EVALUATION OF THE FISSION AND CAPTURE CROSS SECTIONS OF ^{240}Pu AND ^{241}Pu FOR ENDF/B-V

L. W. Weston and R. Q. Wright
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830, USA

Since there were appreciable new data which were not available for ENDF/B-IV, new evaluations for ^{240}Pu and ^{241}Pu were carried out for ENDF/B-V. The evaluation of the fission and capture cross sections will be reviewed and problem areas discussed. The neutron energy range of concern was from 10^{-5} eV to 20 MeV. Significant changes were made over the entire neutron energy region because of the new experimental data available. The problems in the evaluations due to discrepancies in the nuclear data will be emphasized, particularly the 1-eV resonance in ^{240}Pu and the 0.3-eV resonance in ^{241}Pu. The evaluation of the fission and capture cross sections for ENDF/B-V represents an improvement over the previous evaluation; however, there continues to be a need for accurate experimental data.

(neutron nuclear data evaluations, fission, capture, ^{240,241}Pu)

Introduction

An evaluation of the neutron capture and fission cross sections for ^{240}Pu and ^{241}Pu for ENDF/B-V was necessary because significant new data on these isotopes have become available. Some of these new data were discrepant with previous evaluations,[1,2,3] for example, resonance region of ^{241}Pu and the capture cross section of ^{240}Pu in the keV neutron energy region. In the case of the high energy fission cross sections the new data were superior to those previously available. Also, parts of the ENDF/B-IV evaluation were carried over from earlier versions of ENDF and therefore required updating.

Thermal Energy Range

Plutonium-240

A severe uncertainty in the evaluation of ^{240}Pu was the thermal neutron energy region which is dominated by the 1-eV resonance. About 99% of the thermal capture cross section is due to the influence of the 1-eV resonance. Because of the small quoted uncertainty, a single measurement[4] of the thermal cross section dominates the accepted 2200 m/s cross section as well as the determination of the product of the neutron width and radiation width, $\Gamma_n\Gamma_\gamma$, for the 1-eV resonance. This measurement is that of Lounsbury et al.[4] who report a 2200 m/s capture cross section of 289.5 ± 1.4 barns.

Plutonium-241

There have been two measurements[5,6] of the fission cross section and one measurement[6] of the capture cross section since the previous ENDF evaluation. The fission cross section was raised 2% at the 0.25-eV resonance based predominately on the experiments of Wagemans and Deruytter,[5] and Weston and Todd.[6]

The measurements of Weston and Todd[6] indicated a significantly higher capture cross section (14%) over the 0.25-eV resonance than the previous ENDF evaluation. The ENDF/B-IV evaluation was based on the total and fission cross section measurements of Simpson and Schuman[7] and Watanabe and Simpson.[8] The ENDF/B-V capture in the thermal neutron energy region is based on the measurements of Weston and Todd[6] for the following reasons; 1) the smaller uncertainty of the more direct measurement of the capture cross section, 2) the ENDF/B-IV capture in the resonance region was also determined to be low and 3) integral experiments suggest a higher capture cross section in the thermal neutron energy range. Figure 1 illustrates ENDF/B-V fission compared to Wagemans and Deruytter,[5] and the capture cross section of Weston and Todd[6] compared to ENDF/B-IV and V.

