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REVIEW OF MICROSCOPIC NEUTRON CROSS SECTION DATA FOR THE HIGHER PLUTONIUM ISOTOPES IN THE RESONANCE REGION

L. W. Weston

Oak Ridge National Laboratory
Oak Ridge, Tennessee 37830, U.S.A.

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ABSTRACT

The microscopic neutron cross section data for plutonium-240, -241, and -242 in the resonance region are reviewed. In the context of importance to reactors the data on plutonium-240 are acceptable except for the resonance parameters of the 1-eV resonance. Plutonium-241 has discrepancies in the cross sections from thermal to 3 eV and to a lesser extent in the fission and capture cross section in the unresolved resonance region. The plutonium-242 cross sections appear to be known with sufficient accuracy as compared with the other plutonium isotopes.

INTRODUCTION

The higher plutonium isotopes are important both from the point of view of reactor core physics and waste management. The typical composition [1] of the plutonium discharge from a light water reactor is as follows:

59%	^{239}Pu
26%	^{240}Pu
12%	^{241}Pu
3%	^{242}Pu

The implication for reactor core physics is obvious. Plutonium-240 is a poor reactor fuel with neutron absorption leading to

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plutonium-241. Plutonium-241 which has a 15-year half-life beta decays to americium-241. This decay is important because plutonium-241 is an excellent reactor fuel and americium-241 is not very fissionable. Neutron capture in americium-241 leads to curium-242 which has a high spontaneous fission rate and is a waste management problem. Plutonium-242 is not very fissionable and neutron capture leads to higher actinides which are waste management problems.

PLUTONIUM-240

From the point of view of importance to a thermal reactor plutonium-240 is dominated by one very large capture resonance at 1 eV. This resonance is over 10^5 barns in height and determines more than 99% of the thermal capture cross section. This resonance is illustrated in Fig. 1.

This resonance is so large that it has been ignored in recent years as far as experimental measurements are concerned. The reason it has been ignored is that a special sample is needed for measurements on this one resonance. There are total cross section measurements [2-8] dating from the period of 1955 to 1959 and one additional measurement [9] in 1970. The partial widths (Γ_n and Γ_γ) of this resonance have only been known to about 10%. The most accurate constraint on the resonance parameters has been the measurement of the thermal cross section by Lounsbury et al. [10] [$\sigma_\gamma(0.0253 \text{ eV}) = 289.5 \pm 1.4$ barns]. This measurement puts an accurate constraint on the product of the resonance parameters, $\Gamma_n \Gamma_\gamma$. There is no corroboration of this accuracy since this is the only measurement of comparable accuracy of the thermal cross section.

Thompson and Leonard [11] have preliminary results from a least-squares fit of the major part of the experimental data from thermal over the 1-eV resonance which indicates a rather high precision for the fit. Renormalization of the cross sections and energy scale was allowed in the fit. These results may significantly improve the situation; however, new measurements of the total and scattering cross sections from 0.01 to 3 eV would be of great value.

The resonance parameters of the higher resonances of plutonium-240 are in reasonably good agreement. For a time the total cross section data of Kolar and Bockoff, [12] Ashgar et al., [13] and Hockenbury et al. [14] were discrepant, however, this was principally a normalization problem which is now rectified. The review paper of Weigman [15] contains an extensive review of the resonance parameters.

The average resonance parameters from the resonance region (1 eV to ~ 1.5 keV) are in good agreement with the capture cross section in the keV neutron energy region as measured by Weston and Todd [16] and in fair agreement with the data of Hockenbury et al.

[14] and Käppeler et al. [17] Between 200 eV and 3 keV the average capture cross section calculated from the resonance parameters is lower ($\sim 17\%$) than that measured by Weston and Todd, [16] however, this could be the effects of missed small resonances in the total cross section. Figure 2 illustrates the average capture cross section of plutonium-240. The curve marked ENDF/B-V from 3.9 to 40 keV was calculated from the average resonance parameters in the resonance region.

Plutonium-240 has a small but measurable subthreshold fission cross section. Below 10 keV it is characterized by several groups of resonances which are enhanced by the second well in the fission barrier. This is illustrated in Fig. 3. The parameters of these resonances are known reasonably well from the measurements of Auchampaugh and Weston [18] and Migneco and Theobald. [19] Above 10 keV there is a great deal of structure as indicated in Fig. 4 which illustrates very preliminary data of Weston and Todd. These data also confirm the gross structure found by Wisshak and Käppeler [20] in that there is grouping of resonances centered at about 12, 20, 38, and 70 keV. This gross structure is superimposed upon the well known structure imposed by the second well of the fission barrier. It is not clear whether this gross structure is additional effects of the complex fission barrier or due to other causes.

PLUTONIUM-241

In plutonium-241 the major important discrepancy is also at low neutron energies. The fission cross section data of Wagemann and Deruytter [21] and Weston and Todd [22] are in good agreement from 0.02 to 0.5 eV as indicated in Figs. 5 and 6. The agreement ($\sim 3.5\%$) of these fission plus the capture cross section with the total cross section measurements of Simpson and Schuman [23] is not good. As in the case of plutonium-240 accurate total and scattering cross section from 0.01 to 3 eV would be of great value.

