EANDC(US)-139 "L" INDC(USA)-17 "L"

INDC-361

## REMARKS ON THE NEUTRON-INDUCED FISSION SPECTRUM\*

C. R. Lubitz Knolls Atomic Power Laboratory, Schenectady, New York

and

L. Stewart University of California, Los Alamos Scientific Laboratory Los Alamos, New Mexico

#### I. INTRODUCTION

The purpose of this note is to discuss experimental results pertaining to the shape of the <sup>235</sup>U neutron-induced fission spectrum with particular reference to the controversy surrounding activation measurements. A review of all pertiment data is beyond the scope of this paper. Instead, representative experiments have been chosen and areas which require further study are pointed out. It is believed that the results cited give a reasonable view of the present situation.

For the benefit of those not familiar with the topic under discussion, it can be summarized by stating that two groups of experiments have yielded different shapes for the fission spectrum of <sup>235</sup>U. The first group concerns microscopic or energy-differential spectral functions,  $\varphi(E)$ , which are determined directly usually relative to the hydrogen scattering cross section. Macroscopic or integral quantities of the type,  $\int \sigma(E) \ \varphi(E) \ dE$ , are the concern of the second group, where  $\varphi(E)$  is an indirect measurement which must be unfolded from the experimental data by rather complex techniques. In contrast to the conceptual simplicity of the direct measurements, these techniques contribute in a complicated way to ambiguities in the final result. A discussion of these problems will be found in Section III; here we will simply state our conclusions:

Over a long period of time, experimentalists at various laboratories using different techniques have shown remarkable convergence in measuring about 1.9 or 2 MeV as the mean energy of the <sup>235</sup>U fission spectrum. Furthermore, individual shape measurements are in rather good agreement when suitably normalized and fitted to either a simple Maxwellian,

 $\varphi(E) \sim E^{1/2} \exp(-E/T),$ 

or to the Watt form,<sup>1</sup>

 $\sigma(E) \sim \sinh(AE)^{1/2} \exp(-E/B).$ 

On the other hand, the activation analyses, which give a mean energy of  $\sim 2.2$  MeV, are subject to so many errors, the worst probably being in the detector cross sections themselves, that they simply cannot at the present time pinpoint the fission spectrum as being solely responsible for the

Work supported by the U. S. Atomic Energy Commission.



000388

discrepancies they have highlighted.\*

#### II. MICROSCOPIC MEASUREMENTS OF THE FISSION SPECTRUM

The experiments included in this section are characterized by an attempt to isolate the fission source from perturbing influences and to measure the neutrons directly, by time-of-flight or other methods. As such, they represent the most definitive group of experiments bearing on the fission spectral shape.

### A. Early Measurements

Three of the earliest measurements by Watt,<sup>1</sup> Bonner et al.,<sup>3</sup> and Hill<sup>4</sup> were reasonably well fit to the Watt spectrum over the energy range from 50 keV to 17 MeV. Although the experimental results exhibited fairly large error bars, these different detection techniques, cloud chamber and proton recoil counting, used in conjunction with fission plates, gave concordant results, namely

$$\varphi(E) = 0.484 e^{-E} \sinh \sqrt{2E},$$

with a mean energy of 2.0 MeV.

#### B. Cranberg and Nereson and Frye and Rosen Experiments

Using 5-80-keV neutrons from the <sup>7</sup>Li(p,n) reaction and TOF techniques, Cranberg and Neregon<sup>5</sup> measured the <sup>235</sup>U fission spectrum from about 160 keV to <sup>4</sup> MeV. An independent observation by Frye and Rosen, with nuclear emulsions exposed to a fission plate bombarded by thermal neutrons, is reported in the same paper.<sup>5</sup> These results, when normalized to equal areas and fitted to the Maxwellian and Watt forms of the fission spectrum, give a Watt-type spectrum:

$$\varphi(E) \approx e^{-E/0.965} \sinh \sqrt{2.29 E},$$

while a Maxwellian distribution, <sup>†</sup> that is,

$$\varphi(E) \approx \sqrt{E} e^{-E/T},$$

with T = 1.29 MeV, also fitted their data quite well.

\_1 \_1

This situation requires a solution and a first step would be to improve the accuracy of activation cross sections and to eliminate any sources of error in foil counting techniques. Another possibility is that the fission cross sections are in error; for example, 235U fission could be too high at low energies and too low at high energies. Although this suggestion is speculative, such errors in the fission cross sections would produce results quite similar to those presently interpreted in terms of a hardened fission spectrum.

<sup>†</sup>In the Maxwellian, the mean energy is 1.5 T; the most probable energy is T/2; T = 1.29 corresponds to  $\overline{E}$  = 1.935 MeV.

<sup>\*</sup>The above-mentioned difference in spectral shape is large enough to have an important bearing on reactor design and calculations have shown that it produces results at variance with many accepted measurements. Since foil activation techniques are widely used for determining power levels and flux shapes, the authors of the macroscopic papers, in particular McElroy,<sup>2</sup> are rightly concerned that many activation experiments do not reproduce the long-accepted fission spectral shape.