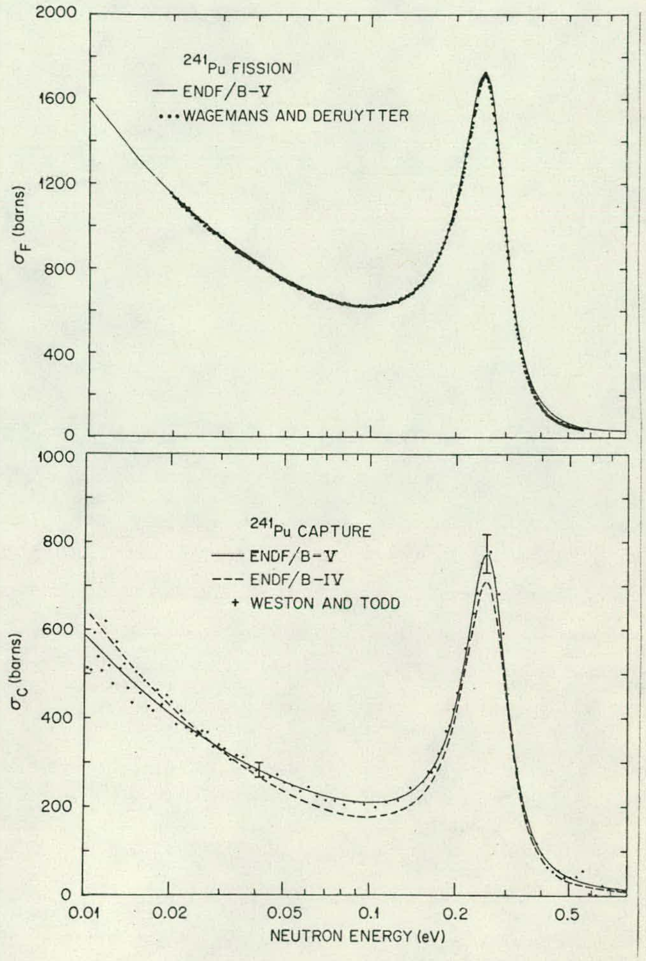

Fig. 1. The fission and capture cross sections of ^{241}Pu from 0.01 to 0.8 eV.

Resonance Region

Plutonium-240

The most severe uncertainty in the evaluation as far as thermal reactors are concerned is in the resonance parameters of the 1-eV resonance. The product of the neutron and radiation widths, $\Gamma_n\Gamma_\gamma$, is determined with an uncertainty of about 1% by the thermal cross section measurement of Lounsbury et al.[4] The individual resonance parameters, Γ_n and Γ_γ, have been measured in

DISCLAIMER

a number of experiments with discrepant results; these are described by Caner and Yiftah[10] and in a review paper by Weigmann et al.[11] In essence, there are two groups of experiments which predict radiation widths which are about 10% apart. This evaluation favors a high value of Γ_γ. The ENDF/B-V values of Γ_n and Γ_γ were taken to be 2.28 and 33.3 meV, respectively, as evaluated by Caner and Yiftah.[10]

Integral experiments yield little information on these resonance widths because they are usually carried out with low ^{240}Pu content as compared to ^{239}Pu and they are most sensitive to the product, $\Gamma_n\Gamma_\gamma$. Thompson and Leonard[12] are using least squares fitting techniques to more accurately determine the widths, Γ_n and Γ_γ, from the available data.

There is a need for careful, accurate experimental measurements to determine the resonance parameters of the 1-eV resonance of ^{240}Pu. Without such additional experimental measurements, the large uncertainty in the cross section of ^{240}Pu below a neutron energy of a few eV will continue to be a problem.

The resonance parameters from 20 to 665 eV for ENDF/B-V are the weighted average of the parameters of Weigman et al.,[11] Asghar et al.[13] and Hockenbury et al.[14] Above 665 eV, the neutron widths of Kolar and Bockhoff[15] were used as was the case for ENDF/B-IV. The fission widths for the subthreshold fission resonances were evaluated from the data of Auchampaugh and Weston,[16] Migneco and Theobald,[17] and Weigman et al.[11]