The higher resolved resonance region of plutonium-241 is known with reasonable certainty as indicated in Figs. 7 and 8. The fission cross section in this energy region has been measured in a number of experiments. [21-22, 24-29] The only differential capture cross section measurement is that of Weston and Todd. [22] The resonance parameters derived from total [23, 30-32] and fission cross sections are in agreement within experimental uncertainties with the measurement [22] of capture cross section.

There have been a number of measurements [22, 24-27, 29] of the fission cross section in the unresolved resonance region from 100 eV to 40 keV, however, the agreement between the measurements is only about 10% as indicated in Fig. 9. Integrals over larger neutron energy ranges also indicate similar discrepancies. [22] As seen in Fig. 9 there is structure in the unresolved range as characterized by the peak at ~ 200 eV and the dip at 800 eV. Apparently this structure is not due to modulation of the fission

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widths caused by the second well in the fission barrier since the capture cross section exhibits the same type structure as seen in Fig. 10. This indicates the modulation must arise from variations in the strength function, Γ_n/D .

An additional problem with plutonium-241 is the value of $\bar{\nu}$, the number of neutrons per fission, between thermal and a few hundred keV. Measurements [33-34] of $\bar{\nu}$ in the MeV region of neutron energies extrapolate to a lower thermal $\bar{\nu}$ than the currently accepted one as can be seen in Fig. 11. This could be due to the californium-252 standard for $\bar{\nu}$ or other experimental effects which have nothing to do with plutonium-241. If there is such an effect in plutonium-241, it is most probably in the thermal and resonance region.

PLUTONIUM-242

Plutonium-242 is similar to plutonium-240 in that the sub-threshold fission is small up to a few hundred keV. There are no severe problems near thermal for plutonium-242 as there are for plutonium-240. There have been a number of total cross section measurements [35-40] from thermal through the resolved resonance region for plutonium-242 as indicated in Fig. 12. Particularly noteworthy are the measurements of Poortmans et al. [39] which include total, scattering, and capture cross sections. The measurements of Auchampaugh and Bowman [38] were principally a study of the subthreshold fission cross section. Unfortunately the high resolution measurements of Simpson [40] with samples at liquid nitrogen temperature have only been partially analyzed and published. Since plutonium-242 is produced in reactors at a relatively low abundance as discussed earlier, the cross sections appear to be sufficiently well known at the present time as compared to other nuclides.

CONCLUSIONS

Table I gives an "Evaluators Wish List for the Resonance Region of the Higher Plutonium Isotopes." In the opinion of the author, these are the most important needs for cross sections for the higher plutonium isotopes from thermal through the unresolved resonance region. This does not mean there are not other problems, which there are, but simply that these are the most outstanding problems in an evaluation of the cross sections for reactor core physics and waste management purposes.

The need for accurate total and scattering cross section from 0.01 to 3 eV for plutonium-240 and -241 is to resolve severe discrepancies which exist in these important cross sections. In the case of plutonium-241 the problem could be in the capture and fission cross sections, however, until a modern total cross section measurement is carried out this is impossible to determine.

An accurate measurement of the plutonium-241 fission cross section in the unresolved region would be useful on a second priority basis because of the large scatter of previous measurements. Such a measurement should be carefully done and precise ($\sim 2\%$) so as not to simply add to the confusion. A thorough theoretical interpretation of the cross section in this neutron energy region might help to determine the shape of this cross section.

Since there is only one published differential capture cross section measurement [22] for plutonium-241, a corroborative experiment would be useful. This measurement is difficult and more than one measurement should be carried out.

In general the cross sections of the higher Pu isotopes are well known as compared to those of plutonium-239. The exception to this is the 1-eV resonance of plutonium-240. The uncertainties in the plutonium-239 cross sections tend to dominate over the uncertainties in the higher plutonium isotopes. If the uncertainties in the plutonium-239 cross sections are reduced then there may be a need for further effort on the higher Pu isotopes.

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TABLE I

EVALUATORS WISH LIST FOR THE RESONANCE REGION OF THE
HIGHER PLUTONIUM ISOTOPES

First Priority

TOTAL CROSS SECTION MEASUREMENTS WITH MULTIPLE SAMPLE THICKNESSES OF ^{240}Pu AND ^{241}Pu FROM 0.01 TO 3 eV TO ACCURATELY DETERMINE THE LOW-ENERGY RESONANCE PARAMETERS. SCATTERING CROSS SECTIONS IN THE SAME NEUTRON ENERGY REGION WOULD BE OF GREAT VALUE.

Second Priority

1. AN ACCURATE (< 2%) MEASUREMENT OF THE AVERAGE FISSION CROSS SECTION IN THE UNRESOLVED RESONANCE REGION OF ^{241}Pu BETWEEN 100 eV AND 50 keV. A BETTER THEORETICAL INTERPRETATION OF THIS NEUTRON ENERGY REGION WOULD ALSO BE HELPFUL.
2. CORROBORATIVE MEASUREMENTS OF CAPTURE CROSS SECTIONS, PARTICULARLY IN THE NEUTRON ENERGY RANGE FROM 100 eV to 10 keV.

Fig. 1. The 1-eV Resonance of Plutonium-240.

Fig. 2. The Average Capture Cross Section of Plutonium-240 from 100 eV to 1 MeV.

Fig. 3. The Subthreshold Fission Cross Section of Plutonium-240.

Fig. 4. The Complex Structure in the Plutonium-240 Subthreshold Fission Cross Section Between 10 and 100 keV as Indicated by Preliminary Results of Weston and Todd.

