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Fission-Neutron Spectra; Perspective and Suggestion^{*}

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by

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Preface

Recently reported macroscopic and spectrum-average measurements¹⁻⁵ and the analysis of fast-critical experiments⁴⁻⁹ have suggested an uncertain knowledge of prompt-fission-neutron spectra. These suggestions re-kindled the author's long-term interest in the fission spectrum.¹⁰⁻¹⁴ Specifically, the applied importance of these uncertainties stimulated additional work at this Laboratory with the objective of testing certain of the postulates put forth as the result of the macroscopic studies. In preparation for new experimental work the status of prompt-fission-neutron spectra was assayed and the results of that survey have received a limited distribution.¹⁵ The planned experimental work at this Laboratory is now complete.¹³ Essentially concurrently a number of new experimental results have become available at other laboratories. In the following resumé the present status of prompt-fission-neutron spectra is outlined inclusive of both the new and the older results. Primary emphasis is given to basic microscopic information with attention to associated experimental problems and limitations. Where appropriate, recommendations for future work are made.

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I. Macroscopic Characteristics

A. Fission-Neutron Spectra

Current uncertainties arise, to a large extent, from the results of recent macroscopic studies employing various reaction indices¹⁻⁵ and from the detailed analysis of fast critical assemblies.⁵⁻⁸

The interpretation and adjustment of basic data from the analysis of fast critical experiments, illustrated by the work of Campbell and Rowlands,⁶ is difficult. While the uncertainties in the deduced parameters are appreciable, Campbell and Rowlands suggest that the average-fission-neutron spectrum energy is 5-10% higher than indicated by microscopic measurement and that the spectrum shape may differ from a Maxwellian form. Similar uncertainties have been discussed in relation to the fast critical assemblies ZEBRA-2 and ZPR-3 by Okrent et al.⁷ The details of the fast critical analyses are too complex for description here. However, it appears that most of the calculations give no consideration to the dependence of the fission-neutron energy and/or spectrum on the energy of the fission-inducing incident neutron. This dependence maybe significant, particularly in very fast assemblies.¹⁶ Therefore:

Recommendation No. 1

The analysis of fast critical experiments should be inclusive of the dependence of fission-neutron spectra on incident neutron energy.

Some reaction-rate measurements in fission-neutron spectra tend to support the suggestions resulting from critical-assembly studies.¹⁻⁵ However, the interpretation is again neither simple nor unique as a number of uncertain physical parameters are involved in addition to the inherent experimental error. The interpretation of such reaction-rate studies has recently been reviewed by Fabry et al.¹ and by Grundl.⁴

The status of available macroscopic results is outlined in Table 1. The recent reaction-rate results of Refs. 2, 4 and 1 (to a lesser extent) all tend to indicate a larger average-fission-neutron energy than microscopic measurements. However, a number of other reaction-rate results are consistent with microscopic values including the recent work of Refs. 17 and 18. Generally the uncertainties in the macroscopic average energies are large or ill-defined. In contrast some ratios of average energies are given to within very small (fractional %) errors. The ratio values appear to be more reliable as is true in most microscopic measurements. Thus the fact that the macroscopic ratio values are consistently smaller than the corresponding microscopic values is particularly disturbing.

Over a number of years Fabry and co-workers¹ have experimentally studied the response of a wide range of detectors (mostly threshold reactions) in fission-spectrum environments. Apparently the initial work was similar to that of Depuydt and Neve de Mevergnies.²⁵ In any case, the fission-spectrum measurements were made with an "in-cavity" arrangement. The early Fabry results indicated a 15-20% harder fission neutron spectrum than is usually obtained from microscopic measurements. Fabry concluded that his results were consistent with those of Grundl⁴ and of Leachman and Schmidt.²⁶ The results were sensitive to the exact microscopic cross sections used in the interpretation. This critical matter is extensively discussed by Fabry in Ref. 1.

Grundl^{4,5} carefully studied reaction rates using both a monoenergetic neutron source (Van de Graaff) and a fission source. His work has the merit of careful relative detector calibration free of many cross section-associated uncertainties. However, Grundl points out the importance of an accurate absolute energy scale and notes that uncertainties in energy of as little as 150 keV can lead to gross errors in subsequently determined average-fission-neutron energy values. The importance of careful calibrations should not be underestimated. Therefore:

Recommendation No. 2

Reaction detectors employed in macroscopic studies should be well calibrated using a carefully controlled mono-energetic neutron source; e.g., similar to the method employed by Grundl.

Grundl, like Fabry, used an "in-cavity" arrangement for producing a fission-neutron spectrum. Forty percent of the return flux from the surrounding material (a D₂O water tank) was above 0.1 MeV. Grundl considered in detail this cavity perturbation using DSN calculations. However, the return-flux correction may not have been perfect and it is noted that the same laboratory went to considerable trouble to hang GODIVA (a bare metal critical) well above the ground to obtain a high-fidelity leakage spectrum.

The cavity problem has been serious since Chadwick's time and it remains so. Therefore:

Recommendation No. 3

Generally, fission-neutron studies should give careful attention to the spectral fidelity with as near an "in-vacuo" environment as possible.