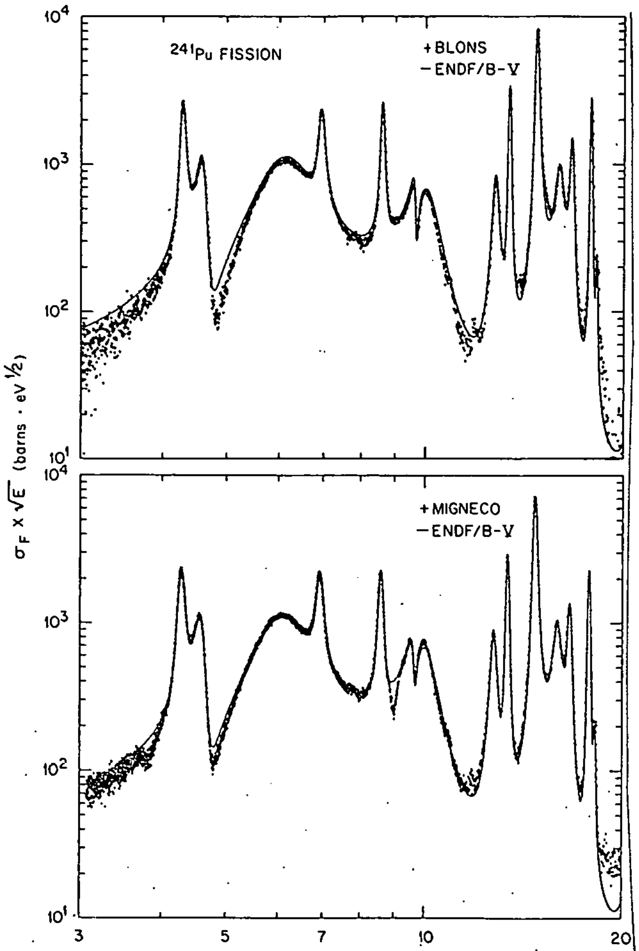

Fig. 2. The fission cross section of ^{241}Pu from 3 to 20 eV.

Plutonium-241

Because of appreciable level-level interference in the fission cross section, this evaluation is in the form of the Adler-Adler[18] multilevel formalism. The data of Weston and Todd[19] had been fit in the Adler-Adler formalism. Fits to the data of Blons,[20] Simpson and Shuman,[7] James,[21] and Moore et al.[22] which were in the Reich-Moore formalism were converted to the Adler-Adler formalism using the computer code, POLLA, and techniques developed by de Saussure and Perez.[23] This evaluation was carried out using these sets of parameters. The resulting capture cross section, and thus the average radiation width, is higher (~40%) than the ENDF/B-IV evaluation. Indications that the capture cross section was low in the previous evaluation had been noted previously.[10,20] Figure 2 illustrates the ENDF/B-V fission cross section compared to the measurements of Blons,[20] and Migneco et al.[24] Figure 3 compares the ENDF/B-V fission and capture cross sections with the measurements of Weston and Todd.[6]

Unresolved Resonance Region

Plutonium-240

The capture cross section of ^{240}Pu in the unresolved resonance region is illustrated in Fig. 4. The average resonance parameters[11] as derived from the resonance region were in good agreement with the average capture cross section measurements of Weston and Todd[25] so these parameters were used for the evaluation. This