C. Recent Time-of-Flight Results

1. Using the <sup>7</sup>Li(p,n) reaction and time-of-flight techniques, Condé and During<sup>6</sup> measured the spectra from 235U, 239Pu, and 252Cf and fitted their results to Maxwellian distribution functions. From their work on 235U at 40 keV and 1.5 MeV they report  $T = 1.24 \pm 0.04$  and  $1.25 \pm 0.04$  MeV, respectively.

2. Barnard et al.<sup>7</sup> (Harwell) performed TOF experiments for 100-keV neutrons incident upon  $^{235}$ U and detected the neutrons from 300 keV to <sup>4</sup> MeV. These data are compared in Fig. 1 with the TOF and nuclear emulsion data of Cranberg et al.<sup>5</sup> A fit to the Harwell data gave T = 1.297 MeV while the smooth curve in Fig. 1 has been calculated with T = 1.29 MeV. It is clear that these three independent measurements (by Cranberg and Nereson, Frye and Rosen, and Barnard et al.) show remarkable agreement with each other and with the Maxwellian distribution. Barnard et al. also compiled results on <sup>235</sup>U and other fissionable isotopes and report the value, T = 1.30 ± 0.01 MeV, obtained from an average of eight independent measurements on <sup>235</sup>U for incident energies  $\leq 100$  keV.

Other measurements of this type could be quoted. Since they are of lower accuracy and almost always agree within the stated errors with a T of 1.29-1.30 MeV, they do not add to the averall picture.

## III. THE GRUNDL-MCELROY-FABRY SPECTRAL ANALYSES

Papers by these authors are usually quoted in connection with the fission spectrum controversy; therefore their analyses of activation results obtained with a thermal fission spectrum are briefly summarized:

A. Grundl<sup>8</sup> measured spectral indices\* for various detectors with <sup>233</sup>U, <sup>235</sup>U, and <sup>239</sup>Pu sources in a cavity with source and detector surrounded by D<sub>2</sub>O and mounted above the "Hydro core reactor." The source neutrons were assumed to be thermal. A comparison with photoplate data from Grundl's Table X is reproduced below after renormalizing both sets of data to give an integral flux of 100%.

Energy Groups <u>MeV</u>	Grundl's Activation Flux in Percent	TOF and Photoplate Flux in_Percent	Percent Difference OF
0 - 1.4	37.31	45.98	-18.9
3 - 6	23.99	18.50 2.45	+ 9•5 +29•7 + 4 4
> <u>11</u>	0.0688	. 0.0499	+37.9

The striking features are the 20% "depletion" below 1.4 MeV and the 30% "bulge" from 3-6 MeV.

B. From an extensive study of activation experiments, McElroy<sup>2</sup> concluded that the average energy of the <sup>235</sup>U fission spectrum should be about 200

<sup>\*</sup>A term denoting ratios of detector cross sections, or ratios of ratios, usually measured in a known (or unknown) spectrum but sometimes with monoenergetic neutrons.

to 300 keV higher than the microscopic value, that is,  $\overline{E} \sim 2.2$  MeV; he also concurs with Grundl's flux shape tabulated above. While a Maxwellian does not fit this shape at all, the magnitude of the discrepancy with microscopic measurements can be qualitatively illustrated by "converting"  $\overline{E} = 2.2$  MeV to T = 1.47 MeV. This is 17 standard deviations outside the average value,  $T = 1.30 \pm 0.01$  MeV, quoted by Barnard et al.<sup>7</sup>

C. Fabry and DeCoster<sup>9</sup> also performed a cavity-type experiment using a  $^{235}$ U fission source. They quoted the following absolute integral cross sections based on the  $^{115}In(n,n'\gamma)$  activity:

$$\overline{\sigma}_{235} = 1335 \pm 130$$
 mb,

and

$$\overline{\sigma}_{238} = 353 \pm 30 \text{ mb},$$

or a 235/238 ratio of 3.78 in excellent agreement with Grundl's determination of the same quantity (3.85). The integral cross section Fabry and Decoster found for  $^{238}$ U, however, is higher than the Leachman-Schmitt value by 13.5 percent (see Section IV.A). These authors suggest that their measurement supports higher fission cross sections than those evaluated by Davey, <sup>10</sup> when integrated over the Grundl spectrum.

## D. Comments on These Analyses

In order to deduce a shape for the spectrum irradiating their samples, the above authors must perform certain steps which will be enumerated here together with difficulties possibly encountered along the way:

### 1. Measurement of the foil activities

Fission-spectrum-averaged activation cross sections reported in the literature often show fairly large discrepancies, commonly 20-25% and sometimes more. This implies that the underlying activity measurements are inherently difficult.

