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*International Evaluation Co-operation*

## *VOLUME 9*

# FISSION NEUTRON SPECTRA OF URANIUM-235

*A report by the Working Party*
*on International Evaluation Co-operation*
*of the NEA Nuclear Science Committee*

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# FOREWORD

A Working Party on International Evaluation Co-operation was established under the sponsorship of the OECD/NEA Nuclear Science Committee (NSC) to promote the exchange of information on nuclear data evaluations, validation and related topics. Its aim is also to provide a framework for co-operative activities between members of the major nuclear data evaluation projects. This includes the possible exchange of scientists in order to encourage co-operation. The working party compiles requirements for experimental data resulting from this activity and determines common criteria for evaluated nuclear data files with a view to assessing and improving the quality and completeness of evaluated data.

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The following report has been prepared by Subgroup 9 which was set up in 1998 with the aim of investigating discrepancies found between microscopic and macroscopic data for the uranium-235 fission neutron spectrum. In addition, it was noted that the most recent evaluation of this spectrum had been performed in 1988 and had been based on only one experiment. It was thus felt necessary to review the existing evaluations, taking into account new experimental data and improved calculation methods.

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# SUMMARY

Subgroup 9 of the NEA Nuclear Science Committee Working Party on International Nuclear Data Evaluation Co-operation (WPEC) was formed to review and improve the existing prompt fission neutron spectra of $^{235}$U. The re-evaluation work included theoretical calculations of new spectra using the latest models and comparisons of these calculations with experimental data, both from differential and integral measurement.

A new prompt fission neutron spectrum matrix for the n + $^{235}$U system is proposed. It should be noted, however, that the thermal spectrum in this matrix is still preliminary. The need for a highly accurate measurement of this spectrum remains, as discrepancies in the existing database can only be solved through the application of such data.

# FISSION NEUTRON SPECTRA OF URANIUM-235

## 1. Work accomplished

Modern measurements of the differential spectrum, together with uncertainties, were collected for 12 different experiments by Kornilov and Staples. The experiments span an incident neutron energy range of thermal to 5.0 MeV, which means that no experimental measurements used to calculate the matrix include the physical effects of second- and third-chance fission. However, other experimental measurements on the compound fissioning nuclei occurring in second- and third-chance fission were used as constraints in the calculations.

A set of 30 integral cross-section measurements in the n(thermal) + $^{235}$U system were compiled by Mannhart following a careful analysis and evaluation of the existing experimental database. These provide integral tests of the prompt fission neutron spectrum for thermal neutron-induced fission under the assumption that the corresponding cross-sections for the specific reactions are known exactly. Mannhart also studied the various available cross-section evaluations for these reactions and chose, based upon his experience with $^{252}$Cf(sf) integral cross-section studies, the best evaluated data to use for comparisons of calculated and measured integral cross-sections here.

The Los Alamos model was then used to calculate a new prompt fission neutron spectrum matrix for the n + $^{235}$U system. Energy-dependent compound-nucleus formation cross-sections for the inverse process were used throughout. The matrix includes first-, second- and third-chance fission components and also includes the neutrons evaporated prior to fission in second- and third-chance fission. It has been calculated for 19 incident neutron energies ranging from 0.0 to 15.0 MeV. The model parameters used in the calculations were determined in least-squares adjustments to the measured differential spectra assembled by Kornilov and Staples.

Following the calculation of the new matrix, Kornilov provided an additional experimental measurement of the spectrum at 14.7 MeV by G.S. Boykov, *et al.* in 1991 [1], together with pertinent details about the measurement. At 14.7 MeV the total spectrum is made up of five components: three multiple-chance fission spectra (1st, 2nd, 3rd) and two evaporation spectra (n′,n′n″). A comparison of this experiment with the 14 and 15 MeV vectors of the new matrix is a test of its predictive power and the result gives good agreement (Figure 1). Note that the inclusion of pre-equilibrium effects at this incident neutron energy yield even better agreement (not shown here).