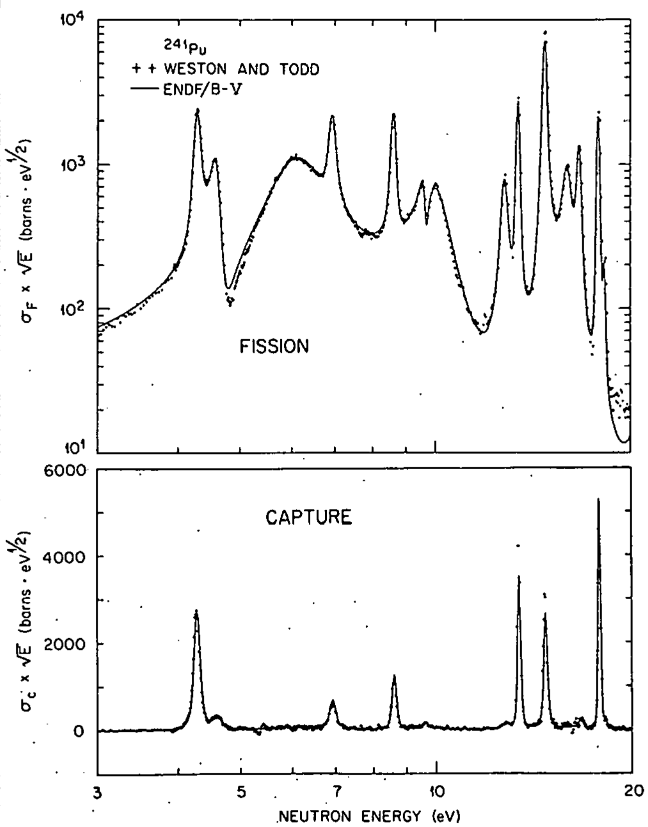

Fig. 3. The fission and capture cross section of ^{241}Pu for ENDF/B-V compared to Weston and Todd from 3 to 20 eV.

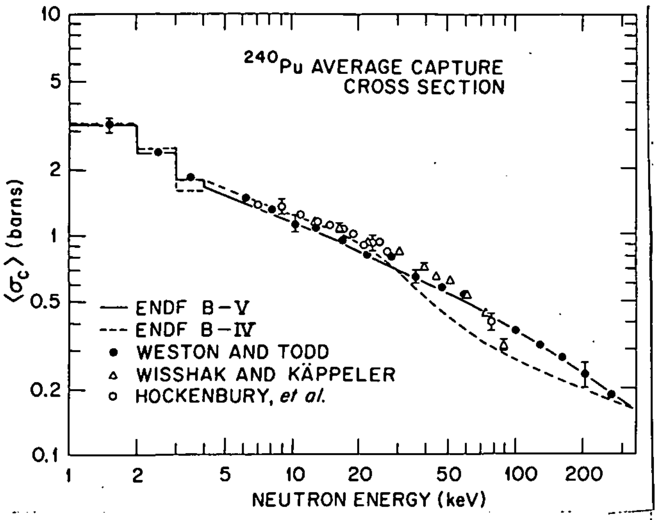

Fig. 4. The average capture cross section of ^{240}Pu from 1 to 300 keV.

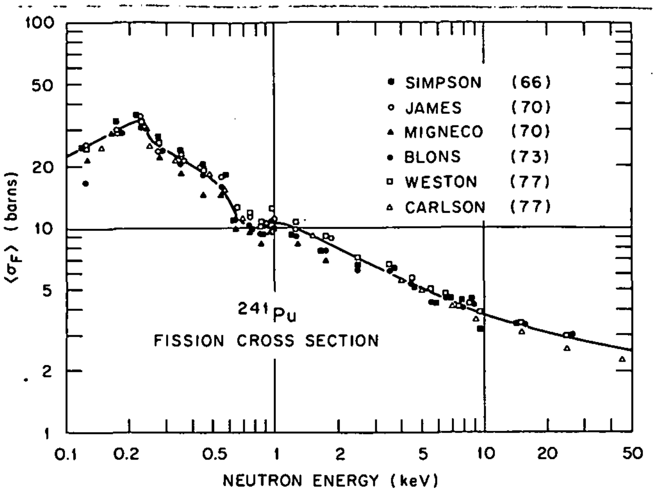

Fig. 5. The fission cross section of ^{241}Pu in the neutron energy range from 0.1 to 50 keV.

evaluation is lower than the measurements of Hockenbury et al.:[14] however, these experimenters[26] indicated their results may have been high because the canning of the sample was not properly taken into account. The measurements of Wisshak and Käppeler were available only in very preliminary form at the time of the evaluation and were not considered. The points of Wisshak and Käppeler[27] in Fig. 4 were converted by the author from ratio measurements with respect to Au using the ENDF/B-V Au cross section. These points are only part of the data and are only intended to be representative of the data. The Wisshak and Käppeler [27] and the Weston and Todd[25] measurements are not very discrepant on a point-by-point basis, but the difference in shape is disturbing.