Grundl deduced ratios of the average-fission-neutron energies of U-233 and Pu-239 to that of U-235 (see Table 1). The associated errors are very small. The Grundl ratios are very similar to other macroscopic ratio values; for example those of Bonner,²¹ Harris¹⁹ and Kovalev et al.²⁴ However, all macroscopic-ratios remain seriously discrepant with those deduced from the majority of microscopic measurements. Grundl indicates an average-fission neutron energy for U-235 of ~ 2.2 MeV, appreciably higher than that deduced from the majority of the microscopic and macroscopic measurements. He further suggests that the spectrum at low neutron energies is less intense than that given by a Maxwellian form in contrast to the opposite trend in some microscopic measurements.^{27,28,41}

McElroy^{2,3} has extensively analyzed reaction-rate measurements, primarily those of Grundl⁴ and of Fabry.¹ He employs an iterative procedure with the computation code SAND-II and a library of selected microscopic cross sections to deduce detailed spectral distributions from the measured reaction rates. He suggests an average U-235 fission neutron energy of ~ 2.24 MeV, similar to that proposed by Grundl. In addition the spectrum deduced by McElroy is 20-40% lower than that indicated by a Maxwellian distribution at neutron energies of ~ 800 keV. This is a large difference in a region where microscopic measurements are reliable. Indeed, a recent specifically designed microscopic experiment failed to verify the McElroy spectrum at these low energies.¹³ To what extent the large discrepancy is due to uncertainties in the SAND-II procedures, the cross section library or the respective measurements is a moot question. However, it has been suggested that the SAND results are sensitive to small and essentially unknown sub-threshold reaction cross sections.²⁹ Therefore:

Recommendation No. 4

The sensitivity of macroscopic results to microscopic cross section values should be carefully assayed with recommendations as to what measurements and to what accuracies are required to give a reliable interpretation of important macroscopic measurements.

Macroscopic measurements, particularly those dealing with critical assemblies, may be limited by the design of the system which is often oriented towards specific engineering tests not necessarily compatible with a sensitive data assay. Therefore:

Recommendation No. 5

Apparatuses for macroscopic studies should be optimized for data test purposes particularly in the area of fast-critical experiments. (The extensive Los Alamos studies of bare critical-spheres are illustrative of the concept of this recommendation).

B. Fission-Neutron Cross Section of U-238 in a U-235 Fission-Neutron Spectrum.

The quantity $\sigma_f(\chi_{235}^{238}\text{U})$ is frequently cited as an index of the fission-neutron spectrum (and of $\sigma_f(238)$). Representative experimental and calculated values are outlined in Table 2. The experimental values of Leachman and Schmidt,²⁶ of Richmond³¹ and of Nikolaev et al.³⁰ are in the range 310-315 mb. The measured results of Fabry et al.¹ are somewhat larger. All of the experimental values are appreciably larger than the result calculated by Grundl⁴ using microscopic spectra and selected U-238 fission cross sections. However, the calculated result of Campbell and Rowlands⁶ based upon the data of Hart³² is in reasonable agreement with the measurements although the fission spectrum used in the calculation is not clear from Ref. 32. The results depend on the fission cross section of U-238 in the MeV region where recent microscopic measurements³³ indicate larger values than found in widely used evaluations.²⁰

$\sigma_f(\chi_{235}^{238}\text{U})$ measurements have generally employed steady-state neutron beams or fluxes. Precision microscopic fission cross section measurements employ pulsed-neutron sources and fast timing techniques to control background effects. Therefore:

Recommendation No. 6

$\sigma_f(\chi_{235}^{238}\text{U})$ should be determined using pulsed-source and fast timing techniques to insure against background distortions.

When recommendation No. 6 has been carried out and when the fission cross section of U-238 is well known the parameter $\sigma_f(\chi_{235}^{238}\text{U})$ will be a far more valuable fission-spectrum index.

C. Age to Indium Resonance in H₂O

The study of the age of U-235 fission neutrons to indium resonance in H₂O has not received the attention warranted by its basic importance and relation to the fission-neutron spectrum. The measured and calculated results are outlined in Table 3. Experimental values prior to 1961 tend to be large; 27-31 cm².³⁵ More recent experimental results are in the range 26-28 cm².^{36,37} The experiments did not generally use point sources but rather combinations of plane and other complex reactor-driven sources. As a consequence the deduction of the fundamental parameter from the measured values entailed considerable correction. Recent theoretical calculations based upon microscopic fission-neutron spectra^{22,38} yield calculated ages very close to the later experimental values. Further, from age considerations Story¹⁸ concluded that the average-fission-neutron energy is very similar to the microscopic value. Harris¹⁹ has pointed out that age is sensitive to changes in the average-fission-neutron energy; a 10% change in age corresponding to a ~ 200 keV change in average energy at 2.0 MeV. He further concludes that $\bar{E}(\text{Pu-239})/\bar{E}(\text{U-235}) \sim 1.04$, a value similar to other macroscopic results.^a

Age is a good index of the fission-neutron spectra both relatively and absolutely. Therefore:

Recommendation No. 7

The age of Cf-252 fission-neutrons to indium resonance in H₂O should be determined to ± 1 cm² using an ideal point source.

^aThroughout this document $\bar{E}$ denotes average-fission-neutron energy.

Carrying out this recommendation will; a) provide a good "standard" age with an ideal point source geometry free of many experimental corrections, b) provide a standard for sensitive determination of the ratio of U-235 and Pu-239 age to that of Cf-252 with consequent definition of differences in average-fission-neutron energies, and c) contribute to the resolution of the current discrepancies in the average energy of the Cf-252 spectrum. The recommendation is for a relatively simple measurement and sources of sufficient intensity can be obtained.

II. Microscopic Characteristics

A. Average-Fission-Neutron Energies and their Ratios.

More than sixty microscopic measurements of fission-neutron spectra are reported in the literature. These results are outlined in Table 4 with the associated references giving an indication of method, range, quality and unusual properties.³⁹ More than thirty of the measurements pertain to U-235 and Pu-239 with the remainder distributed over the mass region $A = 229 - 252$. The incident neutron energy range extends from thermal to 14.3 MeV with additional spontaneous fission processes, principally Cf-252. The experimental techniques employed generally fall into three categories; 1) time-of-flight (TOF) using proton-recoil or reaction detectors, 2) proton-recoil spectrometers either in the form of counters or emulsions, and 3) reaction spectrometers such as Li-6 and He-3 counters. The measurements are most straightforward when obtained with thermal or low-energy (< 1.0 MeV) neutron-induced or spontaneous fission. At higher incident neutron energies the observed spectrum is complicated by contributions from other neutron emitting processes, such as inelastic scattering, and the requisite corrections lead to greater uncertainties. This is particularly true for emulsion and other techniques which do not explicitly identify the origin of the observed neutron. Some of the more careful TOF measurements are not subject to this criticism, for example those of Refs. 47 and 49.