Fig. 5. Comparison of the Preliminary ENDF/B-V Evaluation of the Plutonium-241 Fission Cross Section with the Data of Wagemans and Deruytter [21].

Fig. 6. Comparison of the Preliminary ENDF/B-V Evaluation of the Plutonium-241 Fission and Capture Cross Sections with the Data of Weston and Todd [22].

Fig. 7. The Fission Cross Section Data of Blons [24] and Migneco [26] Between 3 and 20 eV as Compared to the ENDF/B-V Evaluation.

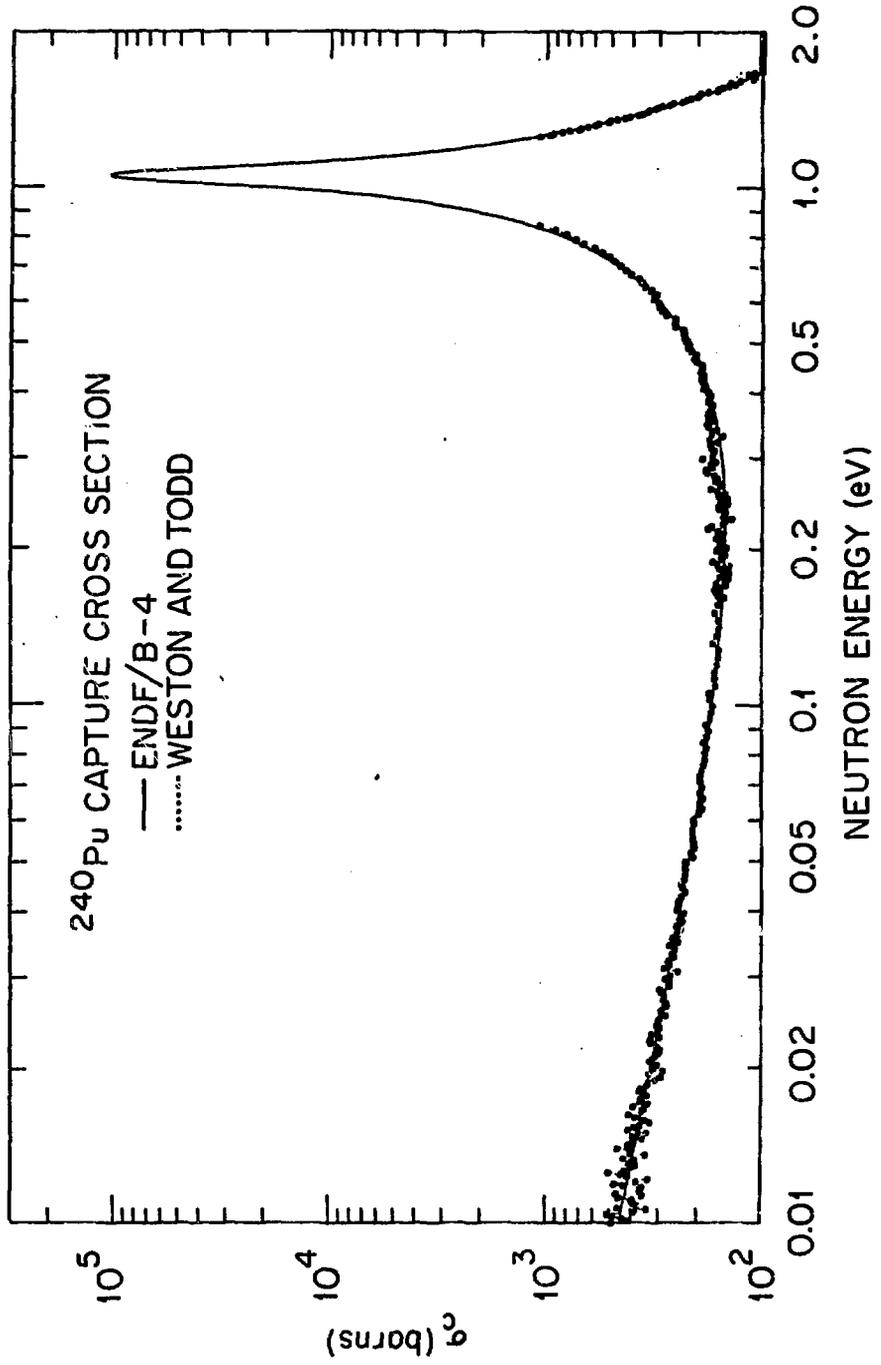
Fig. 8. The Fission and Capture Cross Section Data of Weston and Todd [22] Between 3 and 20 eV as Compared to the Preliminary ENDF/B-V Evaluation.

Fig. 9. The Plutonium-241 Average Fission Cross Section in the Unresolved Resonance Region.