The new fission spectrum matrix for the n + $^{235}$U system is now complete except for two remaining points, the first of which *must* be addressed, and the second of which *should* be addressed:

- The measurements of the thermal-neutron-induced prompt fission spectrum are not in agreement in the peak region (Figure 2) or in the tail region. This means that the calculated thermal spectrum depends upon which measurement, or measurements, is used to determine the model parameters for this case. Four candidate thermal spectra have therefore been calculated for testing against the set of 30 integral cross-section measurements. The candidate spectra were then used by Mannhart to calculate the 30 integral cross-sections for each of the candidates. His results have been studied and yield the following conclusions:

    - The thermal spectrum determined by least-squares adjustment (Figure 3) to the differential measurement of Starostov, *et al*. [2] reproduces the integral cross-section measurements to within 10% (C/E = 1.0 ± 0.1) out to approximately 8 MeV, but fails beyond, falling to C/E values near 0.6 (Figure 4).

    - The thermal spectrum determined by least-squares adjustment (Figure 5) to the differential measurement of Wang, *et al*. [3] reproduces the integral cross-section measurements to within 10% out to approximately 11 MeV, but fails beyond, falling to C/E values just under 0.8 (Figure 4).

    - The ENDF/B-VI thermal spectrum, calculated using the Los Alamos model earlier, reproduces the integral cross-sections to within 10% out to approximately 13 MeV, but fails beyond, falling to C/E values near 0.85 (Figure 4), and is somewhat hard in the tail region in comparison with the two modern differential measurements. Here, the downward trend in C/E values begins at just under 10 MeV.

    - Increasing the average nuclear level density parameter used to calculate the ENDF/B-VI thermal spectrum by 1.8% results in a thermal spectrum that reproduces the integral cross-section measurements to within 10% over their entire range (Figure 4). This spectrum, however, is much too hard in the tail region in comparison to both the Starostov, *et al*. and Wang, *et al*. differential experimental thermal spectra.

Therefore, no calculated thermal spectrum has been found that simultaneously reproduces either of the two modern thermal differential measurements [2,3] and the set (30) of measured integral cross-sections to within an acceptable level. Given the importance of the thermal spectrum this contradiction *must be resolved*.

- The question of the influence of possible scission neutrons on the prompt fission neutron spectrum has been studied by Kornilov and Hambsch for the spontaneous fission of $^{252}$Cf. Further studies were performed by Kornilov using the experimental data on n(thermal) + $^{235}$U measured by Skarsvag [4]. Another study on the same thermal system was performed by Samant, *et al*. [5]. These studies all indicate about a 10% effect. However, Skarsvag in the analysis of his own experiment finds "no convincing evidence for scission neutrons..." and this same conclusion has been reached in other experiments as well. Nevertheless, sufficient analysis exists at this time to conclude that new experimental work on the scission neutron question should be pursued, together with corresponding theoretical studies. This issue *should be resolved* as it may shed light on the thermal spectrum problem.

## 2. Possible sources of the thermal spectrum problem

1) The two differing thermal spectrum measurements?

2) The set of measured integral cross-sections?

3) The set of pointwise threshold reaction cross-sections used in the corresponding set of calculated integral cross-sections?

4) The Los Alamos model?

Our assessment is that items 2) and 3) are, for the most part, satisfactory. That is, if one has the physically correct thermal spectrum, the ratios of the calculated to measured integral cross-sections, plotted against a suitable measure of the reaction threshold [Mannhart's E(50%) point], should not depart from unity by more than 10%. In particular, a monotonic departure from unity as a function of E(50%) that exceeds 10% should not occur. Therefore, items 1) and/or 4) are more likely to be suspect.