A qualitative inspection of Table 4 reveals several general characteristics. 1) No thermal or low-energy-neutron-induced fission spectrum for $A \leq 244$ has been microscopically observed to have an average energy as large as the ~ 2.2 MeV ascribed to some of the macroscopic results. 2) Uncertainties assigned to the average-energy values are not consistent with the discrepancies between measurements. The experimentalists have apparently been optimistic with a certain affinity for errors of 40-60 keV. 3) The ratio $\bar{E}(\text{Pu-239})/\bar{E}(\text{U-235})$ deduced from microscopic measurements is, with one exception, consistently larger than the comparable macroscopic value. The exception is in doubt as the same group obtained a larger value using an alternate technique.²⁷ 4) There maybe some tendency for microscopic $\bar{E}$ values to grow with time but probably by less than 50-100 keV at most.

The average-neutron energies of U-235 thermal and low-energy neutron-induced fission are outlined in Table 5. The weighted average is 1.979 MeV with an RMS deviation of 4.3%. This average is not consistent with the larger macroscopic results. A similar outline of the average-fission-neutron energies of Pu-239 is given in Table 6. Two values of this Table are lower than the rest of the set with that of Ref. 50 being from a generally low set of values. The weighted average of Table 6 is 2.084 MeV or 2.093 MeV if the low value of Ref. 50 is omitted. The ratio of the average values of Tables 5 and 6 is $\bar{E}(\text{Pu-239})/\bar{E}(\text{U-235}) = 1.053 \pm 0.050$. The relatively large error does not make a comparison with the macroscopic results particularly rewarding and the explicitly measured ratios of Table 7 are strongly preferred for definitive comparisons. The average of the directly measured values is $\bar{E}(\text{Pu-239})/\bar{E}(\text{U-235}) = 1.084 \pm 0.006$. This average is not inclusive of the exceptionally low value of Ref. 27 (which could not be verified by other work at the same institution). Thus the directly measured microscopic Pu-239/U-235 ratios are significantly higher than the comparable macroscopic values given in Table 1. This is disturbing as the measured ratios should be experimentally reliable and, as was

pointed out in Ref. 13, the results are not particularly dependent on the spectral shape.

Of the spontaneous-fission-neutron spectra that of Cf-252 has been the most extensively studied. The results are outlined in Table 8. The spread in the experimental values is, in part, the consequence of weak sources available for some of the early work (for example Ref. 61). However, more recent values obtained with stronger sources differ by far more than their respective errors. It has been suggested by Jéki et al.⁷² that backgrounds have seriously perturbed experimental Cf-252 average energies generally reducing the true values by 100-200 keV. Some values of the ratio $\bar{E}(\text{Cf-252})/\bar{E}(\text{U-235})$ have been determined (see Table 4). These ratios are generally associated with measurements giving lower Cf-252 average-energy values. The discrepancies between the various Cf-252 results present a serious problem. Accurate Cf-252 results are important to the determination of an easily used "standard" fission-neutron spectrum and they effect the determination of other important quantities such as Nu-bar. The Cf-252 results strongly influence the determination of spectral dependence on Nu-bar and, indirectly, on incident neutron energy. Therefore:

Recommendation No. 8

High priority should be given to the determination of the Cf-252 fission-neutron spectrum to the best possible precision from a few keV to at least 10 MeV. The quality of the results should be such as to make this THE "standard" fission-neutron spectrum.

The experimental procedures of Recommendation No. 8 are simpler than those associated with neutron-induced fission. Suitable sources can be obtained. The result will be an easily applied "standard" spectrum not requiring relatively intense neutron beams or other complex facilities. Associated with Recommendation No. 8 are:

Recommendation No. 9

The ratios $\bar{E}(X)/\bar{E}(\text{Cf-252})$ should be determined to 1% precision particularly where "X" is U-235 and Pu-239. The measurements should be triads $\bar{E}(X)/\bar{E}(Y)$, $\bar{E}(X)/\bar{E}(\text{Cf-252})$ and $\bar{E}(Y)/\bar{E}(\text{Cf-252})$ so as to provide a check of internal consistency.

Recommendation No. 10

The spectral ratio $X_N(E)/^{252}\text{N}(E)$ should be determined throughout the range several-keV to 10 MeV where "X" is U-235 and Pu-239. The measurements should involve two energy resolutions; 1) coarse (~ 100 keV) with high precision for general energy dependence and 2) fine ($\sim$ tens of keV) for assay of possible structure.

Recommendation No. 11

Selected reaction-rate indices useful in macroscopic measurements should be calibrated in the Cf-252 spectrum. Particularly important is the quantity $\sigma_f(\chi_{252}^{238}\text{U})$.

Recommendation No. 12

Generally, microscopic fission-neutron-spectrum measurements should emphasize techniques that specifically identify the observed neutron as being of fission origin.

B. Spectrum Shape and Structure.

The observed fission-neutron spectra are usually described by either the "Watt"⁴⁵ expression

$$N(E)dE \sim \exp(-bE) \operatorname{sinc} \sqrt{cE}dE \quad (1)$$

or a Maxwellian distribution

$$N(E)dE \sim \sqrt{E} \exp (-E/T). \quad (2)$$

The latter was theoretically proposed by Terrell⁶² from consideration of neutron evaporation from the moving fragment described by a Weisskopf "temperature", T . As noted by Weisskopf,⁶³ the temperature concept is only qualitative and one could reasonably expect considerable deviation from this simple approximation when dealing with the complex fission process. Indeed, experiment indicates that some of the fission neutrons are emitted at the actual scission rather than subsequently from the moving fragments.⁶⁴ Furthermore, the fragments are very highly excited and measurements indicate that multiple emission with varying temperatures does occur.^{14,60} Despite these complexities, either of the above forms has been shown to be qualitatively descriptive of microscopic measurements. From a pragmatic point of view the difference between the two spectral forms is small; less than 5% below 6 MeV. At 10 MeV the difference is 25% but at this energy the spectral intensity has decreased by more than two orders of magnitude from the most probable value and as a consequence the relatively poor experimental statistical accuracy cannot clearly differentiate between the two above expressions. From the standpoint of numerical manipulation the Maxwellian is probably to be preferred.