### 2. Choice of activation cross sections

An essential part of the activation method is to calculate the integrals weighted with some trial spectrum. For this purpose, precise point-wise cross sections are required. Despite considerable activity in recent years, the level of accuracy achieved is not high enough to preclude 10-15% errors in the "best sets," whereas not more than a few percent can be tolerated in accurate flux unfolding.

#### 3. Fast flux spectral calculations

Because of scattering and absorption, the flux irradiating the foils usually differs from a virgin fission spectrum. To deduce a shape for the latter, one has to work backward in principle from the observed unfolded spectrum; such calculations are also beset by difficulties. A comparison of independent shielding calculations<sup>11</sup> and, more recently, the work of the Cross Section Evaluation Working Group's Data Testing Subcommittee<sup>12</sup> have shown that independent calculations carried out at different laboratories often produce serious discrepancies. These can be resolved but only by extensive intergroup cooperation.

The above difficulties imply that activation measurements cannot yet be considered definitive in assessing the fission spectral shape. In fact, some activation measurements do not agree with this hardened <sup>235</sup>U fission spectrum derived from macroscopic experiments.

E. In particular, Bresesti et al.<sup>13</sup> constructed a set of intercalibrated activation cross sections for use in fast flux analysis from measurements with thermal neutrons. The <sup>235</sup>U fission plate and threshold detectors were placed in a cavity and the resultant spectral distortion corrections calculated by Monte Corlo. Using these pointwise evaluated cross sections and the Watt spectrum, they show a standard deviation of about 3 percent between their calculated and measured spectrum-weighted-average ratios. This implies that nothing is seriously wrong with the microscopic data on the <sup>235</sup>U fission spectrum.

These authors state that the 3-6 MeV "bulge" in the Grundl spectrum is inconsistent with their  $5^{4}$ Fe(n,p) and  $5^{8}$ Ni(n,p) cross sections which they believe to be better known in this energy region than the Al(n,p) and P(n,p) used by Grundl. Rydin<sup>14</sup> also feels that the short half life of Al along with inconsistencies which could be introduced between  $\beta$ -counting and  $\gamma$ -counting could be troublesome. In order to avoid the latter difficulty, Bresesti et al. employed  $\gamma$ -counting exclusively in their measurements.

F. As further evidence against the harder fission spectrum, Staub and Swaja<sup>15</sup> recently reported on calculations made at Bettis using various fission spectra. The Grundl-McElroy-Fabry spectrum gave results for the age in zirconium-water mixtures, ratios of 238U/235U fission, \* and eigenvalues of some Oak Ridge homogeneous spheres which were about 10%, 18%, and 1.5% different from experiment, respectively. The conventional spectra were much closer, generally within the accuracy of the comparisons, which were about 1%, 2.5%, and 0.25%, respectively. Similar results have been obtained at AI and ANL.

#### IV. PERTINENT FISSION CROSS SECTION AND RATIO MEASUREMENTS

#### A. The Leachman-Schmitt Integral Experiment

The serious discrepancy which exists between macroscopic experiments and the microscopic energy-dependent <sup>238</sup>U fission cross sections integrated over the <sup>235</sup>U fission spectrum is of recent origin. Since microscopic fission cross sections commonly employed through the early 1960's gave results in excellent agreement with integral measurements, little note was made of the following macroscopic experiment which highlights this discrepancy:

Leachman and Schmitt<sup>16</sup> measured  $312 \pm 5 \text{ mb}^{\dagger}$  for the 2380 fission cross section integrated over the 2350 thermal neutron-induced fission spectrum. This value was obtained in a  $2\pi$ -geometry with a thin coat of 2380 in a hemispherical shape and a 2350 fission source at the center. Scattering and

<sup>†</sup>Adjusted to  $\overline{v} = 2.42$ .

Ì

<sup>\*</sup>The data were basically from ENDF/B, version I, and these sets are currently under revision.

background corrections were applied even though the experiment was designed to minimize these effects. This cross section is about 11-13% higher than currently accepted  $^{238}$ U fission cross sections when integrated over the  $^{235}$ U fission spectrum (with T = 1.29 or 1.3 MeV); Davey's evaluation<sup>10</sup> of  $^{238}$ U fission based on most recent measurements, is typical of these. While the Leachman-Schmitt value could be in error, two groups\* at different laboratories have reported good agreement with this integral cross section.

## B. Fission Cross Sections

The activation analysts view the above discrepancy between integral and microscopic data as support for hardening the  $^{235}$ U fission spectrum. It should be pointed out, however, that the currently accepted  $^{235}$ U and  $^{238}$ U fission cross sections have undergone significant changes in the last few years and these changes have produced the above discrepancy. Experiments most prominent in lowering the  $^{235}$ U and  $^{238}$ U fission cross sections are summarized below:

1. In 1965, White<sup>17</sup> reported absolute <sup>235</sup>U fission cross sections which were several percent lower than an average of many independent measurements from a few keV to 15 MeV. Since <sup>235</sup>U is the "accepted" fission standard, the cross sections for other fissionable elements were consequently lowered in order to retain the absolute values of the fission ratios with respect to <sup>235</sup>U.