The fission cross section of ^{240}Pu is small and relatively insignificant from thermal through the unresolved resonance region. The evaluation of this cross section in the unresolved resonance region was based on the data of Behrens et al.[28]

Plutonium-241

The fission cross section of ^{241}Pu in the unresolved resonance region is illustrated in Fig. 5. Most of the experimental measurements[6,17,20,21,29,30] shown in Fig. 5 indicate an unexpected rise between 100 and 200 eV and a dip in the cross section at about 800 eV. These measurements show disturbing discrepancies (~10%) when their quoted uncertainties are considered (~3%). The ENDF/B-V evaluation reproduces the major structure which is common to the experimental results.

The data on the capture cross section of ^{241}Pu by Weston and Todd[6] in the energy region shown in Fig. 5 indicate the same structure (rise between 100 and 200 eV and dip at about 800 eV) which appears in the fission cross section. This is somewhat surprising because if the structure were due to the usual effect of the well-known double fission barrier the fission widths would be modulated. Since the structure appears in the capture cross section as well as in the fission cross section it must be due to one or both components of the strength function, $\langle \Gamma_n/D \rangle$. The evaluation of the capture cross section was based on the data of Weston and Todd[6] since no other differential data are available.

High Energy Region

Plutonium-240

The evaluation of the fission cross section of ^{240}Pu was based upon the ratio to ^{235}U measurements of Behrens et al.[28] These data appeared superior to other available data at the time of the evaluation. Since the evaluation, additional data by Kari and Cierjacks,[31] and Budtz-Jorgensen and Knitter[32] have become available which are in reasonable agreement with the measurements of Behrens. The data of Behrens[28] converted to the fission cross section with the use of the ENDF/B-V fission cross section for ^{235}U are compared in Fig. 6 with the ENDF/B-V evaluation for ^{240}Pu and the ENDF/B-IV evaluation for ^{240}Pu.

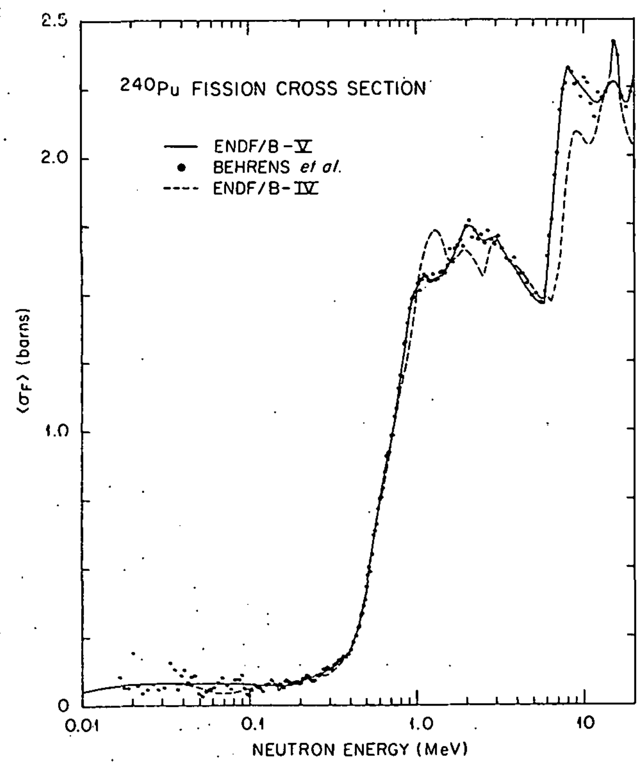

Fig. 6. The fission cross section of ^{240}Pu at high-neutron energies.

The capture cross section of ^{240}Pu above 300 keV is unchanged from Version IV. The changes below this neutron energy were illustrated in Fig. 4.

Plutonium-241

Figure 7 illustrates the experimental data,[30, 33-36] the ENDF/B-V, and the ENDF/B-IV evaluations for the high-energy fission cross section of ^{241}Pu. Again the ratio data of Carlson and Behrens[33] were given the principle weight in the evaluation because of accuracy, consistency with ^{240}Pu, and complete coverage of the high-energy neutron range.

