CONF-850507--30

RECEIVED BY OSTI JUN 07 1985

LA-UR--85-1674

DE85 012699

TITLE: STATUS OF FAST NEUTRON NUCLEAR DATA FOR ^{239}Pu

AUTHOR(S): Edward D. Arthur, T-2

SUBMITTED TO: THE INTERNATIONAL CONFERENCE ON NUCLEAR DATA FOR BASIC AND APPLIED SCIENCE, SANTA FE, N.M., MAY 13-17, 1985

STATUS OF FAST NEUTRON NUCLEAR DATA FOR ^{239}Pu

EDWARD D. ARTHUR
Los Alamos National Laboratory, Los Alamos, N.M., U.S.A.

Abstract Recent energy-averaged data (fission, total cross section, scattering and $\bar{\nu}_p$) describing the interaction of fast neutrons with ^{239}Pu are discussed, with emphasis on neutron energies higher than the inelastic scattering threshold. The linkage between such experimental data and new theoretical calculations is described in context of their influence on a new revision to the ENDF/B-V ^{239}Pu evaluation. Results from this new revision are are compared with other current ^{239}Pu evaluations. Incorporation of such information pertaining to neutron emission and $\bar{\nu}_p$ in the new ENDF revision had a profound impact on calculations performed for fast critical assemblies. Such calculational results are presented with emphasis on systematic improvements achieved in eigenvalues and fission ratios.

INTRODUCTION

The accuracy requirements of nuclear data for the major fissile nuclei and their impact on reactor design have been the subject of several recent reviews.[1-3] Within the context of fast breeder applications, ^{239}Pu nuclear data plays a dominant role and affects not only reactor criticality, but breeding ratios as well. This paper will review the present state of knowledge concerning neutron-induced data for ^{239}Pu for fast neutrons, i.e., at energies above the resonance region where cross sections exhibit a smooth variation with energy. Because of space limitations, we will primarily discuss $\sigma(n,f)$, scattering, prompt fission yield $\bar{\nu}_p$ and prompt fission neutron spectra. Recent data (generally since 1978) will be emphasized along with results from new ^{239}Pu evaluations. Of particular importance is the linkage between new experimental results and theoretical model calculations of them. As recently applied, these sources of information have complemented each other, thereby strengthening results obtained for evaluated data. Finally, new information, particularly that related to neutron emission, has impacted significantly the ability to calculate critical assembly parameters in a more consistent fashion. Results of such calculations will be presented along with observations of the role of critical assembly information in overall data testing.

FISSION CROSS SECTIONS

Much of the recent data applicable to $^{239}Pu(n,f)$ comes from measurements relative to $^{235}U(n,f)$. We consider recent measurements[4-8] that generally cover a wide energy region from low energies up to 10 or 20 MeV. Such results are presented in Fig. 1. Experimental errors have not been shown to maintain clarity in the figures but are usually in the range of 1.5-2.5% for the measurements of Refs. 4, 6, 7, and 8. Some comments are in order. It has been noted[1,9] that the energy scale of the Kari measurements appears in error, relative to the majority of the other measurements. In a recent Livermore evaluation[9] of the $^{239}Pu(n,f)/\ ^{235}U(n,f)$ ratio, the energy scale of the Kari measurements was shifted by 8 ns in neutron time of flight. This adjustment, though large, does bring the Kari data into good agreement with the Carlson,[4] Meadows,[7] and Weston[8] results in the energy region around 6 MeV where a sharp descent occurs in the fission ratio. The data presented in Fig. 1 reflect this adjustment. One finds that below 1.6 MeV the data of Carlson,[4] Kari,[5] Fursov,[6] Meadows,[7] and Weston[8] generally agree to within ± 1% of each other. The Kari[5] data have a lower energy limit of 0.5 MeV, exhibit significant statistical scatter, and are less reliable in this energy region. Between 1.6 and 10 MeV the Meadows and Fursov results lie systematically higher (by about 4%) than the Weston, Carlson, and adjusted Kari data. Again, the Weston and Carlson data also agree with each other to within 2-3%. Above 10 MeV there are three data sets (Weston, Carlson, Kari) that cover this energy range. These agree to better than 3% up to 15 MeV. Shown by the curves are evaluated ratios from a recent Livermore evaluation,[9] that of Antsipov[10] and the ENDF/B-V evaluation.[11] The Livermore and Antsipov evaluations exhibit a lower limit for smooth cross-section representation of 0.1 MeV, while the ENDF/B-V curve extends down to 0.025 MeV. While all three evaluations agree with each other to within approximately 2% between 0.15 and 2 MeV, the ENDF/B-V results lie significantly higher (up to 20-25%) than the data at energies between 0.025 and 0.15 MeV. For energies between 2 and 6 MeV, the Antsipov evaluation follows the Fursov data and thus is 3-4% higher. It also exhibits a shape difference in the energy region between 5 and 6 MeV. For energies between 7 and 13 MeV the evaluated data sets agree to within 3-4% with the LLNL values being lower but in better agreement with the Carlson, Weston, and Kari results. Around and above 14 MeV, the LLNL and Antsipov evaluations are in good agreement with data and each other. In contrast, the ENDF/B-V evaluation diverges significantly and its shape is not supported by any recent measurements.