### *2.1 Experiment and the thermal spectrum problem*

The most certain way to address item 1) is to perform a new and highly accurate measurement of the thermal spectrum. Ideally, the energy range of the measured spectrum would be about 100 keV to 15 MeV. Clearly, good statistics and an exhaustive treatment of systematic error are essential. Great confidence in the measurement would result if the same experimental team were to perform the identical measurement at two different, but well understood, thermal neutron facilities and obtain the same result at each. Josch Hambsch has some initial ideas on such a measurement.

### *2.2 New work on the Los Alamos model and the thermal spectrum problem*

The best way to address item 4) is to examine all of the physics content of the existing Los Alamos model, keeping in mind the successes of that model in calculating and predicting fission neutron spectra for a variety of fissioning systems for a range of excitation energies (incident neutron energies). For example, the prompt fission neutron spectrum matrix for the n + $^{235}$U system in ENDF/B-VI, calculated using the Los Alamos model, was based upon only one measurement of the differential spectrum, that of Johansson and Holmqvist [6] at 0.53 MeV incident neutron energy, together with the existing experimental database of $\bar{\nu}$ values ranging from thermal energy to 15 MeV. This matrix has performed very well in comparisons with measurements at other incident energies and in a number of different applications. Similar results have been obtained in calculating the n + $^{239}$Pu and n + $^{238}$U matrices with the Los Alamos model (summary article on the three matrices in preparation). At this time, nevertheless, two physics issues in the Los Alamos model have been re-examined and a new physics issue has been introduced:

- The shape of the prompt fission neutron spectrum depends partly upon the energy dependence of the process inverse to neutron emission, namely, compound nucleus formation. The energy-dependent compound nucleus formation cross-section is calculated with a physically realistic optical potential. Once the potential is chosen, however, for low-energy neutrons the question arises as to whether width fluctuation corrections would affect the energy dependence of the sum over all open channels, that is, the compound nucleus formation cross-section. The answer, due to flux conservation (unitarity), is no. Therefore, this consideration has no obvious impact upon the thermal spectrum problem.

- The Los Alamos model calculates the contributions to the prompt fission neutron spectrum separately from each fragment mass peak and then adds them together to obtain the total spectrum. The correct weights for this addition are constructed from the average prompt neutron multiplicities due to the separate fragment mass peaks, namely, $\overline{\nu}_L$ (light mass peak) and $\overline{\nu}_H$ (heavy mass peak), leading to weights $\overline{\nu}_L / (\overline{\nu}_L + \overline{\nu}_H)$ and $\overline{\nu}_H / (\overline{\nu}_L + \overline{\nu}_H)$ for the light and heavy fragment peaks, respectively. These quantities are very difficult to measure, but are reasonably well determined for $^{252}$Cf(sf). They are less well determined for neutron-induced fission and, as a consequence, the default assumption of equal values is typically used. This is the case for the present n + $^{235}$U fission spectrum matrix calculated with the Los Alamos model. However, a recent measurement by Nishio, *et al*. [7] for the n(thermal) + $^{235}$U system yields $\overline{\nu}_L$ = 1.42 and $\overline{\nu}_H$ = 1.01. This means that the weight on the spectrum contribution from the light fragment mass peak vs. that from the heavy fragment mass peak is 58.4% vs. 41.6%. This impacts the shape of the total fission neutron spectrum because the tail region of the contribution from the light peak is harder than that from the heavy peak, due to the fact that the light peak fragments move with much higher kinetic energy than those from the heavy peak. So if the light peak contribution is weighted more than the heavy peak contribution, then the total fission spectrum becomes harder in the tail region. This is exactly the type of change needed to reproduce the higher threshold integral cross-sections*.

The thermal spectrum was re-calculated with the Los Alamos model using the weights inferred from the Nishio, *et al*. experiment yielding the following results:

- This spectrum reproduces the integral cross-section measurements to within 10% out to approximately 13 MeV, where the downward trend begins falling to C/E values near 0.9 (Figure 4), which is an improvement over the ENDF/B-VI spectrum discussed above.

- Use of these same weights improves the agreement with differential spectrum measurements for incident neutron energies of 0.4 and 0.5 MeV.