2. In 1968, Stein et al.<sup>18</sup> reported fission ratios from 1.5 to 5 MeV for 236U, 238U, and 237Np with respect to 235U with accuracies approaching 1% and found the 238U/235U ratio to be approximately 6% lower than commonly employed up to that time.

3. In 1969, Hansen et al.<sup>19</sup> reported scattering calculations had been made in order to correct old LASL proton telescope data. Again, <sup>235</sup>U and <sup>238</sup>U fission cross sections were decreased in addition to "lower" <sup>238</sup>U/<sup>235</sup>U ratios in the MeV range - in somewhat better agreement with the ratios of Stein et al. and the absolute fission cross sections of White.

4. Poenitz<sup>20</sup> also measured the  $^{235}$ U fission cross sections from 30 keV to 1.5 MeV. Except for the normalization point at 30 keV, his results are even lower than White's measurements and the differences, at some energies, are as large as 15-20%. The serious lack of agreement between these shape measurements and those of White could be due to the experimental geometry employed by Poenitz. First, he shielded a <sup>7</sup>Li(p,n) source from his monitor and TOF fission detectors. Both the monitor and fission counters were unshielded but, more importantly, hydrogen capture was chosen as the monitor reaction thereby precluding the use of TOF. The counts recorded by the monitor,

\*Complete descriptions of these experiments are not available, the results having been quoted only in secondary references. D. W. Allen and R. L. Henkel [Progr. Nucl. Energy, Series I, Phys. and Math. Vol. 2, p. 29, Pergamon Press (1958)] give  $307 \pm 7$  mb for the cross section measured by Richmond at Harwell; this value, however, should probably be corrected using recent values of  $\overline{\nu}$ . A Russian paper by M. N. Nikolaev, V. I. Golubev, and I. I. Bondarenko, published in Sov. Phys.--JETP 7, 517 (1958), contains the comment, "...  $310 \pm 10$  mb, a value previously obtained.", without further reference to or description of the experiment involved.

<sup>†</sup>Such low cross sections for <sup>235</sup>U fission make it difficult to calculate criticality for Lady Godiva, for example. For this reason, Poenitz' data have yet to be widely used and accepted for fast flux calculations.

therefore, are not easily related to the number of neutrons/cm<sup>2</sup> per unit solid angle passing the fission detector within a given time interval  $\Delta t$ . Perhaps this experiment should be repeated with an unshielded source and TOF employed on shielded fission and monitor counters.

## C. Fluctuations in Fission Cross Sections

While the experiments of White, Stein et al. and Hansen et al. have been instrumental in lowering the absolute fission cross sections for 2350 and 2380 in recent years, difficulties in interpreting fission data still remain. For example, 2350 fission has been assumed to be a smoothly varying function of energy up to 50-80 keV when necessary to obtain normalizations for higher energy data; the following experiments show this to be an incorrect assumption.

During the past year, independent experiments performed at Harwell,<sup>21</sup> LASL,<sup>22</sup> and LRL<sup>23</sup> show pronounced structure in <sup>235</sup>U fission up to the region of 50 keV. From 20-30 keV, energies often chosen for normalization purposes, LASL data indicate fluctuations as high as 15-20% and these are essentially borne out by both the LRL and Harwell measurements. Such structure necessitates significant changes in higher energy cross sections normalized to <sup>235</sup>U fission in the keV range. It is interesting to note that ascribing these fluctuations to genuine physical effects rather than to experimental errors follows closely the theoretical and experimental elucidation of intermediate structure in subthreshold fission. The fact that 20 percent fluctuations in the cross sections have been dismissed in the past as random reflects poorly on the often-heard statement that the <sup>235</sup>U fission cross section is known to a few percent.

## D. Point-wise Calibrations for Several Activation Cross Sections

Finally, another microscopic experiment which was undertaken in an effort to place activation cross sections on a firm foundational basis should be mentioned.

In 1967, Grundl<sup>24</sup> measured the cross sections for eight activation detectors from 1.7 to 14 MeV.\* Absolute cross sections were obtained by measuring ratios with respect to  $^{235}$ U or  $^{238}$ U fission foils whose cross sections were assumed to be knowr. As an example of the difficulties sometimes encountered with these techniques, consider the fact that Grundl measured a cross section for  $^{31}P(n,p)$  at one energy which differed by a factor of two from a measurement reported by Grundl et al. in 1958.<sup>25</sup> Both experiments, however, were performed using  $^{238}$ U fission as the monitor cross section even though different shapes and magnitudes for  $^{238}$ U fission were chosen for normalization in the two experiments. Suffice to conclude that the energy dependence of most activation cross sections is yet to be determined with reasonable precision, especially on an absolute basis.

<sup>\*</sup>The various detector thresholds, of course, prohibited measurements on all materials over the entire energy region.