In addition to these ratio results, recent absolute measurements of $^{239}Pu(n,f)$ cross sections have been made by Kari,[5] Xianjian,[12] Arlt,[13,14] Cance,[15] Mahdavi,[16] Li Jingwen,[17] and Adamov.[18] These are shown in Fig. 2. At energies between 1 and

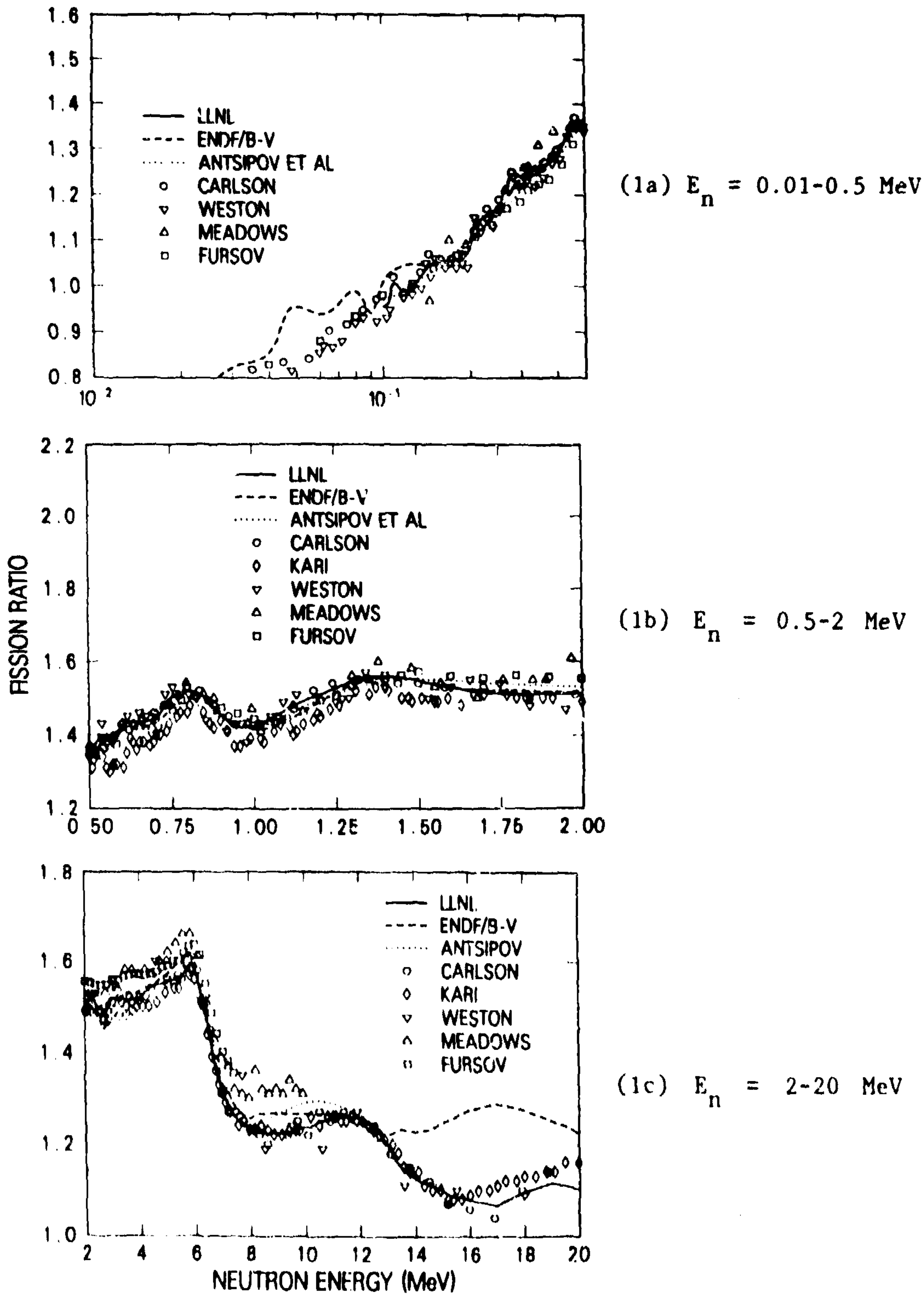

FIGURE 1 Experimental[4-8] and evaluated[9-11] data for the $^{239}Pu(n,f)$ to $^{235}U(n,f)$ ratio.