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* *Note: Calculations have been performed by others using the Los Alamos model where the fission-fragment mass distributions have been deconvoluted into an odd number of Gaussian distributions and separate spectrum contributions have been calculated for each Gaussian. This approach introduces more parameters into the model calculation and therefore more degrees of freedom to utilise in fitting a given experimental measurement. However, the theoretical basis justifying such a procedure has yet to be demonstrated on fundamental grounds.*

  - The agreement with the two differential thermal measurements of Starostov, *et al*. and Wang, *et al*., however, is worse than the earlier results using equal weights.

  - Therefore, the thermal spectrum problem has yet to be resolved.

- Plans are underway to develop a scission neutron emission mechanism to install into the Los Alamos model. At this point, however, it is not possible to estimate when this might be accomplished because the emission mechanism needs to be developed, quantified and tested.

## 3. Conclusions

A new prompt fission neutron spectrum matrix exists for the n + $^{235}$U system. It contains spectra for 19 incident neutron energies ranging from thermal to 15 MeV. It is based upon 12 differential and 30 integral measurements of the spectrum and the Los Alamos model for the spectrum. Initial tests of the predictive power of this matrix yield good results. The matrix is available at http://t2.lanl.gov/data/fspect.html.

The thermal spectrum in this matrix is temporary and is to be replaced. It has been calculated using weights for the light and heavy fission fragment mass peak contributions that are inferred from the Nishio, *et al*. experiment. The temporary thermal spectrum reproduces the measured integral cross-sections reasonably well, but not ideally, and it is too hard in the tail region in comparison to the two modern measured thermal spectra used in this study. The temporary spectrum is a compromise.

The two modern measured thermal spectra used in this study, that of Starostov, *et al*. [2] and that of Wang, *et al*. [3] are incompatible with each other and are both incompatible with the set of 30 integral cross-section measurements used in this study.

Therefore, a new and highly accurate measurement of the prompt fission neutron spectrum for the n(thermal) + $^{235}$U system should be undertaken as soon as possible. This measurement is the best – and perhaps only – way to resolve the existing discrepancies in the thermal spectrum.