## V. SPECTRAL MEASUREMENTS ON CRITICAL ASSEMBLIES AND REACTORS

## A. Microscopic Measurements

1. Of some interest are nuclear emulsion studies of the point-wise leakage spectra from several LASL bare critical assemblies. The data on  $^{2330}$  (Jezebel-23),  $^{26}$   $^{2350}$  (Lady Godiva),  $^{27}$  and  $^{239}$ Pu (Jezebel-49) $^{28}$  are displayed in Fig. 2 along with the average energies determined from these measurements. While the average energy of the Lady Godiva spectrum is markedly lower than for the  $^{2330}$  and  $^{239}$ Pu assemblies, all three assemblies show that the neutrons above ~2 MeV are in essential accord with a conventional Maxwellian distribution function. While some ambiguity exists in extrapolating to zero energy, the integral yields and therefore the normalizations chosen for presentation are more sensitive to this extrapolation than the average energies obtained from the measurements. However the comparisons are made, these data show no "Grundl-like" enhancement in the 3-6 MeV range.

Many measurements, performed on "Godiva-like" assemblies, are described in the literature. The following experiments are of special interest since the results have been interpreted in terms of a virgin fission spectrum and detailed comparisons made with other data.

2. Neill<sup>29</sup> (GGA) made TOF measurements on the APFA-III,  $^{235}$ U fueled, fast-critical assembly which is quite similar to Lady Godiva. He obtained an excellent Maxwellian fit to his data above 2 MeV with T = 1.318 MeV, in good agreement with the tail of a conventional fission spectrum.

## B. Macroscopic Measurements

McElroy et al.<sup>30</sup> made extensive studies of the same assembly, APFA-III, by placing activation detectors at several radial positions and deriving the spectra using various calculational procedures. The results of these measurements are summarized as follows:

First, McElroy et al. reported good agreement with Neill's integral fluxes above various reference energies from 10 keV to 6 MeV, which these authors report as "core center values." Second, Neill's results are converted to core surface and compared with surface activation measurements. While the methods employed by McElroy et al. in converting Neill's data are not completely understood, it should be noted that the "central" comparisons agreed to within 4-5% while the "corrected" surface results differ by as much as 20%, a value typically encountered in comparing "direct" versus "activation" measurements.

Third, McElroy et al. included comparisons of the leakage spectrum reported by Frye et al.<sup>27</sup> which were measured <sup>4</sup>2" from the Lady Godiva core center. Again he "converted" these measurements to a surface flux and found agreement to within a few percent with the "converted" Neill data, but, as noted above, both showed 20% discrepancies with the foil activation results.

Still larger discrepancies are observed when the foil results are unfolded and compared with a conventional fission spectrum. These authors present various average energies generally about 200 keV higher from foil activation results. The inability of these experiments to carefully define the low-energy end of the spectra, however, weakens these average-energy comparisons.

## C. Reactor Experiments

A host of experiments have been carried out in various reactor spectra; only two representative measurements will be included here.

1. Sherwood and King<sup>31</sup> determined the high-energy leakage spectrum from a 95% enriched 235U-core pool reactor using the proton-recoil technique. They found that a trial fission spectrum with a Maxwellian temperature of 1.3 MeV produced no significant deviations from the measured spectrum to within 1% in slope above 6 MeV.

2. Kimura et al.<sup>32</sup> measured the Kyoto University Reactor spectrum with seven threshold detectors and found activation cross sections in surprisingly good agreement with those of Bresesti et al. [with the possible exception of the  $^{56}$ Fe(n,p)], allowing for the arbitrary overall normalization of the latter. These authors conclude that their spectrum is fission-like, but presumably this means above a few MeV.

## VI. CONCLUSIONS AND RECOMMENDATIONS

Discrepancies between microscopic and macroscopic experiments were not apparent until both the 235U fission cross sections and the 238U/235U fission ratios were reduced considerably below those used 10 years ago. The excellent agreement among microscopic measurements of the fission spectrum, therefore, does not permit drastic changes to be made until possible major errors are removed in the absolute fission cross sections and fission ratios. The 235U fission spectrum averaged 235U/238U ratio, computed from point-wise cross sections, is approximately 4.61 in disagreement with Grundl's measured value of 3.85. Hardening the spectrum preferentially weights 235U and reduces the computed ratio. It is obvious, however, that raising the high-energy  $^{235U}$  fission cross section and, with it the  $^{238}U$ , has the same effect since the increase in 235U is partially offset by its large low-energy contribution to its spectrum-averaged value. In fact, the fluctuation effects now seen in 235U fission in the keV range could infer changes in the low-energy cross sections. The situation would be improved if these low-energy cross sections could be decreased but a significant increase would wipe out any agreement obtained by raising the high-energy cross sections. Suffice to remark that the current situation dictates that further study is required, quite possibly along the following lines:

## A. Activation Experiments

Activation measurements do not appear to have the precision required to pinpoint the fission spectrum as the cause of the observed discrepancies between microscopic and macroscopic data. The Grundl-McElroy-Fabry results are valuable, however, in bringing to a wide audience the discrepancy between differential and integral data in this particular area. This may serve to promote activity in producing less discordant microscopic and integral activation and fission cross sections. Experience in the resonance integral field, \* however, suggests that widespread improvement in agreement between microscopic and macroscopic approaches is not just around the corner.