Neither Eq. (1) or (2) fully describe all of the experimental results. Several measurements indicate an abundance of low energy neutrons well above that predicted by either expression.^{28,41,70} The low-energy excess is particularly evident in Cf-252 fission as measured by Meadows²⁸ and by Zamyatin et al.⁴¹ A careful inspection of a number of other measured spectra reveals a consistent tendency for a low-energy neutron excess for several of the fission processes. Good examples are the spectra of U-235, Pu-239 and Cf-252 shown in Ref. 27. Furthermore, some fast critical studies have suggested a similar low-energy excess.⁶⁵ Low-

energy measurements that do not show the excess tend to be of poorer accuracy (cloud chamber studies, for example). Therefore:

Recommendation No. 13

The low energy non-maxwellian behavior of the Cf-252 spectrum should be investigated to accuracies of $\sim 2\%$.

The above is redundant with Recommendation No. 8 but can be followed with a less comprehensive experimental program and thus is separately stated. The Recommendation should be extended to other fissile isotopes through Recommendation No. 10.

Neutron emission from highly excited fragments can be complex and a small portion of the emitted neutrons may arise from $(f;x,n)$ processes. These could give a structure to the fission-neutron spectrum. Indeed, Zamyatin et al.⁴¹ and Nefedov⁷¹ reported a structure in the measured Cf-252 and U-235 fission neutron spectra. Nefedov associates this structure with specific fragment energies. This structure has escaped notice in other detailed work such as that of Meadows.²⁸ Furthermore, no structure was observed in a search of a number of U-235 and Pu-239 fission spectra by Smith¹³ and it seems unlikely that the phenomena is peculiar to Cf-252. Structure of the type reported in Ref. 41 should be cautiously considered as a number of experimental artifacts can contribute to the observed effect and have done so in similar processes such as $n-n'$ scattering. Thus, before the reported structure is accepted it is suggested that Recommendation No. 14 be pursued.

Recommendation No. 14

Structure in the fission spectrum should be assayed with a specifically designed ratio experiment. Properly executed, a ratio determination will cancel most experimental artifacts while revealing differences in structure. It is unlikely that the structure will be identical for each fissile isotope therefore it will be apparent in the ratios.

Recommendation No. 14 is redundant with Recommendation No. 10 but is individually stated due to its simpler and more explicit nature.

Several measurements^{14,60} observed the fission-neutron spectra in correlation with the mass and direction of motion of the fission fragments. The results are largely explained in terms of evaporation postulates with a light/heavy fragment neutron-emission ratio approximately 1.2 - 1.4. The results are not particularly sensitive to any reasonable anisotropy of the neutron emission from the moving fragment. The direction of fission fragment motion is known to be correlated with incident-neutron direction at some incident-neutron energies dependent upon the particular fission channels involved. The correlation is particularly pronounced near threshold, for example in U-238 fission. In these selected regions one would expect a correlation between the incident neutron direction and the fission spectrum if the evaporation hypothesis holds true. A possible indication of such an effect has been observed in U-238 by Knitter et al.⁶⁹ However, the incident neutron energies of the Knitter and of other work do not exactly correspond to those of a strong incident-neutron to fragment correlation and the incident resolutions employed in the fission spectrum measurements are usually coarse compared to the energy dependence of the fragment correlations. An incident-neutron to fission-neutron angular correlation could contribute to an anomalous structure observed in some measurements made at selected incident energies and reaction angles. This possible effect was, of course, avoided in the more detailed studies such as those of Refs. 13 and 49 where the spectrum was observed at a number of reaction angles. In any case, an angular correlation, localized in incident energy, will not be of appreciable applied significance.

C. Spectral Dependence on Nu-bar* and Incident Energy.

There are a number of microscopic results obtained at incident neutron energies well above thermal (see Table 4). However, as noted above, some of these results are open to considerable question as the measured values may be heavily contaminated with neutrons originating in processes other than fission. The number of measurements at high incident energies where the fission origin of the neutrons is assured is too limited for a good definition of spectral dependence on incident neutron energy. However, there are a number of spontaneous fission results available, notably for Cf-252. These are not as subject to ambiguous interpretation and span a wide range of Nu-bar values. Nu-bar is fairly well known both for spontaneous fission and as a function of incident neutron energy. Therefore the dependence of microscopic fission-neutron spectra on Nu-bar and, indirectly, on incident neutron energy can reasonably be examined. The Nu-bar route is that followed here.

From basic considerations of kinetics and the equation-of-state Terrell⁶² relates the average-fission-neutron energy, $\bar{E}$, to Nu-bar through the relation

$$\bar{E} = A + B \sqrt{\nu + 1} \quad (3)$$

where $A = 0.75$ and $B = 0.65$.

The values of A and B were determined from a comparison with experimental results. The expression of Eq. 3 is compared with the experimental values of Table 4 in Fig. 1. The requisite Nu-bar values were taken primarily from the IAEA tabulations and the work of Soleihac et al.⁶⁶ and from a few additional sources where necessary. $\bar{E}$ is not a strong function of Nu-bar and thus the comparison is not appreciably influenced by the relatively small uncertainties in Nu-bar.

* Herein Nu-bar is defined as the average number of prompt neutrons emitted per fission.