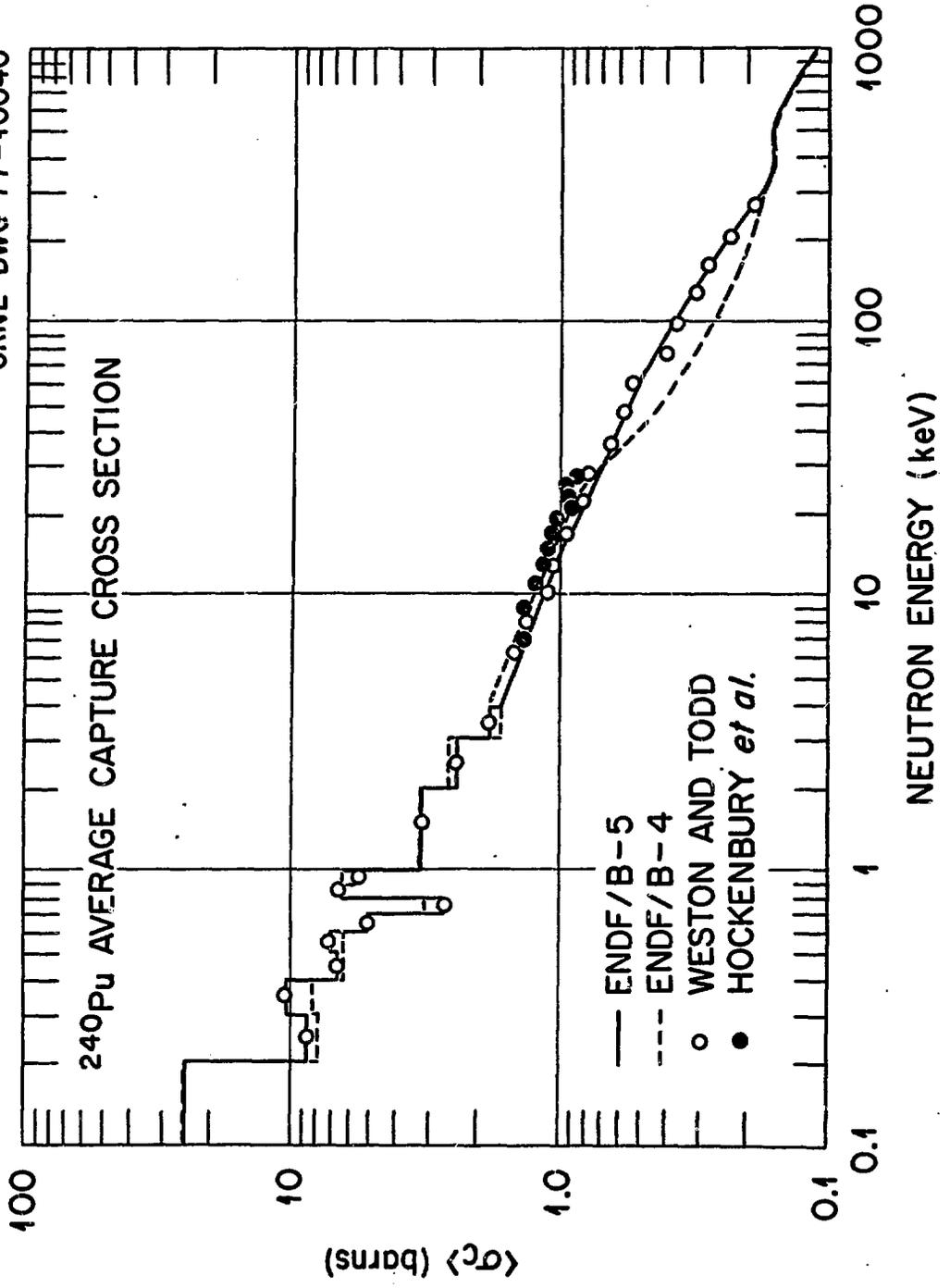
Fig. 10. The Average Capture Cross Section of Plutonium-241 in the Unresolved Resonance Region.

Fig. 11. The Average Number of Prompt Neutrons per Fission from Thermal to 20 MeV. Note that the Preliminary ENDF/B-V Evaluation does not Extrapolate Smoothly to the Thermal Value.

Fig. 12. Total Cross Section Measurements on Plutonium-242 in the Resolved Resonance Region.