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# FIGURES

**Figure 1. Los Alamos model prediction near 14.7 MeV**

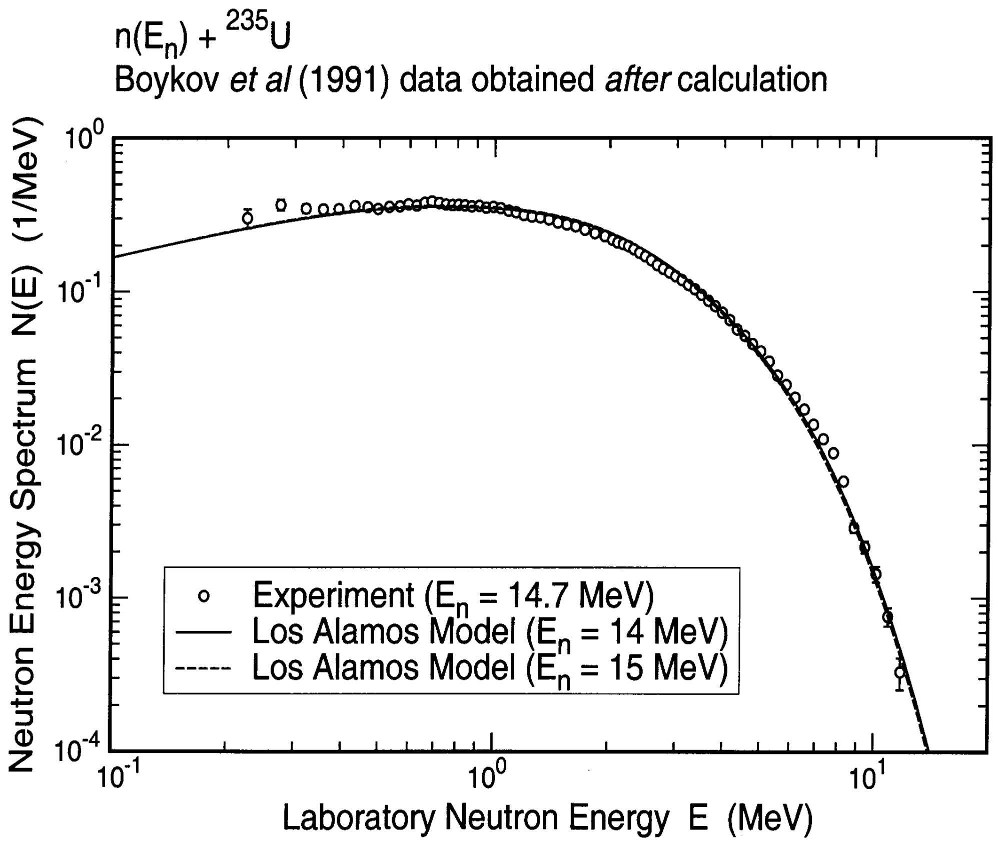


**Figure 2. Comparison of experimental thermal spectra in the peak region**

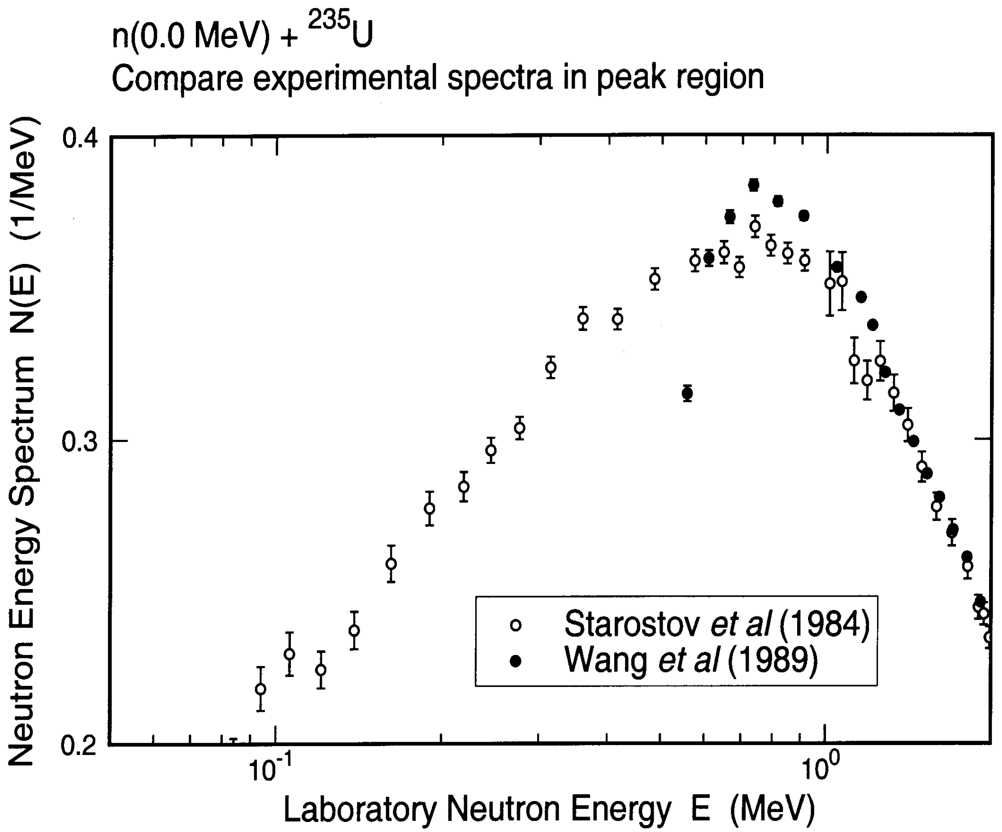


**Figure 3. Least-squares adjustment of the Los Alamos model to the experiment of Starostov, *et al*. [2]. The ENDF/B-VI spectrum is an earlier Los Alamos model calculation.**