6 MeV these absolute data and the evaluated results agree reasonably well (to within 4%), although the new Livermore evaluation exhibits apparently artificial structure. Likewise, at the onset of second chance fission, the agreement between the three evaluations and the Kari data is good. Between 8.5 and 13 MeV all three evaluations agree with this data, although the Antsipov curve intersects the upper portion of the error bars, while the Livermore results pass through the lower bounds. This translates into a 6.5% difference in evaluated values. Around 14 MeV there is a considerable spread in absolute measurement results that is well outside the errors quoted. Differences of up to 10% occur. Only the Cance, Arlt, and portions of the Kari results agree reasonably with the TOF measurements described earlier.

In Ref. 19, Patrick compared absolute ^{239}Pu data[20-21] with (n,f) values obtained from the Carlson ratio data. In the range between 0.1 and 1 MeV, there was good agreement, a conclusion which still stands. However, the ratio data do diverge with the Meadows and Fursov results being higher in the region between 2 and 6 MeV. The absolute data of Kari agree with (n,f) values based on these ratios, while the Xianjian results lie slightly higher (~ 2.5%). At energies between 8.5 and 12.5 MeV (at the second chance fission plateau), the Kari data lie about 3% higher

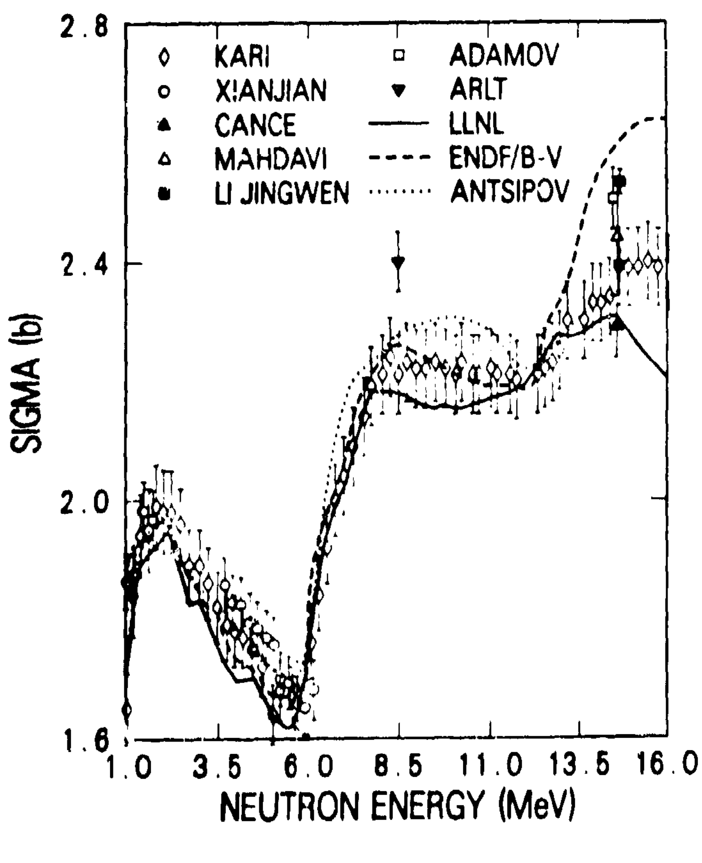

FIGURE 2 Recent absolute measurements of $^{239}Pu(n,f)$ are compared with evaluated results.[9-11]

than values deduced via consideration of the Weston and Carlson ratio data. Finally, as indicated earlier, the existence of the large spread in 14-MeV absolute measurement data is at odds with the precision quoted for these measurements. It is also at odds with the fact that for several of them,[15-18] the agreement obtained for 14 MeV $^{235}U(n,f)$ was on the order of 1%.

PROMPT FISSION NEUTRON EMISSION

Two recent experimental results have served to clarify $\bar{\nu}_p$ information for ^{239}Pu. These are the measurements by Gwin et al.[22] and the 1980 revised $\bar{\nu}_p$ values of Frehaut.[23] Note that the earlier Frehaut measurements had been adoped in ENDF/B-V as the basis for $\bar{\nu}_p$ in the MeV energy region. Figure 3 illustrates results from these new measurements as well as the ENDF/B-V evaluation and its recent revision, REV2.[24] The REV2 values are based on the new data sets and deviate from ENDF/B-V by up to 1-3% at energies above 1.5-2 MeV. The REV2 results agree with older measured data[25-27] and with the new Antsipov[10] and Howerton and White[9] evaluations, with the spread in recommended $\bar{\nu}_p$ values being less than 1%. The differences in $\bar{\nu}_p$ values (with respect to ENDF/B-V) occur primarily for neutron energies above 1.25 MeV, and therefore may not significantly affect fast breeder reactor performance parameters.[1] However, they do significantly affect calculations made for fast critical assemblies, especially when used with new results for inelastic scattering data.