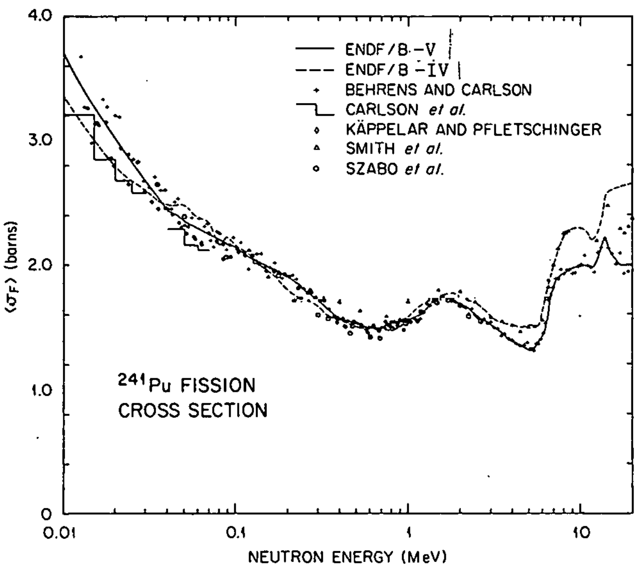

Fig. 7. The fission cross section of ^{241}Pu at high-neutron energies.

Conclusions

The capture and fission cross sections for ^{240}Pu and ^{241}Pu were evaluated from 10^{-5} eV to 20 MeV for ENDF/B-V. Due to new experimental data which have become available the new evaluation is placed on a firmer basis than the earlier evaluation.

In the authors' opinion, three problem areas remain which would require new, careful experimental measurements to resolve. These are listed in decreasing order of importance:

1. The resonance parameters of the 1-eV resonance of ^{240}Pu are not known with a sufficient accuracy. The 1-eV resonance also controls the 2200 m/s capture cross section and so a confirmation of the experiment of Lounsbury et al.[4] would be worthwhile.

2. The capture cross-section measurements on ^{241}Pu of Weston and Todd[6] carried an inordinate weight because they were the only extensive differential capture cross-section measurements from 0.01 eV to 200 keV neutron energy. Another independent measurement would be of value.

3. The ^{241}Pu fission cross section in the unresolved resonance region is uncertain by about 5%. There are a number of experimental measurements but they are discrepant. A careful high-precision measurement of this cross section would be most useful.

Acknowledgment

Research sponsored by the Reactor Research and Technology Division, U. S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

References

1. E. Pennington and H. Hummel, ENDF/B-IV, MAT 1265, File 1, Brookhaven National Laboratory (1974).

2. H. Hummel and E. Pennington, ENDF/B-IV, MAT 1266, File 1, Brookhaven National Laboratory (1974).

3. R. E. Hunter, L. Stewart, and T. S. Hirons, "Evaluated Neutron-Induced Cross Sections for ^{239}Pu and ^{240}Pu," LA-5172, Los Alamos Scientific Laboratory (1973).

4. M. Lounsbury, R. W. Durkam, and G. C. Hanna, Proc. Conf. on Nuclear Data for Reactors, Helsinki, IAEA, Vienna (1970) p. 287.

5. C. Wagemans and A. J. Deruytter, Proc. Conf. Nuclear Cross Sections and Technology, Washington, National Bureau of Standard Special Publication, 425, II, p. 603 (1975).

6. L. W. Weston and J. H. Todd, Nucl. Sci. Eng. 65, 454 (1978); for resonance parameters see Nucl. Sci. Eng. 68, 125 (1978).

7. O. D. Simpson and R. P. Schuman, Nucl. Sci. Eng. 11, 111 (1961).

8. T. Watanabe and O. D. Simpson, IDO-16995, Phillips Petroleum, Idaho (1964).

9. H. D. Lemmel, Proc. Symp. Neutron Standards and Applications, National Bureau of Standards, Washington D. C., NBS-493 (1977) p. 170.