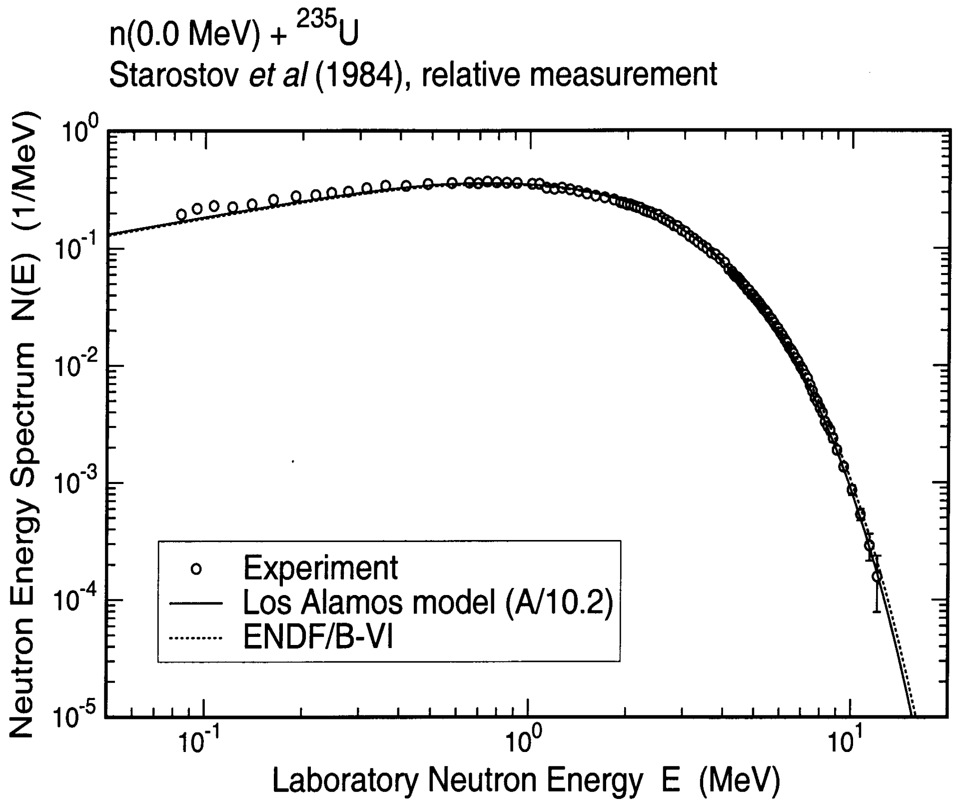


Figure 4. Ratios of calculated to experimental integral cross-sections, shown without error bars (for clarity) and joined by straight-line segments. The calculated integral cross-sections all use a Los Alamos model thermal spectrum determined as per the constraint identified in the legend.