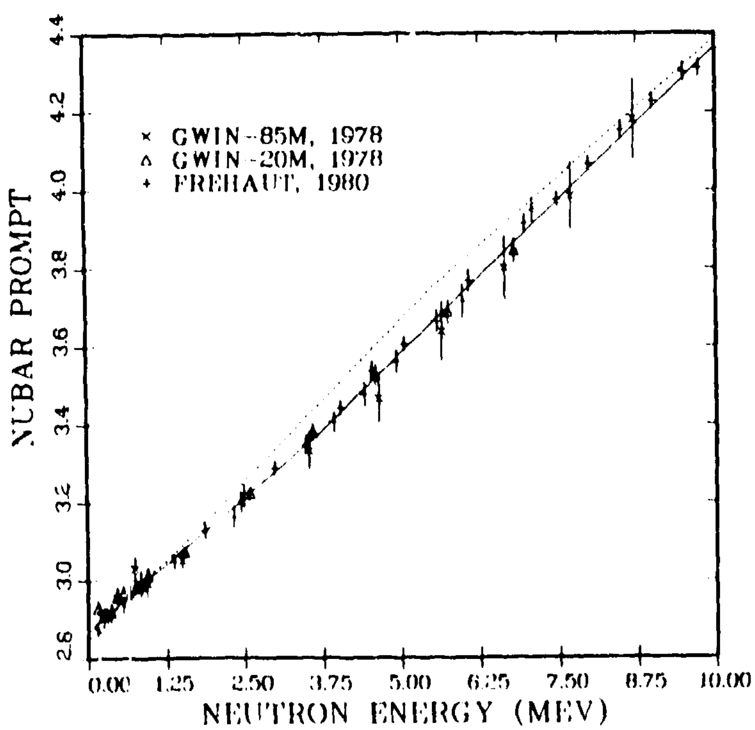

FIGURE 3 The $\bar{\nu}_p$ values of Gwinn[22] and Frehaut[23] are compared with ENDF/B-V[11] (dashed curve) and a new revision[24] to it, REV2 (solid curve).

TOTAL, ELASTIC, AND INELASTIC CROSS SECTIONS

These are areas where recent progress has been made in obtaining more accurate data or in relieving a situation where there was previously a paucity of data. Figure 4 illustrates results of new total cross-section measurements made by Poenitz et al.[28] and compares them with the three evaluated sets discussed earlier. The REV2 and Antsipov evaluations agree well with the Poenitz measurements but the ENDF/B-V evaluation lies significantly below them. Such data differences affect calculation of smaller critical assemblies where neutron leakage effects can be important.

There has been a relative lack of experimental data pertaining to ^{239}Pu elastic and nelastic scattering processes. This occurs because of its ground state band structure where close spacings exist between the ground and low-lying excited states. Data are also lacking for scattering from higher states that are members of higher-lying vibrational bands. However, data for elastic and inelastic scattering processes have improved significantly because of recent measurements of Haouat.[29] These results (along with their measurements of other actinide nuclei) have prompted theoretical analyses of neutron scattering from the ground-state rotational band in ^{239}Pu. Another series of measurements by Smith[30] have provided complementary information that relate

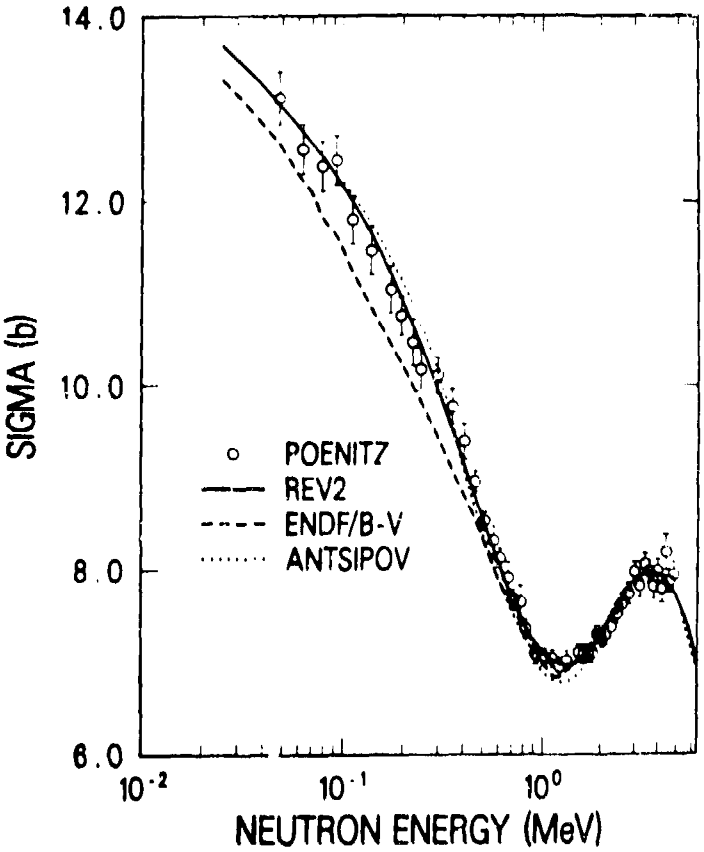

FIGURE 4 The ^{239}Pu total cross measurements of Poenitz[28] are compared with evaluated results from REV2[24] (solid curve), ENDF/B-V[11] (dashed curve) and Antispov[10] (dotted curve).