10. M. Caner and S. Yiftah, "Nuclear Data Evaluation for Plutonium-240," IA-1243; "Nuclear Data Evaluation for Plutonium-241," IA-1276, Israel Atomic Energy Commission (1972).

11. H. Weigmann, G. Rohr, and F. Pourtmans, Proc. Conf. Resonance Parameters of Fertile Nuclei and ^{239}Pu, Saclay, Commissariat a L'Energia Atomique, NEANDC(E) 163U (1974) p. 219.

12. J. K. Thompson and B.,R. Leonard, Jr., private communication (1978).

13. M. Asghar, M. C. Moxon, and N. J. Pattenden, Proc. Conf. Nuclear Data for Reactors, Paris, IAEA, Vienna (1966); see ref. 7 for revised parameters.

14. R. W. Hockenbury, W. R. Moyer, and R. C. Block, Nucl. Sci. Eng. 49, 153 (1972).

15. W. Kolar and K. H. Bockhoff, J. Nucl. Energy 22, 299 (1968).

16. G. F. Auchampaugh and L. W. Weston, Phys. Rev. C12, 1850 (1975).

17. E. Migneco and J. P. Theobald, Nucl. Phys. A112, 603 (1968).

18. D. B. Adler and F. T. Adler, Proc. Conf. Breeding Economics and Safety in Large Fast Power Reactors, Argonne, Illinois, ANL-6792, Argonne National Laboratory (1963) p. 695.

19. L. W. Weston and J. H. Todd, Nucl. Sci. Eng. 68, 125 (1978).

20. J. Blons, Nucl. Sci. Eng. 51, 130 (1973).

21. G. D. James, Proc. Conf. Nuclear Data for Reactors, Helsinki, 1, p. 267, International Atomic Energy Agency, (1970).

22. M. S. Moore et al., Phys. Rev. 135, B945 (1964).

23. G. de Saussure and R. B. Perez, "POLLA, A Fortran Program to Convert R-Matrix-Type Multilevel Resonance Parameters for Fissile Nuclei into Equivalent Kapur-Peierls-Type Parameters," ORNL-TM-2599, Oak Ridge National Laboratory (1969).

24. E. Migneco et al., Proc. Conf. Nuclear Data for Reactors, Helsinki, 1, p. 437, International Atomic Energy Agency (1970).

25. L. W. Weston and J. H. Todd, Nucl. Sci. Eng. 63, 143 (1977).

26. R. W. Hockenbury, private communication (1978).

27. K. Wisshak and F. Käppeler, Nucl. Sci. Eng. 66, 363 (1978).

28. J. W. Behrens, J. C. Browne, and G. W. Carlson, Nucl. Sci. Eng. 66, 433 (1978).

29. O. D. Simpson et al., Proc. Conf. Neutron Cross Sections and Technology, Washington, CONF-660303, II, p. 910, USAEC (1966).

30. G. W. Carlson, J. W. Behrens, and J. B. Crizz, Nucl. Sci. Eng. 63, 149 (1977).

31. K. Kari and S. Cierjacks, Proc. Conf. Neutron Physics and Nuclear Data, Harwell, p. 905, OECD Nuclear Energy Agency (Paris) (1978).

32. C. Budtz-Jorgensen and H. H. Knitter, Proc. Nuclear Data of Plutonium and Americium Isotopes for Reactor Applications, p. 239, Brookhaven, BNL-50991, Brookhaven National Laboratory (1978).

33. G. W. Carlson and J. W. Behrens, Nucl. Sci. Eng. 68, 128 (1978).

34. F. Käppeler and E. Pfletschinger, Nucl. Sci. Eng. 51, 124 (1973).

35. H. L. Smith, R. K. Smith, and R. L. Henkel, Phys. Rev. 125, 1329 (1962).

36. I. Szabo et al., Proc. Symp. Neutron Standards, Argonne National Laboratory, CONF-701002, p. 257 USAEC (1970).