Where, again, integral-differential discrepancies are the rule.

Since activation measurements continue to be used for intercalibrating fluxes in reactor spectra, every effort should be employed to determine absolute point-wise cross sections. By calibrating activation detectors against other reliable flux monitors and against fission cross sections measured with fission fragment detectors, perhaps more insight into the problems can be found. There is yet little reason to believe that all fission cross sections determined by activation are directly related to those determined from fragment counting since the energy dependence of the particular fragment producing the activation is not yet clearly defined in all cases.

### B. Point-wise Fission Cross Sections

There is every reason to believe that the energy dependence of the <sup>235</sup>U fission cross section is not well known, even though it is widely purported to be a "standard." Unfortunately, fission ratio measurements do not define absolute values of the cross sections. Precision absolute <sup>235</sup>U fission cross-section measurements over the range from a few keV to 20 MeV should be undertaken using modern methods including TOF techniques. If the energy dependence (shape) is well defined over the entire region for other fissionable isotopes, then ratio measurements would suffice as long as absolute values are verified at a few energy points. From 500 keV up, hydrogen scattering should be used to monitor the incoming flux.

## C. Fission Spectral Shape

As far as the shape of the fission spectrum is concerned, the good agreement among the various independent microscopic measurements implies that additional measurements of this type are not now required. In fact, experience shows that new measurements are as likely to confuse the situation as to clarify it. To achieve substantially higher precision would require an expenditure of time and money that is not easily justified.

## $D. \overline{v}$

1. Although a knowledge of  $\sigma_f(^{235}U)$  by itself is enough for many purposes (principally normalization), neutronics calculations require  $\overline{\nu} \sigma_f$ . It would be poor strategy to mount a 1% attack on  $\sigma_f$  and leave  $\overline{\nu}(E)$  in its current state.

2. Hanna et al.<sup>33</sup> have adopted a mean energy of 2.1 MeV for the <sup>235</sup>U fission spectrum for calculating leakage corrections to  $\overline{\nu}$  measurements. This choice is based on the Grundl-McElroy-Fabry papers. While this may be a small effect on their evaluated thermal cross sections, this point should be checked.

# E. Remeasurement of <sup>238</sup>U Fission Spectrum Averaged Cross Section

The  $^{238}$ U(n,f) measurement of Leachman and Schmitt should be repeated at one, and preferably two, other laboratories.

## F. Independent Check of McElroy's Calculation

It might be worthwhile to arrange for independent verification of McElroy's calculations at one or more laboratories. Certainly the technological importance of accurate fast flux measurements warrants considerable attention to the problems which he has described.

## VII. ACKNOWLEDGMENTS

H. Alter, W. G. Davey, J. A. Grundl, W. N. McElroy, J. Neill, T. A. Pitterle, and R. A. Rydin provided many helpful and pertinent discussions throughout the preparation of this paper. This summary report was undertaken at the suggestion of G. A. Kolstad, M. S. Moore, and R. F. Taschek. We wish to express our appreciation for their encouragement in addition to their technical contributions.

## APPENDIX

In any discussion of the shape of the <sup>235</sup>U thermal neutron-induced fission spectrum, questions regarding the dependence upon incident energy and the changes in the spectral shape with various important isotopes arise. Each of these subjects is briefly outlined in the following sections.

# I. INCIDENT-ENERGY DEPENDENCE OF THE <sup>235</sup>U FISSION SPECTRUM

If the incident neutron energy were an important parameter in determining the shape or mean energy of the fission spectrum, then serious experimental difficulties would be encountered in making precise measurements. For fast neutrons, the angular distributions of fission fragments show large anisotropies which vary with incident bombarding energy and isotope. The emitted neutrons are expected to be correlated in energy and angle with the emitted fragments, so that the spectrum measured at a few angles with respect to an incident beam would not necessarily provide correct results. Second, at high energies clean separation of the fission neutrons from those of competing reactions becomes a difficult task thus requiring coincidence measurements with the fission process itself. Such measurements carried out in the presence of fragment and/or neutron angular distribution effects would be a tremendous undertaking. Adding to this difficulty are elastic and, in some cases, inelastic angular distribution effects, since these neutrons must be removed from the emission spectrum without distorting the fission component. It has been shown by several authors that the average energy of fission neutrons should be correlated with  $\overline{v}$ , but most present high-energy measurements are not sufficiently precise to lend much insight into the problem of the energy dependence of the fission neutron spectrum. The relationship derived by Ferrell<sup>34</sup> between  $\overline{E}$  and  $\overline{v}$  seems to work reasonably well, especially if, as shown by Doyas and Howerton,  $35 \overline{v}$  is corrected to remove neutrons from competing, non-fission processes. Belov et al.<sup>36</sup> have recently claimed that  $\overline{v}$  in this relation should also be reduced by the number of neutrons emitted before scission.