The data of Fig. 1 is grouped about the thermal neutron-induced fission of U-235 and Pu-239 and the spontaneous fission of Cf-252. The remainder of the points correspond to a limited number of recent spontaneous fission results, primarily from Ref. 41, and various measurements made with incident neutrons with energies up to 14.3 MeV. The Terrell expression is indicated by the solid curve. A least-squares fit to all of the data, weighted by $1/\text{error}^2$, gives the dashed curve. The dashed-dotted curve was obtained by a similar fitting procedure but omitting those values possibly contaminated with non-fission neutrons. Generally, the slope of the curve rises as the basic data becomes more comprehensive and then more selective. However, none of the curves are consistent with the microscopic measurements of the ratio $\bar{E}(\text{Pu-239})/\bar{E}(\text{U-235})$ (for example, the Terrell ratio = 1.04 and that of Table 7 = 1.084). Further, none of the curves are consistent with the higher $\bar{E}(\text{Cf-252})$ values nor with many of the $\bar{E}$ values corresponding to the Nu-bar range 2.65 - 3.70. The conclusion is that a number of experimental values are systematically in error or that Eq. 3 is not quantitatively descriptive of the physical phenomena; or both. The above recommendation emphasizing the importance of the Cf-252 spectrum and of precise ratio values should resolve the issue.

D. Comments on Flux Normalization and Detector Efficiency

Microscopic fission-spectrum measurements seek to determine the relative energy distribution of a continuum-temperature spectrum over the extended energy range 1 keV to 10 MeV or more. The discrepancies between measured values are of the order of 1-10%. Measurements of this nature are exceedingly difficult requiring detailed attention to the calibration of the detector response regardless of the specific method. Some indication of the difficult nature of the problem is to be found in similar x-n spectral measurements which generally do not provide temperature distributions for nuclear processes approaching the accuracies sought in fission-spectrum measurements.

Many of the fission measurements are directly or indirectly based upon the n-p cross section. This cross section is known to suitable accuracy⁶⁷ but its utilization in the laboratory often leaves much to be desired. Little attention is given to the effects of multiple processes or to the presence of carbon in the hydro-carbons often employed as detectors. The latter may be a source of anomalous structure as carbon scattering is resonant over much of the range of interest. The sensitivity of the detection system is often deduced only from calculation. Such calculations have not proven outstandingly reliable in fast-neutron-flux determinations. In some instances the detection efficiency is "verified" by observation of some "known" source reaction such as $D(d,n)$. It is not clear that these source distributions are sufficiently well known to serve as a standard.

The above indicates that the microscopic measurement of fission spectra is a difficult flux measurement problem and that greatly increased attention should be given to the quantitative calibration of the detectors employed using the n-p cross section over the entire energy range and/or the carbon scattering cross section at lower energies (below 1.8 MeV). Any measurement without such careful calibration of the detection system using a well controlled mono-energetic neutron source may be subject to systematic uncertainties. Therefore:

Recommendation No. 15

Measurement systems employed in microscopic-fission-spectrum experiments should be well calibrated using a controlled mono-energetic neutron source and a standard reaction such as n-proton or n-carbon scattering. The calibration should be inclusive of corrections for multiple processes and other perturbations.

The above statements are not as applicable to spectrum-ratio measurements wherein the detection efficiency does not directly relate to the result.

E. Isomeric Fission*

It has recently been reported that U-235 fission induced by 30 keV neutrons proceeds to an appreciable extent through an isomeric state in U-236 with a half-life in the order of 100 nsec.⁶⁸ Such processes in this and similar fissile nuclei could distort fission-neutron-spectrum measurements based upon the time-of-flight technique wherein the experimental time scale is in the range 0-500 nsec. In view of this possibility a detailed search for such isomeric fission was carried out with 30 keV neutrons incident on U-235. The method employed the fast-pulsed-beam technique and a rapid response gas scintillation counter. Of a total of 3.67×10^4 observed fission events approximately 0.12% were non-prompt and at least 75% of these were attributed to the measured room background. It was concluded that the relative contribution of the U-236 isomeric process at a 30 keV incident-neutron energy was less than 3×10^{-2} percent. This is one to two orders of magnitude lower than that indicated in Ref. 68 and too small to be significant in fission-neutron-spectrum measurements employing fast-neutron-time-of-flight techniques.

III. Concluding Remark

The above is a resumé of current knowledge of fission-neutron spectra with emphasis on microscopic quantities. Clearly, there is a discrepancy between some macroscopic results and the microscopic values which, on the average, have remained relatively static for a number of years. The discrepancy is by no means universal nor is it becoming more acute with time. It should be stressed that microscopic knowledge of the fission-neutron-spectrum is good in the context of the difficult nature of the problem and in comparison with our understanding of other neutron processes. For example, few percent differences between the "Watt" and Maxwellian forms or between measured $\bar{E}(\text{Pu-239})/\bar{E}(\text{U-235})$ ratios are no larger than current discrepancies in fast-fission cross sections of U-235; one of the most basic cross sections. It will not be easy to

*The isomeric studies reported here were carried out by J. W. Meadows and W. P. Poenitz, Argonne National Laboratory.

grossly improve the already relatively good quality of the microscopic information. The recommendations made herein involve many man-years of effort and even if exactly followed will likely not lead to an order of magnitude improvement in the basic information. Hopefully the recommendations will forestall inappropriate or ill-conceived endeavors and improve the basic data by a factor of 2-5.

It has been suggested that an evaluation of the basic data is now in order. The contemporary merit of such an effort can be questioned. Many of the available results, though of good quality, are old and poorly documented. As a consequence the evaluator may tend to equate newness with goodness to the detriment of physical fact. At present an evaluation can massage and renormalize to remove some of the more glaring discrepancies between, for example, average energies and ratios; but it is unlikely that it will resolve the basic issue -- the difference between some macroscopic deductions and microscopic values. It is here suggested that an evaluation would be more pregnant several years hence when, hopefully, the concepts and criticisms of this and similar meetings have born fruit. Until that time, in this author's opinion, the fission-neutron spectra stands as indicated by the microscopic results outlined above.

IV. Summary of Recommendations

- No. 1 The analysis of fast critical experiments should be inclusive of the dependence of fission-neutron spectra on incident neutron energy.