directly to determination of the total inelastic scattering cross section. These experiments measured precisely the cross section for elastic scattering along with contributions due to inelastic scattering from low-lying states that could not be resolved by the experimental apparatus. These data were then subtracted from total, fission, and capture cross sections to determine a total inelastic cross section for excitation of levels above the threshold set by the experimental resolution. Such inelastic results appear in Fig. 5 where they are compared with the three evaluations discussed earlier. Again, there is good agreement between the REV2, Antsipov results and the data, while ENDF/B-V deviates significantly. Both of the newer evaluations utilized the deformed optical model via coupled-channel calculations to account for direct-interaction (DI) cross section components, which should be significant at neutron energies >1-2 MeV. In contrast, such effects were neglected in ENDF/B-V.

Although there are no direct measurements for inelastic scattering cross sections for higher-lying ^{239}Pu levels, there exists indirect evidence that current theoretical methods are adequate to describe such processes. Such data are of concern, since substantial scattering from higher-lying states could significantly affect the spectrum of downscattered neutrons. However, new measurements have been made of ^{238}U(n,n') scattering from levels occupying higher-lying vibrational bands.[31] These data are reproduced well by calculations[32] employing the Hauser-Feshbach model with small amounts of DI components determined using results from charged-particle inelastic scattering. Thus, current theoretical techniques are probably sufficient to deal with the similar problem in ^{239}Pu.

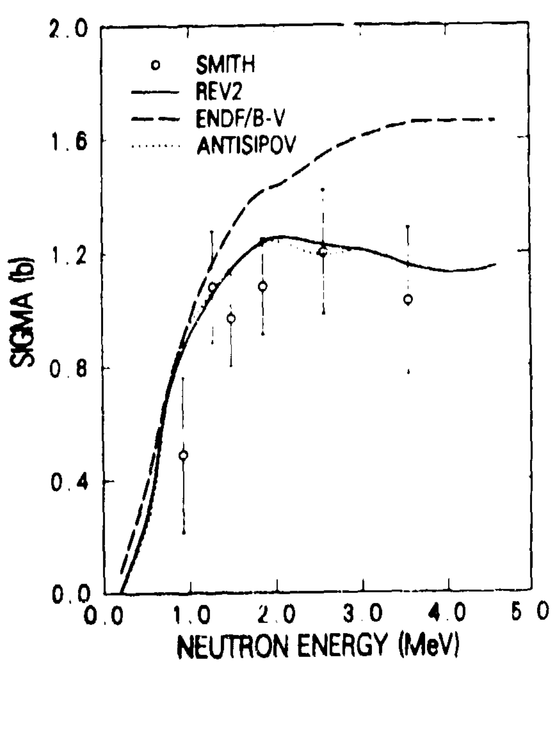

FIGURE 5 Total inelastic scattering data determined by Smith[30] are compared with REV2,[24] ENDF/B-V,[11] and the the Antsipov[10] evaluated results (solid, dashed, and dotted curves).

The impact that the measurements and calculations discussed here have on evaluated data libraries is illustrated in Fig. 6. Here cross-section excitation functions for scattering from low-lying states in ^{239}Pu resulting from ENDF/B-V, the REV2 update, and the Antsipov evaluations are compared. The REV2 and Antsipov results agree well, but the ENDF/B-V values disagree significantly, especially where shapes are concerned. This again occurs because of the neglect of direct-interaction effects in ENDF/B-V. These differences markedly affect the total neutron emission spectrum as well as comparisons of calculated and experimental critical assembly data.