#### II. FISSION SPECTRA OF OTHER ISOTOPES

The fission spectra for isotopes other than  $^{235}$ U are often required for reactor calculations. These data also lend insight into comparisons with similar measurements on  $^{235}$ U. The following summary is reproduced from the 1965 paper of Barnard et al.7 and the 1969 experiment by Belov et al.<sup>36</sup>

Isotope	Incident Neutron Energy	Average Maxwellian Temperature (MeV)	Number of Results Averaged	Reference
233 <sub>U</sub>	thermal	1.36 ± 0.02	9	7
235 <sub>U</sub>	thermal to 100 keV	1.30 ± 0.01	8	7
239 <sub>Pu</sub>	thermal to 130 keV	1.39 ± 0.01	8*	7
2 <sup>140</sup> Pu	spontaneous	1.19 ± 0.03	1	7
2 <sup>41</sup> Pu	thermal	1.34 ± 0.03	1	7
242 <sub>Pu</sub>	spontaneous	1.21 ± 0.07	1	36
2 <sup>1414</sup> Cm	spontaneous	1.37 ± 0.04	1	36
252 <sub>Cf</sub>	spontaneous	1.42 ± 0.02	. 4	7
		-		

\*Belov et al. also reported T =  $1.35 \pm 0.04$  MeV for thermal neutron-induced fission of 239Pu. 36

The above fission spectra tend to be harder than  $^{235}$ U except for spontaneous fission of  $^{240}$ Pu and  $^{242}$ Pu. Since only one measurement is available on each of these, definite conclusions must await further experimental work. It should be pointed out that the temperatures quoted above are based on individual measurements which show significant deviations from the average; for example, the maximum and minimum for  $^{252}$ Cf differ by 18% while  $^{235}$ U and  $^{239}$ Pu show somewhat better agreement. This means that  $^{235}$ U is in considerably better shape than the other isotopes.

#### REFERENCES

- 1. B. E. Watt, "Energy Spectrum of Neutrons from Thermal Fission of U235," Phys. Rev. <u>87</u>, 1037 (1952).
- 2. W. N. McElroy, "Implications of Recent Fission-Averaged Cross-Section Measurements," Nucl. Sci. Eng. <u>36</u>, 109 (1969).
- 3. T. W. Bonner, R. A. Ferrell, and M. C. Rinehart, "A Study of the Spectrum of the Neutrons of Low Energy from the Fission of U235," Phys. Rev. <u>87</u>, 1032 (1952).
- 4. D. L. Hill, "The Neutron Energy Spectrum from <sup>235</sup>U Thermal Fission," Phys. Rev. 87, 1034 (1952).
- 5. L. Cranberg, G. Frye, N. Nereson, and L. Rosen, "Fission Neutron Spectrum of U235," Phys. Rev. 103, 662 (1956).
- 6. H. Condé and G. During, "Fission Neutron Spectra, Part II, Fission Neutron Spectra of 235U, 239Pu, and 252Cf," Arkiv Fysik 29, 313 (1965).
- 7. E. Barnard, A. T. G. Ferguson, W. R. McMurray, and I. J. Van Heerden, "Time-of-Flight Measurements of Neutron Spectra from the Fission of U235, U238 and Pu239," Nucl. Phys. 71, 228 (1965).
- 8. J. A. Grundl, "A Study of Fission-Neutron Spectra with High-Energy Activation Detectors. Part II: Fission Spectra," Nucl. Sci. Eng. <u>31</u>, 191 (1968).
- 9. A. Fabry and M. De Coster, "Integral Test of Capture Cross Sections in the Energy Range 0.1 - 2 MeV," Proc. Second Conf. on Neutron Cross Sections and Technology, Spec. Publ. 299, Vol. 2, p. 1263, National Bureau of Standards (1968).
- 10. W. G. Davey, "Selected Fission Cross Sections for <sup>232</sup>Th, <sup>233</sup>U, <sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, <sup>237</sup>Np, <sup>238</sup>U, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, and <sup>242</sup>Pu," Nucl. Sci. Eng. <u>32</u>, 35 (1968).
- 11. "Neutron Attenuation in Optically Thick Shields," ANS-SD-1, Edited by R. L. Ashley and W. E. Kreger (April 1964); H. G. Goldstein, private communication.
- 12. "Annual Technical Progress Report, IMFBR Physics Programs GFY 1969," AI-AEC-12857 (Physics), (Sept. 30, 1969) and H. Alter, private communication.
- 13. A. M. Bresesti, M. Bresesti, A. Rota, and R. A. Rydin, "Threshold Reaction Excitation Functions Intercalibrated in a Pure Fission Spectrum," Nucl. Sci. and Eng. 40, 331 (1970).
- 14. R. A. Rydin, private communication (1970).