- No. 2 Reaction detectors employed in macroscopic studies should be well calibrated using a carefully controlled mono-energetic neutron source, e.g., similar to the method employed by Grundl.

- No. 3 Generally, fission-neutron studies should give careful attention to the spectral fidelity with as near an "in-vacuo" environment as possible.

- No. 4 The sensitivity of macroscopic results to microscopic cross section values should be carefully assayed with recommendations as to what measurements and to what accuracies are required to give a reliable interpretation of important macroscopic measurements.

- No. 5 Apparatuses for macroscopic study should be optimized for data test purposes particularly in the area of fast-critical experiments. (The extensive Los Alamos studies of bare critical spheres are illustrative of this recommendation).

- No. 6 $\sigma_f(\chi_{235} \text{ } ^{238}\text{U})$ should be determined using pulsed-source and fast timing techniques to insure against background distortions.

- No. 7 The age of Cf-252 fission-neutrons to indium resonance in H₂O should be determined to $\pm 1 \text{ cm}^2$ using an ideal point source.

- No. 8 High priority should be given to the precision determination of the Cf-252 fission-neutron spectrum to the best possible precision from a few keV to at least 10 MeV. The quality of the results should be such as to make this the "standard" fission-neutron spectrum.

Summary of Recommendations (Contd.)

- No. 9 The ratios $\bar{E}(X)/\bar{E}(\text{Cf-252})$ should be determined to 1% precision where "X" particularly is U-235 and Pu-239. The measurements should be triads $\bar{E}(X)/\bar{E}(Y)$, $\bar{E}(X)/\bar{E}(\text{Cf-252})$ and $\bar{E}(Y)/\bar{E}(\text{Cf-252})$ so as to provide a check of internal consistency.
- No. 10 The complete spectral ratio $X_N(E)/^{252}\text{N}(E)$ should be determined throughout the range few-keV to 10 MeV where "X" is U-235 and Pu-239. The measurements should involve two energy resolutions; 1) coarse (few 100 keV) for generally energy dependence and 2) fine (few tens of keV) for assay of possible structure.
- No. 11 Selected reaction-rate indices useful in macroscopic measurements should be calibrated in the Cf-252 spectrum. Particularly important is the quantity $\sigma_f(\chi_{252}^{238}\text{U})$.
- No. 12 Generally microscopic fission-neutron-spectrum measurements should emphasize techniques that specifically identify the observed neutron as being of fission origin.
- No. 13 The low energy non-maxwellian nature of the Cf-252 spectrum should be investigated to accuracies of 2%.
- No. 14 Structure in the fission spectrum should be assayed with a specifically designed ratio experiment. Properly executed a ratio determination will cancel most experimental artifacts while revealing differences in structure. It is unlikely that the structure will be identical for each fissile isotope therefore the differences will be significant.
- No. 15 Measurement systems employed in microscopic-fission-spectrum experiments should be well calibrated using a controlled mono-energetic source and a standard reaction such as n-proton or n-carbon scattering. The calibration should be inclusive of corrections for multiple processes and other perturbations.