PROMPT FISSION NEUTRONS AND TOTAL NEUTRON EMISSION SPECTRA

In 1982 Madland and Nix published an improved theoretical treatment[33] for determination of the neutron energy spectrum from fission. This approach allows use of physics information other than direct measurements to determine spectra, and produces

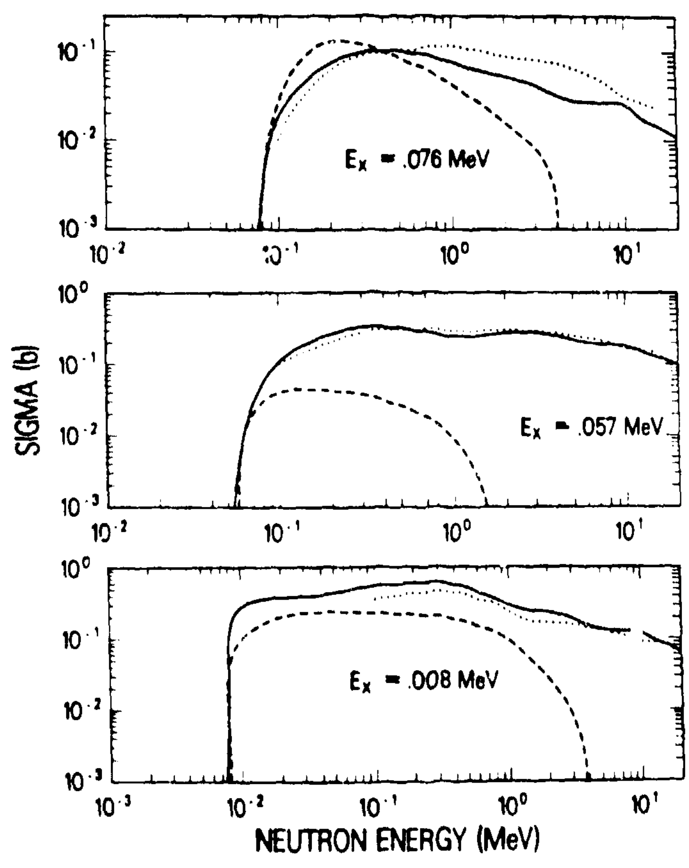

FIGURE 6 Excitation functions for inelastic scattering from the three lowest states in ^{239}Pu are compared for REV2, ENDF/B-V, and the Antsipov results (solid, dashed, dotted curves, respectively).

results that differ significantly from the Maxwellian and Watt shapes generally used in data evaluation. Differential and integral comparisons[34,35] illustrate its superior capabilities in reproducing experimental results.[36,37] Its firm theoretical basis provides a sound method to provide realistic data where experimental results are not available.

This theoretical approach was applied to determination of the prompt fission neutron spectra for REV2 and generally produced higher average emission energies. Both the changes in the shape of the prompt fission neutron spectrum and those resulting from improvement of inelastic scattering data combine together to produce significant changes in evaluated total neutron emission spectra. Figure 7 shows an example in which the total emission spectrum produced by 2-MeV neutrons on ^{239}Pu is compared for ENDF/B-V and its revision, REV2. Both the inelastic scattering changes and the utilization of the Madland-Nix theory for fission neutrons produce a harder emission spectrum, as compared with ENDF/B-V. The next section discusses the significance of these effects on calculated fast critical assembly data.

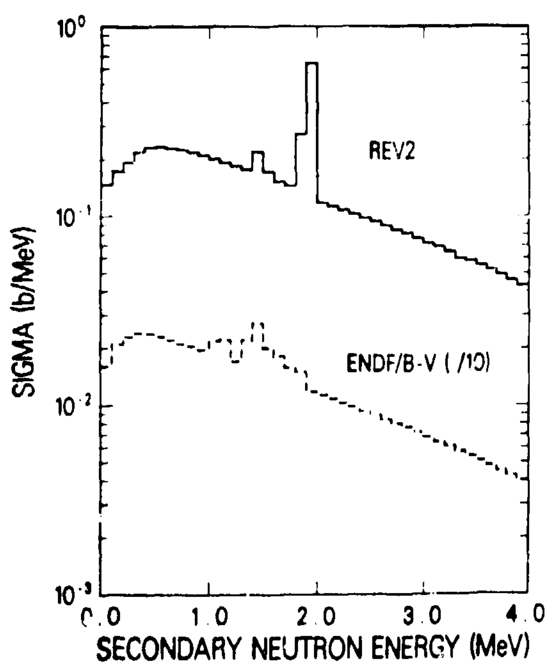

FIGURE 7 A comparison of the total neutron emission spectrum induced by 2-MeV neutrons on ^{239}Pu. The solid histogram results from REV2 and the dashed from ENDF/B-V (divided by 10). The elastic scattering peaks have been removed for clarity.

INTEGRAL TESTING WITH FAST CRITICAL ASSEMBLIES

The revision to the ^{239}Pu data files incorporated significant changes in total and scattering cross sections brought about by improved experimental results and advanced theoretical models. Likewise, a significant decrease in $\bar{\nu}_p$ at MeV energies occurred as a result of the analysis of new experimental results. An independent validation of these evaluated data came through calculation of eigenvalues (k_{eff}) and ratios (C/E values) for several cross-section types measured in a number of fast critical assemblies. Calculations were made for the JEZEBEL, ZPR-6/7, JEZEBEL-PU, FLATTOP-PU, and THOR assemblies.[38 39] Eigenvalue and C/E results are shown in Fig. 8. The solid circles reflect results obtained using REV2, while the triangles are results obtained with ENDF/B-V. Generally all calculated values based on the revised data show marked improvements in agreement with experiment compared with the ENDF/B-V results. This is particularly true for C/E values for threshold fission that are sensitive to the hardening of emission spectra. Also, eigenvalues for these assemblies average around unity with a significant reduction in their scatter.