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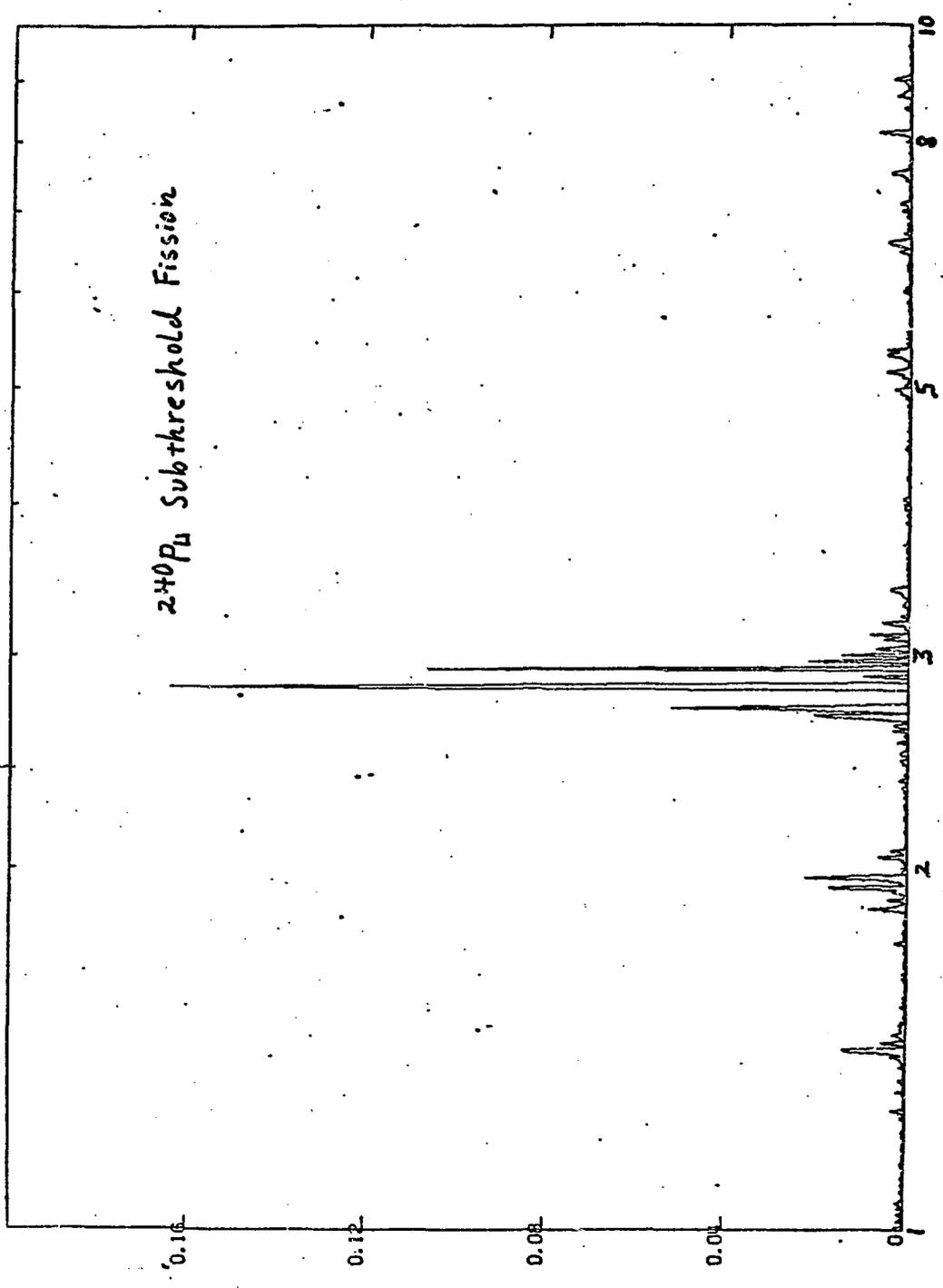