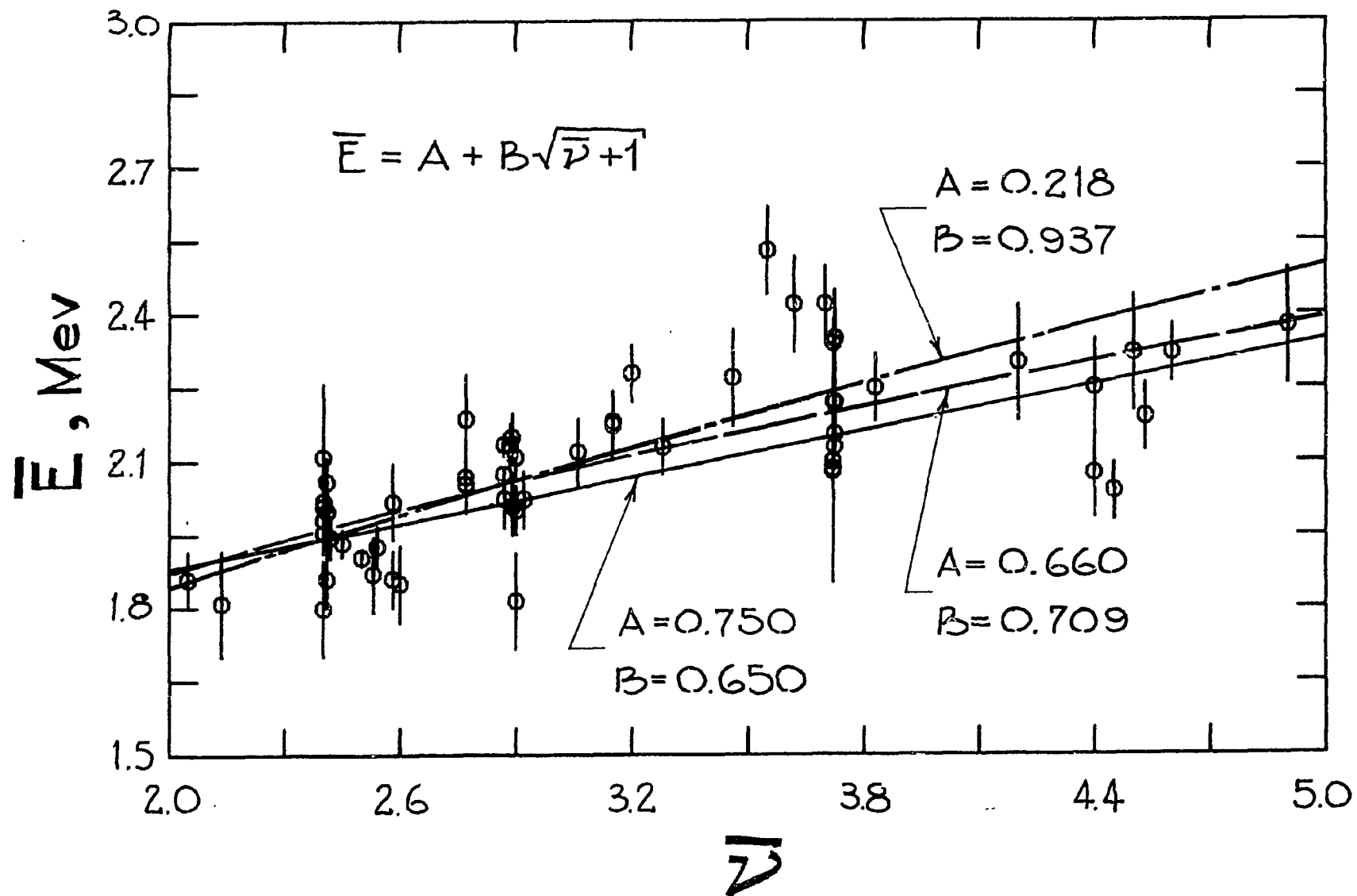


Fig. 1 Dependence of average-fission-neutron energy, $\bar{E}$, on $\bar{\nu}$. Data points are from Table 4. Curves indicate the Terrell⁶² expression with various parameter choices as described in the text.

TABLE 1. MACROSCOPIC FISSION SPECTRUM PARAMETERS.

ISOTOPE	E(AVE), MEV ^A	E(IN), MEV ^B	RATIO ^C	REFERENCE
U-235	1.998	TH	0.982	(21)
	-----	TH	1.015	(19)
	-----	TH	1.021+-0.005	(4)
	-----	TH	1.020+-0.005	(24)
	1.960			(20) ENDF
U-235	2.100 +-0.100	FAST CRIT.	-----	(6)
	2.034	TH	-----	(21)
	1.950	TH	-----	(17)
	1.950-2.025	FAST	-----	(1)
	2.200	TH	-----	(4)
	2.240	TH	-----	(2)
	2.060	TH	-----	(22)
	2.025	TH	-----	(18)
	1.950			(20) ENDF
PU-239	2.077	TH	1.021	(21)
	-----	TH	1.040	(19)
	-----	TH	1.039 +-0.002	(4)
	-----	TH	1.025+-0.0062	(24)
	2.115			(20) ENDF
PU-240	1.738	SP	0.854	(21)
CF-252	2.050	SP	1.007	(21)
	2.085 +-0.06	SP	-----	(23)

FOOTNOTES-

- A. AVERAGE FISSION NEUTRON ENERGY AS GIVEN OR DEDUCED FROM THE BASIC REFERENCE. ERRORS ARE OMITTED WHEN NOT DIRECTLY AVAILABLE.
- B. INCIDENT NEUTRON ENERGY. TH=THERMAL, FAST OR FAST CRIT.= FAST CRITICAL SPECTRUM, SP=SPONTANEOUS FISSION.
- C. RATIO $E(AVE-X)/E(AVE-235)$ IS GIVEN WHEN DIRECTLY MEASURED OR DEDUCED FROM SAME EXPERIMENTAL SET.

Table 2: Measured and Calculated Values of $\bar{\sigma}_f(\chi_{235} \text{ } ^{238}\text{U})$

Ref.	$\bar{\sigma}_f$, mb
<u>Measured</u>	
Fabry et al. ¹	$353 \pm 30, 374 \pm 30$
Nikolaev et al. ³⁰	310 ± 10
Leachman and Schmidt ²⁶	313 ± 5
<u>Calculated</u>	
Grundl ⁴	$273 - 282 \pm 5^a$
Campbell and Rowlands ⁶	301

^aThe lower value was obtained with a Maxwellian temperature = 1.29 MeV.
The higher value from the Cranberg expression³⁴.

Table 3: Age to Indium Resonance in H₂O

Ref.	Age (cm ²)
<u>Measured</u>	
Summary of pre-1961 values ³⁵	27 - 31
Doerner et al. ³⁶	27.9 ± 0.1
Paschall ³⁷	26.6 ± 0.3
<u>Calculated</u>	
Dunford and Alter ²²	26.46 ± 0.32
Staub et al. ³⁸	25.4 ± 26.4
Story ¹⁸	Deduces Average fission energy to be ~ 2.025 MeV

TABLE 4. MICROSCOPIC FISSION SPECTRUM PARAMETERS

ISOTOPE	A E(AVE), MEV	B E(IN), MEV	C RATIO	REFERENCE
TH-229	1.860 $\pm$ 0.060	TH	-----	(41)
TH-232	2.250	14.0	1.068	(42)
U-233	1.870 $\pm$ 0.080	TH	0.960 $\pm$ 0.03	(11)
	EXP. DECADE	TH	-----	(43)
	= ABQU! 4.2 MEV			
	2.300 $\pm$ 0.120	14.0	1.108	(42)
U-235	2.010 $\pm$ 0.00	TH	-----	(40)
	1.956 $\pm$ 0.013	TH	-----	(27)
	2.020 $\pm$ 0.025	TH	-----	(27)
	2.110 $\pm$ 0.150	0.03-0.4	-----	(13)
	1.900 $\pm$ 0.200	TH	-----	(44)
	2.000	TH	-----	(45)
	EXP. DECADE	TH	-----	(46)
	= 3.900 $\pm$ 0.2 MEV			
	2.060	0.03	-----	(34)
	1.946 $\pm$ 0.045	0.1	-----	(47)
	1.800 $\pm$ 0.100	TH	-----	(48)
	1.905 $\pm$ 0.019	0.95	-----	(49)
	1.860 $\pm$ 0.060	0.04	-----	(50)
	1.860 $\pm$ 0.060	1.5	-----	(50)
	2.040 $\pm$ 0.060	14.3	-----	(51)
	2.075 $\pm$ 0.090	14.0	-----	(42)
	1.982	TH	-----	(52)
U-238	1.927 $\pm$ 0.045	2.09	-----	(47)
	2.133 $\pm$ 0.048	4.09	-----	(47)
	1.935 $\pm$ 0.030	1.35	-----	(49)
	2.190 $\pm$ 0.070	14.3	-----	(51)
	2.320 $\pm$ 0.120	14.0	1.118	(42)
	2.019 $\pm$ 0.080	1.9	-----	(69)
	1.850 $\pm$ 0.080	2.3	-----	(69)
PU-238	2.025 $\pm$ 0.060	TH	-----	(41)

TABLE 4, (CONT.)