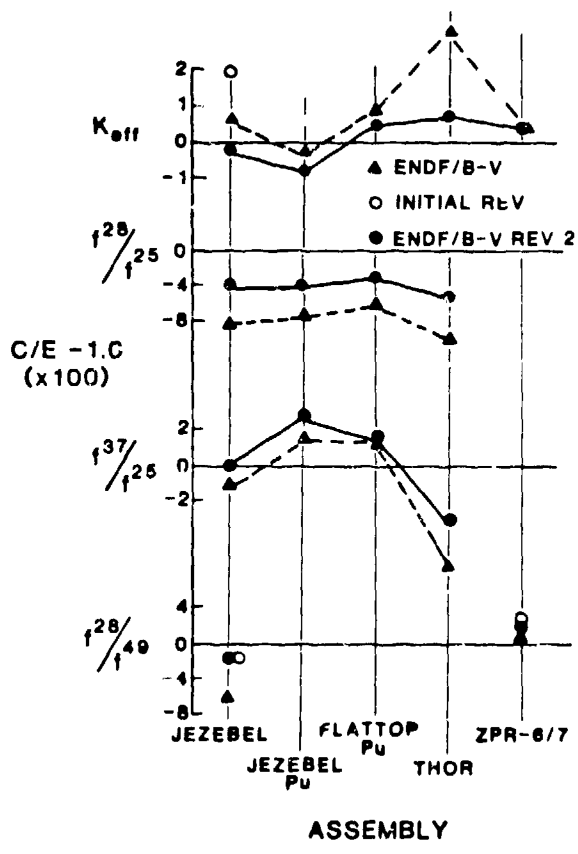

FIGURE 8 A comparison of k_{eff} and several C/E values obtained using ENDF/B-V (triangles) and the revision to it, REV2 (closed circles) for several critical assemblies. The open circles represent a special case encountered during the process of revision that is also described in the text.

The results shown by the open circles point to an interesting problem encountered in implementing the data changes discussed. These calculated results were obtained after only improvement to the inelastic scattering and total cross sections were implemented. (No $\bar{\nu}_p$ changes had been made at this point.) While improvements in some C/E values did occur, the JEZEBEL eigenvalue deviated unacceptably from experimental results. This example illustrates that isolated changes for one class of parameters cannot be made without a similar re-evaluation of other pertinent data. It also illustrates the value of critical assemblies for consistent testing of cross section values.

SUMMARY AND CONCLUSIONS

The review of recent (n,f) experimental data shows good agreement among several ratio measurements for neutron energies below 1.5 MeV. This agreement is on the order of 2% and extends to absolute data as well, based on the results of Ref. 19. At higher energies (2-6 MeV), ratio data exhibit divergences up to 3-4%. However, the ratio data of Meadows and Fursov that appear to be high are in reasonable agreement with absolute (n,f) data. Around 14 MeV, absolute data (most measured recently) exhibit considerable (and excessive) scatter. New evaluations are based primarily on the recent experimental data described previously and show good agreement with them. The older ENDF/B-V evaluation, however, exhibits significant discrepancies for neutron energies between 0.025 and 0.15 MeV and at energies above 14 MeV.

Data for $\bar{\nu}_p$ have improved because of results from the Gwin measurements and the updated data of Frehaut. Agreement to within 1% exists for these data sets. The new evaluations show a similar agreement with only ENDF/B-V having a serious discrepancy (up to 3%) at energies above 1.25 MeV.

The areas of neutron scattering and neutron emission spectra have improved significantly because of the availability of new experimental data (particularly for elastic and inelastic scattering) and the utilization of advanced theoretical models. The new experimental data (even though somewhat restricted in nature) have provided necessary information to appropriate models, which can be applied to produce significant improvement in evaluated scattering data. New models (i.e., Madland and Nix) have been developed to address the problem of prompt fission neutron emission and were applied to improve evaluated data for ^{239}Pu fission spectra. Measurements of the total neutron emission spectra at a few incident energies would be very desirable to provide further tests of these models.

Finally, the utilization of critical assembly data has illustrated the validity of many of the differential data improvements discussed in this paper. As illustrated by the effect on JEZEBEL when only inelastic data improvements were made, such integral assembly results provide an essential method for testing the overall consistency of microscopic data improvements.

In summary, recent trends in data evaluation have utilized information from three key components--new differential measurements, theoretical model calculations, and integral experimental results. These three sources provide complementary methods for improvement of ^{239}Pu fast neutron data in the consistent fashion required for practical applications.