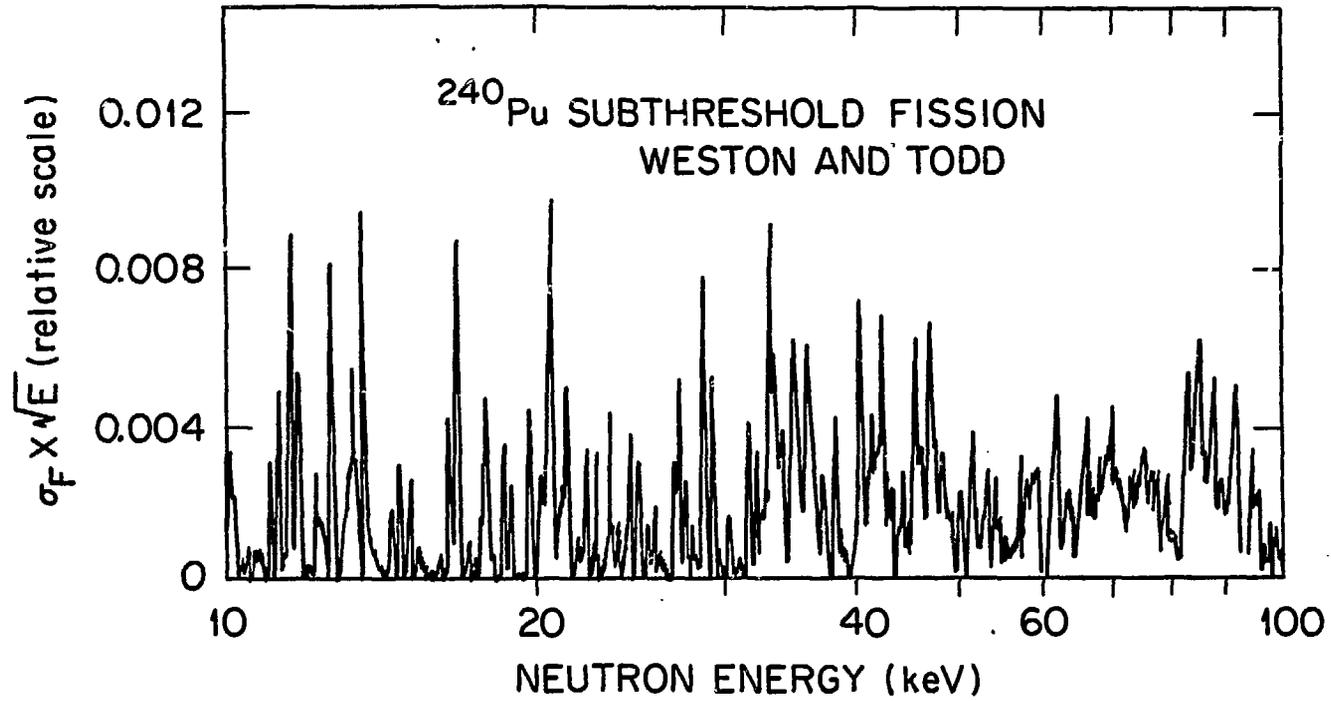
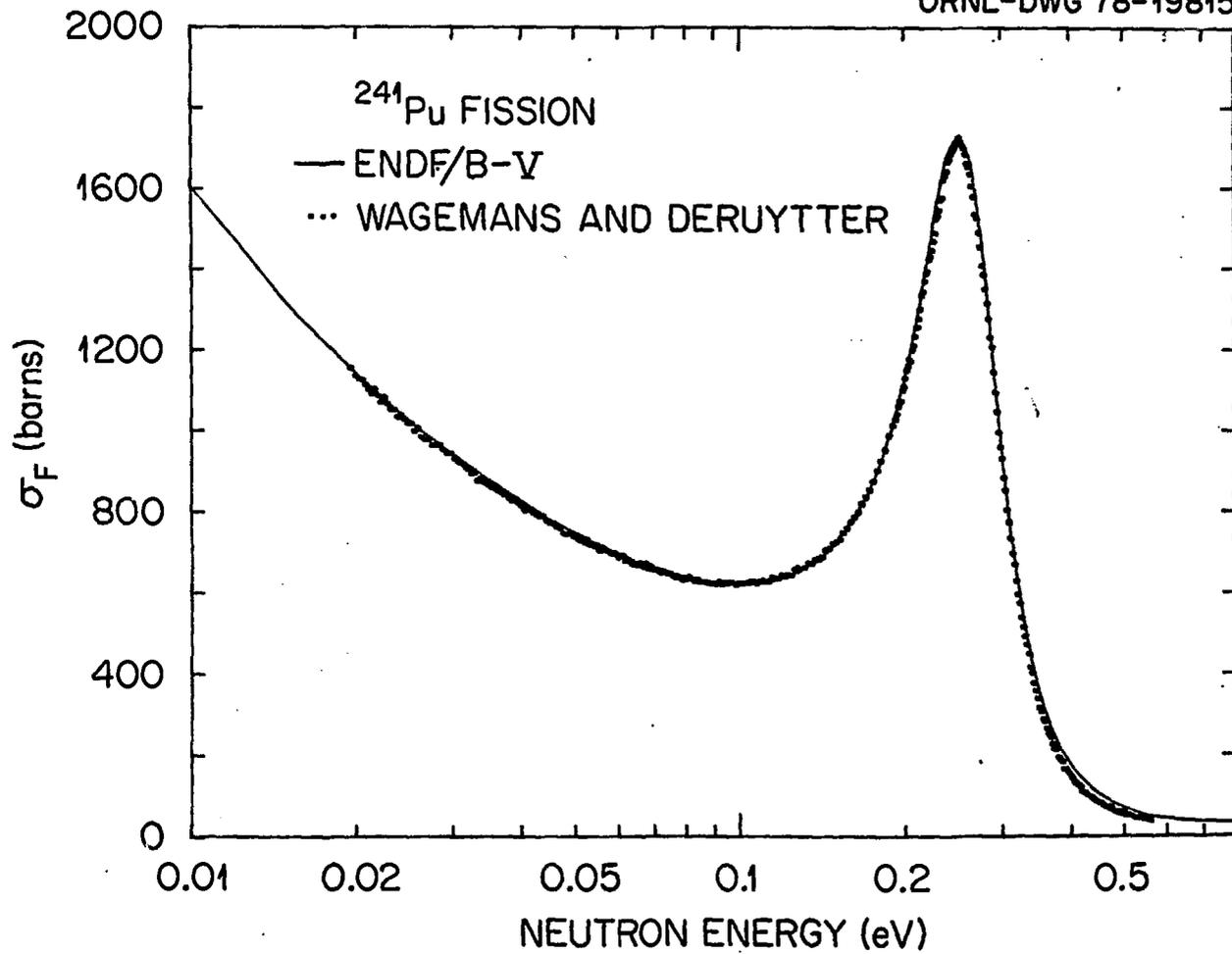