ISOTOPE	E(AVE), MEV	E(IN), MEV	RATIO	REFERENCE
PU-239	2.136 $\pm$ 0.024	TH	1.092 $\pm$ 0.014	(27)
	2.075 $\pm$ 0.017	TH	1.028 $\pm$ 0.012	(27)
	-----	0.03 -0.4	1.075 $\pm$ 0.02	(13)
	EXP. DECADE	TH		(53)
	$\approx$ 4.5 $\pm$ 0.2			
	2.110 $\pm$ 0.044	0.13	1.084 $\pm$ 0.03	(47)
	2.010 $\pm$ 0.060	0.04	1.080 $\pm$ 0.04	(50)
	2.025 $\pm$ 0.060	TH	-----	(54)
	1.815	0.3-1.5	-----	(55)
	2.175 $\pm$ 0.070	+1.9	-----	(56)
	2.120 $\pm$ 0.070	1.5	-----	(57)
	2.180 $\pm$ 0.060	1.9	-----	(57)
	2.280 $\pm$ 0.060	2.3	-----	(57)
	2.270 $\pm$ 0.100	4.0	-----	(57)
	2.530 $\pm$ 0.090	4.5	-----	(57)
	2.420 $\pm$ 0.100	5.0	-----	(57)
	2.420 $\pm$ 0.080	5.5	-----	(57)
	2.375 $\pm$ 0.120	14.0	1.144	(42)
PU-241	2.002 $\pm$ 0.051	TH	1.027	(10)
PU-242	1.810 $\pm$ 0.11	SP	-----	(54)
AM-242M	2.130 $\pm$ 0.050	TH	-----	(41)
CM-244	2.055 $\pm$ 0.060	SP	-----	(54)
	2.070 $\pm$ 0.060	TH	-----	(41)
	2.187 $\pm$ 0.093	SP	-----	(58)
CM-245	2.250 $\pm$ 0.070	TH	-----	(41)
CF-249	2.320 $\pm$ 0.060	TH	-----	(41)

TABLE 4, (CONT.)

ISOTOPE	E(AVE), MEV	E(IN), MEV	RATIO	REFERENCE
CF-252	2.220 $\pm$ 0.050	SP	-----	(41)
	2.350 $\pm$ 0.100	SP	-----	(12)
	2.348 $\pm$ 0.100	SP	-----	(28)
	2.155 $\pm$ 0.024	SP	1.102 $\pm$ 0.014	(27)
	2.130 $\pm$ 0.022	SP	1.054 $\pm$ 0.015	(27)
	2.080	SP	-----	(59)
	2.340 $\pm$ 0.050	SP	-----	(60)
	2.085 $\pm$ 0.060	SP	1.120 $\pm$ 0.050	(50)
	2.100	SP	-----	(61)
	2.220 $\pm$ 0.040	SP	-----	(41)

FOOTNOTES-

- A. AVERAGE FISSION NEUTRON ENERGY AS GIVEN OR DEDUCED FROM THE BASIC REFERENCE. ERRORS ARE OMITTED WHEN NOT DIRECTLY AVAILABLE.
- B. INCIDENT NEUTRON ENERGY. TH=THERMAL, FAST OR FAST CRIT.= FAST CRITICAL SPECTRUM, SP=SPONTANEOUS FISSION.
- C. RATIO $E(\text{AVE-X})/E(\text{AVE-235})$ IS GIVEN WHEN DIRECTLY MEASURED OR DEDUCED FROM SAME EXPERIMENTAL SET.

TABLE 5, U-235 AVE-ENERGY (THERMAL AND LOW ENERGY FISSION)

E-AVE (MEV)	ERROR	REF.
2.010	.060	40
1.956	.013	27
2.020	.025	27
2.110	.150	13
1.900	.200	44
2.000	.060	45*
2.060	.060	34*
1.946	.045	47
1.800	.100	48
a		
AVE (MEV) =	1.979201	RMS DEV. = .086094

Footnotes-

* Errors not available from the Ref. Indicated values are estimates by the present author.

a. Weighted average, weighting factor = 1/error.

TABLE 6. PU-239 AVE-ENERGY (THERMAL AND LOW ENERGY FISSION)

E-AVE (MEV)	ERROR	REF.
2.136	.024	27
2.075	.017	27
2.110	.044	47
2.025	.060	54
2.010	.060	50
a		
AVE (MEV) =	2.084073	RMS DEV. = .049855

Footnote-

a. Weighted average, weighting factor = 1/error

TABLE 7. RATIO PU-239/U-235 AVE-ENERGY(THERMAL AND LOW ENERGY FISSION)

E-AVE(MEV)	ERROR	REF.
1.092	.014	27
1.075	.020	13
1.084	.030	47
1.080	.040	50
AVE(MEV)= 1.084119 ^a RMS DEV.= .006369		

Footnote-

a. Weighted average, weighting factor = 1/error

TABLE 8. Cf-252 AVE-ENERGY (SPONTANEOUS FISSION)

E-AVE (MEV)	ERROR	REF.
2.348	.100	28
2.350	.100	12
2.155	.024	27
2.130	.022	27
2.080	.150	59*
2.340	.050	60
2.085	.060	50
2.100	.200	61
2.220	.040	30

AVE (MEV) = 2.188953^a RMS DEV. = .110669

Footnotes-

* Errors not available from the Ref. Indicated values
are estimates by the present author.

a. Weighted average, weighting factor = 1/error.

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