REFERENCES

1. W. P. Poenitz, G. de Saussure, Prog. in Nucl. Energy 13, 129 (1984).
2. C. R. Weisbin et al., Ann. Nucl Energy 9, 615 (1983).
3. J. L. Rowlands, Proc Conf. Neutron Phys. and Nucl. Data, Harwell, p. 7 (1978).
4. G. W. Carlson, J. W. Behrens, Nucl. Sci. Eng. 66, 205 (1978).
5. K. Kari, Karlsruhe report KFK 2673 (1978).
6. B. I. Fursov et al., Atomnaya Energiya 43, 261 (1977).
7. J. W. Meadows, Argonne report ANL/NDM-83 (1983).
8. L. W. Weston and J. H. Todd, Nucl. Sci. Eng. 84, 248 (1983).
9. R. J. Howerton and R. M. White, Lawrence Livermore National Laboratory report UCID-19973 (1984).
10. G. V. Antsipov et al., IAEA report INDC (CCP)-166/GHJ (1981).
11. E. Kujawski, General Electric report GEFR-00247 (1977).
12. Zhou Xianjian et al., Proc. Int. Conf. Nucl. Data Sci. and Tech., Antwerp (D. Reidel, Dordrech, Holland), p. 36 (1982).
13. R. Arlt et al, Kernenergie 24, 48 (1979).
14. R. Arlt et al., Sixth All Union Conf. on Neutron Physics, Kiev, Vol. 4, p. 129 (1983).
15. M. Cancé and G. Grenier, Nucl. Sci. Eng. 68, 197 (1978).
16. M. Maldavi and G. F. Knoll, Ibid. Ref. 12, p. 58.
17. Li Jingwen et al., Ibid. Ref. 12, p. 55.
18. V. M. Adamov et al., Proc. Conf. on Nucl. Cross Sections and Tech., Knoxville, NBS Spec. Pub. 594, p. 995 (1979).
19. B. H. Patrick, Ibid. Ref. 3, p. 76.
20. G. F. Knoll, Proc. Symp. Neutron Standards and Applications, NBS Spec. Publ. 493, p. 304 (1977).
21. I. Szabo and J. P. Marquette, Proc. NEANDC/NEACRP Specialists Meet. on Fast Neutron Fission Cross Sections, Argonne report ANL-76-90, p. 183 (1976).
22. R. Gwin et al., Oak Ridge report ORNL/TM-6246 (1978).
23. J. Frehaut et al, "Recent Results in Prompt Nubar Measurements between 1.5 and 15 MeV," [SCISRS Accession No. 20490, National Nucl. Data Center, BNL (1980)].
24. E. D. Arthur et al., Nucl. Sci. Eng. 88, 56 (1984).
25. H. Condé et al., J. Nucl. Energy 22, 53 (1968).
26. J. C. Hopkins and B. C. Diven, Nucl. Phys. 48, 433 (1963).
27. D. S. Mather et al., Nucl. Phys. 66, 149 (1965).
28. W. P. Poenitz et al., Nucl. Sci. Eng. 78, 333 (1981).
29. G. Haouat et al., Nucl. Sci. Eng. 81, 491 (1982).
30. A. B. Smith, P. T. Guenther, Argonne report ANL/NDM-63 (1982).

31. Ji Qun Shao, "Fast Neutron Inelastic Scattering Cross Sections of ^{238}U," Ph.D. Thesis, U. of Lowell (1984) (unpub.).
32. J. J. Egan et al., "A Comparison of Measured and Calculated Neutron Inelastic Scattering Cross Sections for Vibrational States from 680 to 1060 keV in ^{238}U, Contribution JB38, Int. Conf. on Nucl. Data for Basic and Ap. Sci., Santa Fe (1985).
33. D. G. Madland and J. R. Nix, Nucl. Sci. Eng. 81, 213 (1982).
34. D. G. Madland et al., "Differential and Integral Comparisons of Three Representations of the Prompt Neutron Spectrum for the Spontaneous Fission of ^{252}Cf," IAEA Advisory Meet. on Nucl. Standard Ref. Data, Geel, 1984.
35. D. G. Madland et al., "Comparisons of Four Representations of the Prompt Neutron Spectrum for the Spontaneous Fission of ^{252}Cf," Ibid. Ref. 32, Contribution B02.
36. W. P. Poenitz and T. Tamura, Ibid. Ref. 12, p. 465.
37. J. W. Boldeman et al., "Measurements of the Prompt Fission Neutron Spectrum from the Spontaneous Fission of ^{252}Cf," submitted to Nucl. Sci. Eng. (1985).
38. H. Alter et al., Brookhaven rept. BNL-19302 (ENDF 202) (1974).
39. R. B. Kidman, Los Alamos Nat. Lab. report LA-9037-MS (1981).