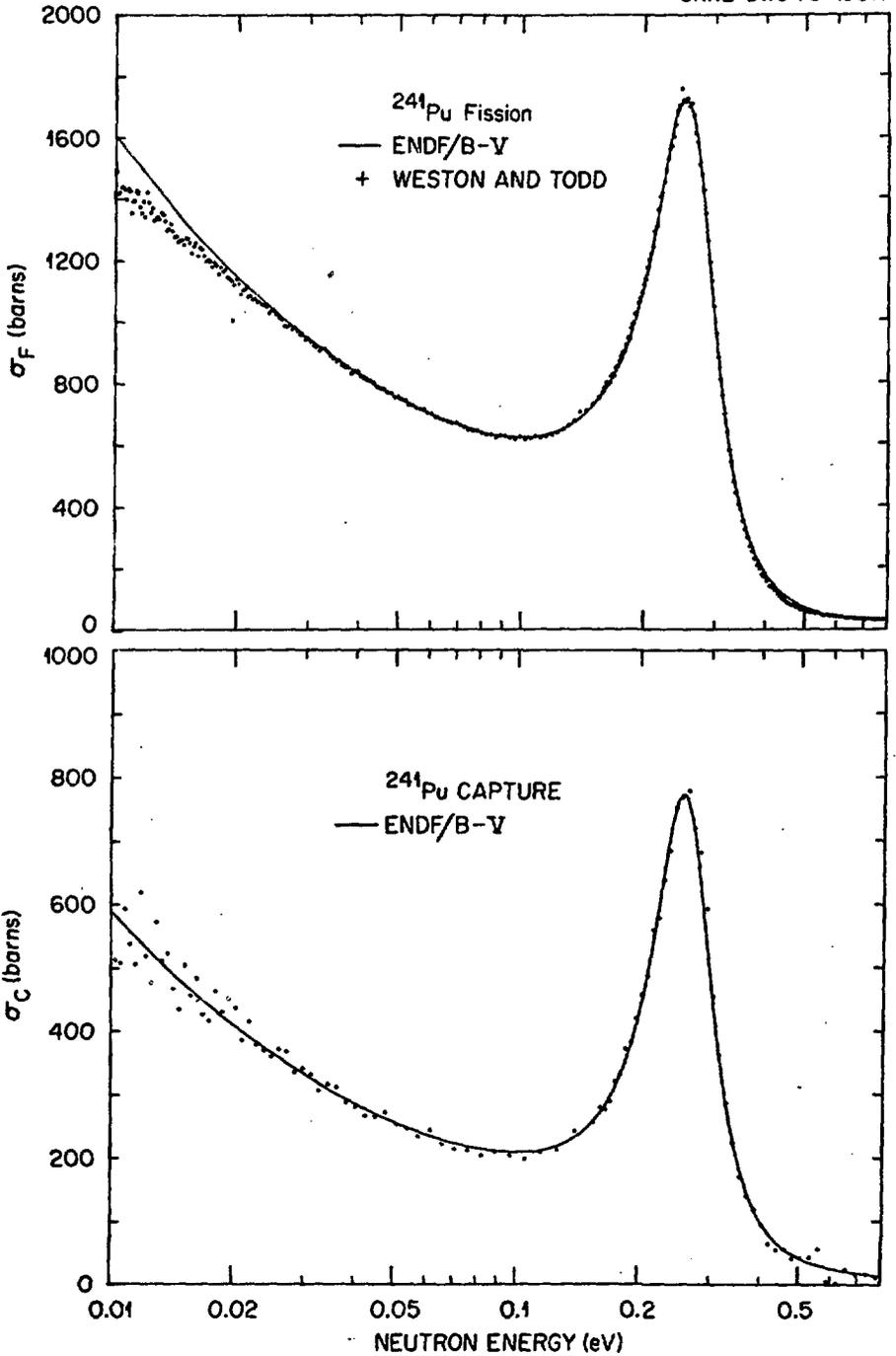
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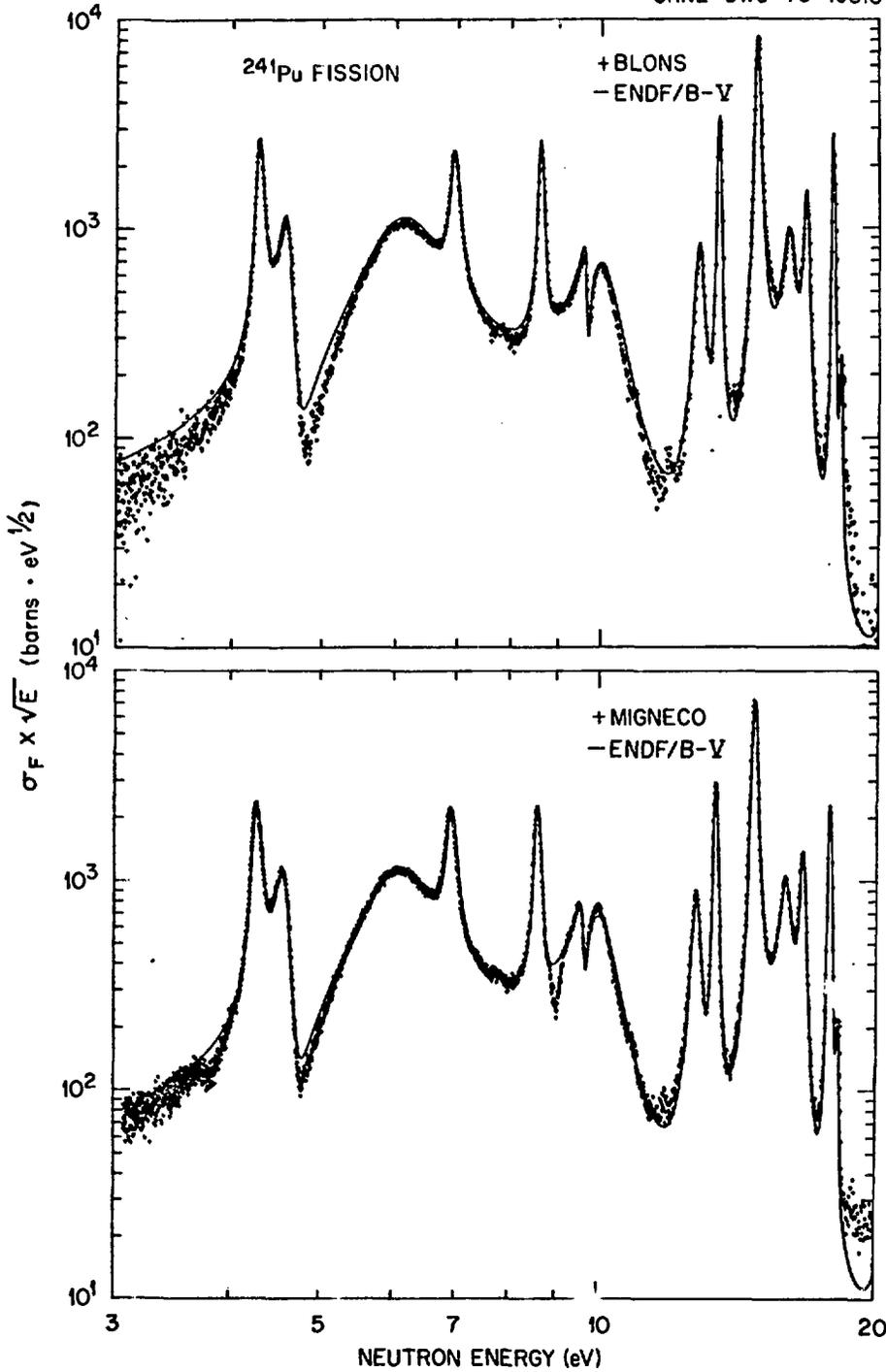
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