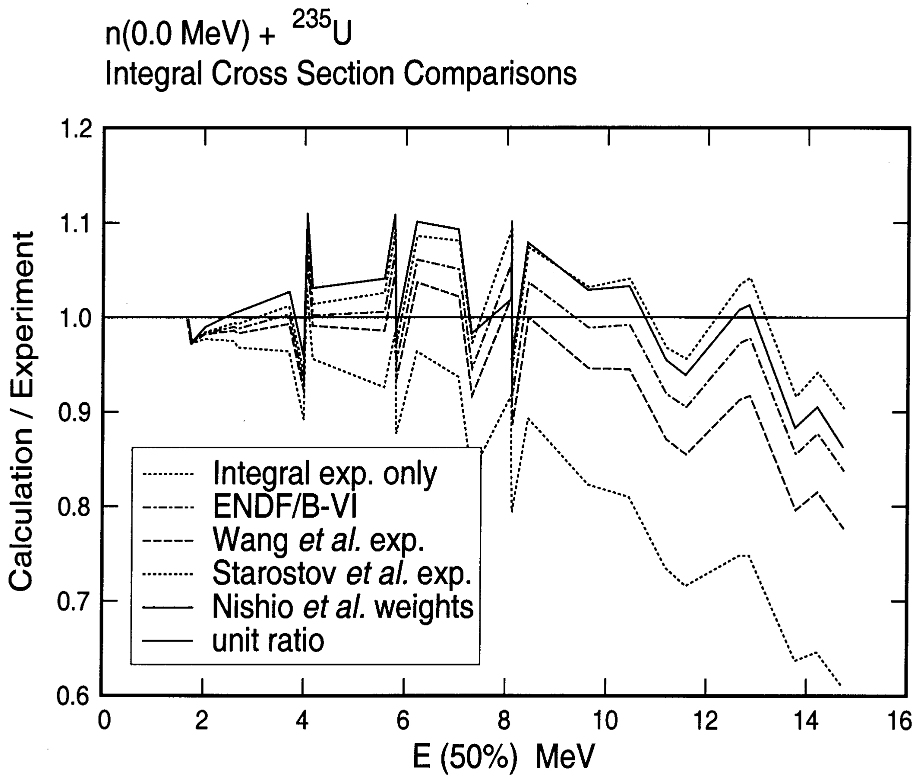
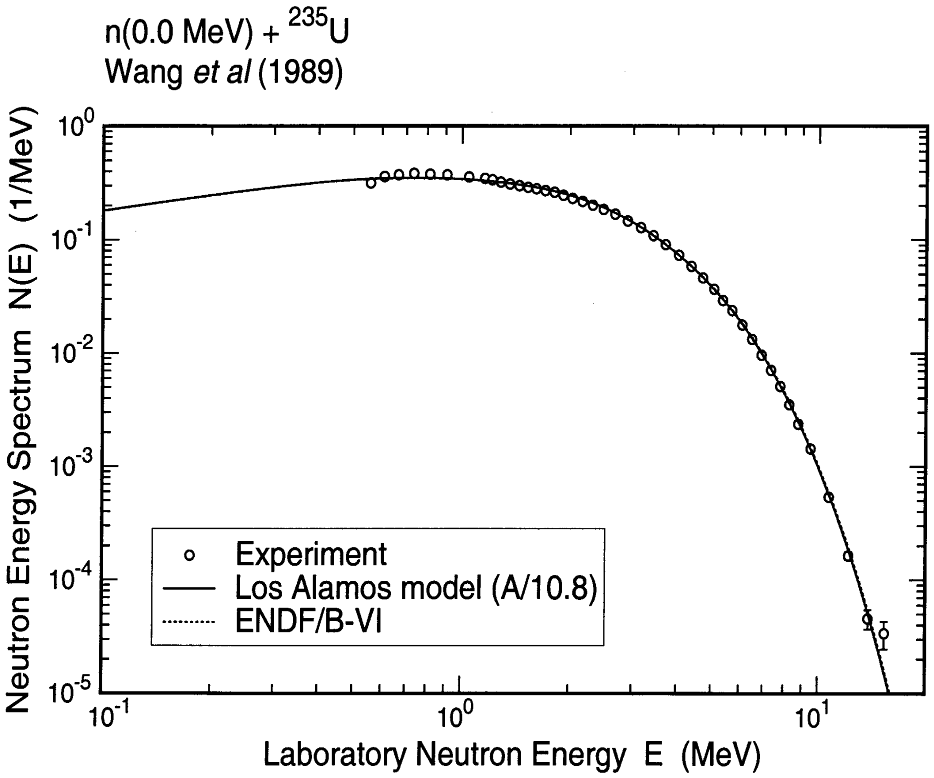


Figure 5. **Least-squares adjustment of the Los Alamos model to the experiment of Wang, *et al*. [3]. The ENDF/B-VI spectrum is an earlier Los Alamos model calculation.**



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