly urges all participants to send their data to the Neutron Data Centre which serves their area.

(7) In consideration of the extensive efforts presently under way at many laboratories, a third panel on neutron standard reference data is recommended for spring 1975.

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CONVERSION TABLE:
FACTORS FOR CONVERTING UNITS TO SI SYSTEM EQUIVALENTS*

SI base units are the metre (m), kilogram (kg), second (s), ampere (A), kelvin (K), candela (cd) and mole (mol). [For further information, see International Standards ISO 1000 (1973), and ISO 31/0 (1974) and its several parts]

Multiply		*by*	*to obtain*
Mass			
pound mass (avoirdupois)	1 lbm	$= 4.536 \times 10^{-1}$	kg
ounce mass (avoirdupois)	1 ozm	$= 2.835 \times 10^{1}$	g
ton (long) (= 2240 lbm)	1 ton	$= 1.016 \times 10^{3}$	kg
ton (short) (= 2000 lbm)	1 short ton	$= 9.072 \times 10^{2}$	kg
tonne (= metric ton)	1 t	$= 1.00 \times 10^{3}$	kg
Length			
statute mile	1 mile	$= 1.609 \times 10^{0}$	km
yard	1 yd	$= 9.144 \times 10^{-1}$	m
foot	1 ft	$= 3.048 \times 10^{-1}$	m
inch	1 in	$= 2.54 \times 10^{-2}$	m
mil (= 10^{-3} in)	1 mil	$= 2.54 \times 10^{-2}$	mm
Area			
hectare	1 ha	$= 1.00 \times 10^{4}$	m^2
(statute mile)^2	1 mile^2	$= 2.590 \times 10^{0}$	km^2
acre	1 acre	$= 4.047 \times 10^{3}$	m^2
yard^2	1 yd^2	$= 8.361 \times 10^{-1}$	m^2
foot^2	1 ft^2	$= 9.290 \times 10^{-2}$	m^2
inch^2	1 in^2	$= 6.452 \times 10^{2}$	mm^2
Volume			
yard^3	1 yd^3	$= 7.646 \times 10^{-1}$	m^3
foot^3	1 ft^3	$= 2.832 \times 10^{-2}$	m^3
inch^3	1 in^3	$= 1.639 \times 10^{4}$	mm^3
gallon (Brit. or Imp.)	1 gal (Brit)	$= 4.546 \times 10^{-3}$	m^3
gallon (US liquid)	1 gal (US)	$= 3.785 \times 10^{-3}$	m^3
litre	1 l	$= 1.00 \times 10^{-3}$	m^3
Force			
dyne	1 dyn	$= 1.00 \times 10^{-5}$	N
kilogram force	1 kgf	$= 9.807 \times 10^{0}$	N
poundal	1 pdl	$= 1.383 \times 10^{-1}$	N
pound force (avoirdupois)	1 lbf	$= 4.448 \times 10^{0}$	N
ounce force (avoirdupois)	1 ozf	$= 2.780 \times 10^{-1}$	N
Power			
British thermal unit/second	1 Btu/s	$= 1.054 \times 10^{3}$	W
calorie/second	1 cal/s	$= 4.184 \times 10^{0}$	W
foot-pound force/second	1 ft·lbf/s	$= 1.356 \times 10^{0}$	W
horsepower (electric)	1 hp	$= 7.46 \times 10^{2}$	W
horsepower (metric) (= ps)	1 ps	$= 7.355 \times 10^{2}$	W
horsepower (550 ft·lbf/s)	1 hp	$= 7.457 \times 10^{2}$	W

* Factors are given exactly or to a maximum of 4 significant figures

Multiply		*by*	*to obtain*
Density			
pound mass/inch^3	1 lbm/in^3	= 2.768×10^4	kg/m^3
pound mass/foot^3	1 lbm/ft^3	= 1.602×10^1	kg/m^3
Energy			
British thermal unit	1 Btu	= 1.054×10^3	J
calorie	1 cal	= 4.184×10^0	J
electron-volt	1 eV	≃ 1.602×10^{-19}	J
erg	1 erg	= 1.00×10^{-7}	J
foot-pound force	1 ft·lbf	= 1.356×10^0	J
kilowatt-hour	1 kW·h	= 3.60×10^6	J
Pressure			
newtons/metre^2	1 N/m^2	= 1.00	Pa
atmosphere[a]	1 atm	= 1.013×10^5	Pa
bar	1 bar	= 1.00×10^5	Pa
centimetres of mercury (0°C)	1 cmHg	= 1.333×10^3	Pa
dyne/centimetre^2	1 dyn/cm^2	= 1.00×10^{-1}	Pa
feet of water (4°C)	1 ftH_2O	= 2.989×10^3	Pa
inches of mercury (0°C)	1 inHg	= 3.386×10^3	Pa
inches of water (4°C)	1 inH_2O	= 2.491×10^2	Pa
kilogram force/centimetre^2	1 kgf/cm^2	= 9.807×10^4	Pa
pound force/foot^2	1 lbf/ft^2	= 4.788×10^1	Pa
pound force/inch^2 (= psi)[b]	1 lbf/in^2	= 6.895×10^3	Pa
torr (0°C) (= mmHg)	1 torr	= 1.333×10^2	Pa
Velocity, acceleration			
inch/second	1 in/s	= 2.54×10^1	mm/s
foot/second (≈ fps)	1 ft/s	= 3.048×10^{-1}	m/s
foot/minute	1 ft/min	= 5.08×10^{-3}	m/s
mile/hour (= mph)	1 mile/h	= 4.470×10^{-1} m/s; 1.609×10^0 km/h	
knot	1 knot	= 1.852×10^0	km/h
free fall, standard (= g)		= 9.807×10^0	m/s^2
foot/second^2	1 ft/s^2	= 3.048×10^{-1}	m/s^2
Temperature, thermal conductivity, energy/area·time			
Fahrenheit, degrees −32	°F − 32	$\frac{5}{9}$	°C
Rankine	°R	$\frac{5}{9}$	K
1 Btu·in/ft^2·s·°F		= 5.189×10^2	W/m·K
1 Btu/ft·s·°F		= 6.226×10^1	W/m·K
1 cal/cm·s·°C		= 4.184×10^2	W/m·K
1 Btu/ft^2·s		= 1.135×10^4	W/m^2
1 cal/cm^2·min		= 6.973×10^2	W/m^2
Miscellaneous			
foot^3/second	1 ft^3/s	= 2.832×10^{-2}	m^3/s
foot^3/minute	1 ft^3/min	= 4.719×10^{-4}	m^3/s
rad	rad	= 1.00×10^{-2}	J/kg
roentgen	R	= 2.580×10^{-4}	C/kg
curie	Ci	= 3.70×10^{10}	disintegration/s

[a] atm abs: atmospheres absolute; atm (g): atmospheres gauge.

[b] lbf/in^2 (g) (= psig): gauge pressure; lbf/in^2 abs (= psia): absolute pressure.

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