· .

15. A. Staub and R. E. Swaja, "Consequences of Various Proposed Nuclear Data," Trans. Am. Nucl. Soc. <u>12</u>, 748 (1969).

- 16. R. B. Leachman and H. W. Schmitt, "The Cross Section for <sup>238</sup>U Fission by Fission Neutrons," J. Nucl. Energy I, 1957, Vol. 4, p. 38, Pergamon Press Ltd., London.
- 17. P. H. White, "Measurements of the <sup>235</sup>U Neutron Fission Cross Section in the Energy Range 0.04 MeV to 14 MeV<sub>s</sub>" J. Nucl. Energy <u>19</u>, 325 (1965).
- 18. W. E. Stein, R. K. Smith, and H. L. Smith, "Relative Fission Cross Sections of 236U, 238U, 237Np, and 235U," Proc. Conf. on Neutron Cross Sections and Technology, Washington, D. C., NBS Publication 299, <u>1</u> (1968) 503.
- 19. G. Hansen, S. McGuire, and R. K. Smith (corrections to the old LASL fission data), private communication (1969).
- 20. W. P. Poenitz, "Measurement of the <sup>235</sup>U Fission Cross Section in the keV Range," Proc. Conf. on Neutron Cross Sections and Technology, Washington, D. C., NBS Publication 299, <u>1</u> (1968) 503.
- 21. B. H. Patrick, M. G. Sowerby, and M. G. Schomberg, "Structure in the Fission Cross Section of 235U," AERE R 6350 (1970).
- 22. G. A. Keyworth et al., LASL, private communication (1970).
- 23. C. D. Bowman, M. L. Stelts, and R. J. Baglan, "keV Fission Cross Section of 235U Measured with High Resolution," Paper CN-26/41 to be presented at IAEA Conf. on Nuclear Data for Reactors, Helsinki, 15-19 June 1970.
- 24. J. A. Grundl, "A Study of Fission Neutron Spectra with High-Energy Activation Detectors - Part I. Detector Development and Excitation Measurements," Nucl. Sci. Eng. 30, 39 (1967).
- 25. J. A. Grundl, R. L. Henkel, and B. L. Perkins,  $"^{31}P(n,p)^{31}$ Si and  $^{27}Al(n,\alpha)^{24}Na$  Cross Sections," Phys. Rev. <u>109</u>, 425 (1958).
- 26. L. E. Bobisud, "The Leakage Neutron Spectrum of a Spherical Critical Assembly of 233U," Master's Thesis, Univ. of New Mexico (1962).
- 27. G. M. Frye, Jr., J. H. Gammel, and L. Rosen, Los Alamos Scientific Laboratory, TID-10073 (1954).
- 28. L. Stewart, "Leakage Neutron Spectrum from a Bare <sup>239</sup>Pu Assembly," Nucl. Sci. Eng. 8, 595 (1960).
- 29. J. Neill, "Analysis of Spectral Measurements," taken from GGA Progress Report, GA-9753 (Sept. 1969), and private communication.
- 30. W. N. McElroy, R. J. Armani, and E. Tochilin, "Integral Flux Spectra and Spectral Averaged Cross Sections for a Bare <sup>235</sup>U Assembly and the Fission Spectrum," BNWL-SA-2684 (1969), and to be published.
- 31. G. G. Sherwood and J. S. King, "<sup>235</sup>U Fission Neutron Spectrum 6 to 20 MeV," Trans. Am. Nucl. Soc. <u>10</u>, 555 (1967).

- 32. I. Kimura, K. Kobayashi, and T. Shibata, "Measurement of Fast Neutron Spectrum with Threshold Detectors," KURRI-TR-61 (1969).
- 33. G. C. Hanna, C. H. Westcott, H. D. Lemmel, B. R. Leonard, Jr., J. S. Story, and P. M. Attree, "Revision of Values for the 2200 m/s Neutron Constants for Four Fissile Nuclides," Atomic Energy Review, Vol. 7, p. 3 (1969).
- 34. J. Terrell, "Fission Neutron Spectra and Nuclear Temperatures," Phys. Rev. <u>113</u>, 527 (1959); "Neutron Yields from Individual Fission Fragments," Phys. Rev. <u>127</u>, 880 (1962); and "Prompt Neutrons from Fission," in Physics and Chemistry of Fission, IAEA, Vienna (1965).
- 35. R. J. Doyas and R. J. Howerton, "Incident Energy Dependence of Fission Spectra, UCRL-50755 (1969).
- 36. L. M. Belov, M. V. Blinov, N. M. Kazarinov, A. L. Krivokhatskiĭ, and A. M. Protopopov, "Spectra of Fission Neutrons of <sup>244</sup>Cm, <sup>242</sup>Pu, and <sup>239</sup>Pu," Sov. J. Nucl. Phys. 9, 421 (1969).





-17-

## Figure 2