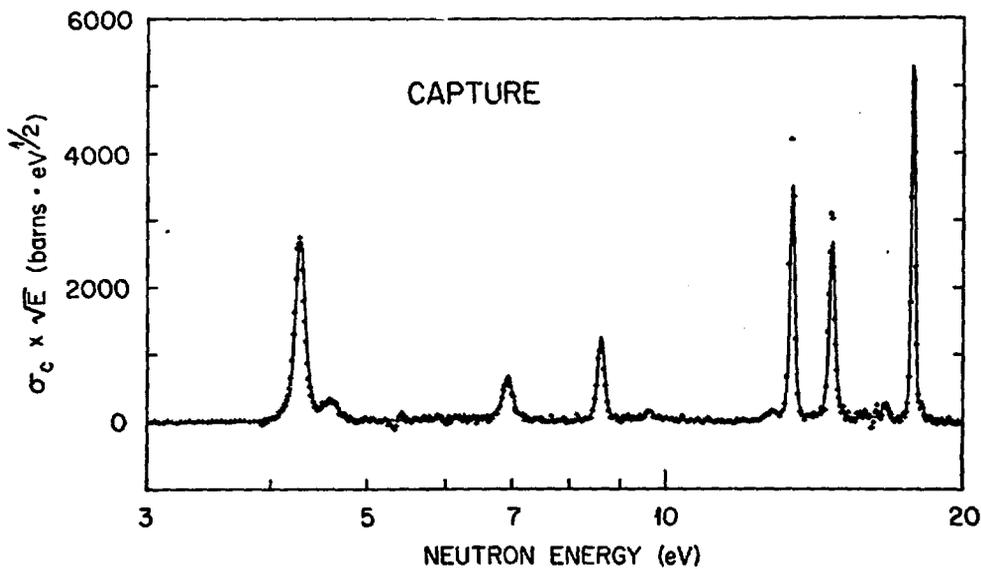
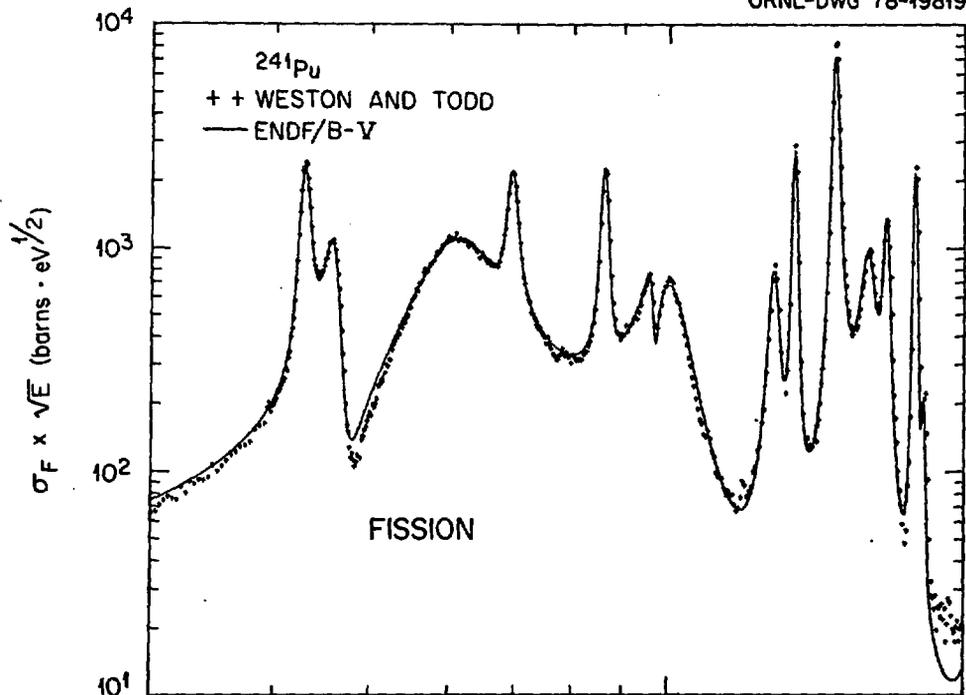


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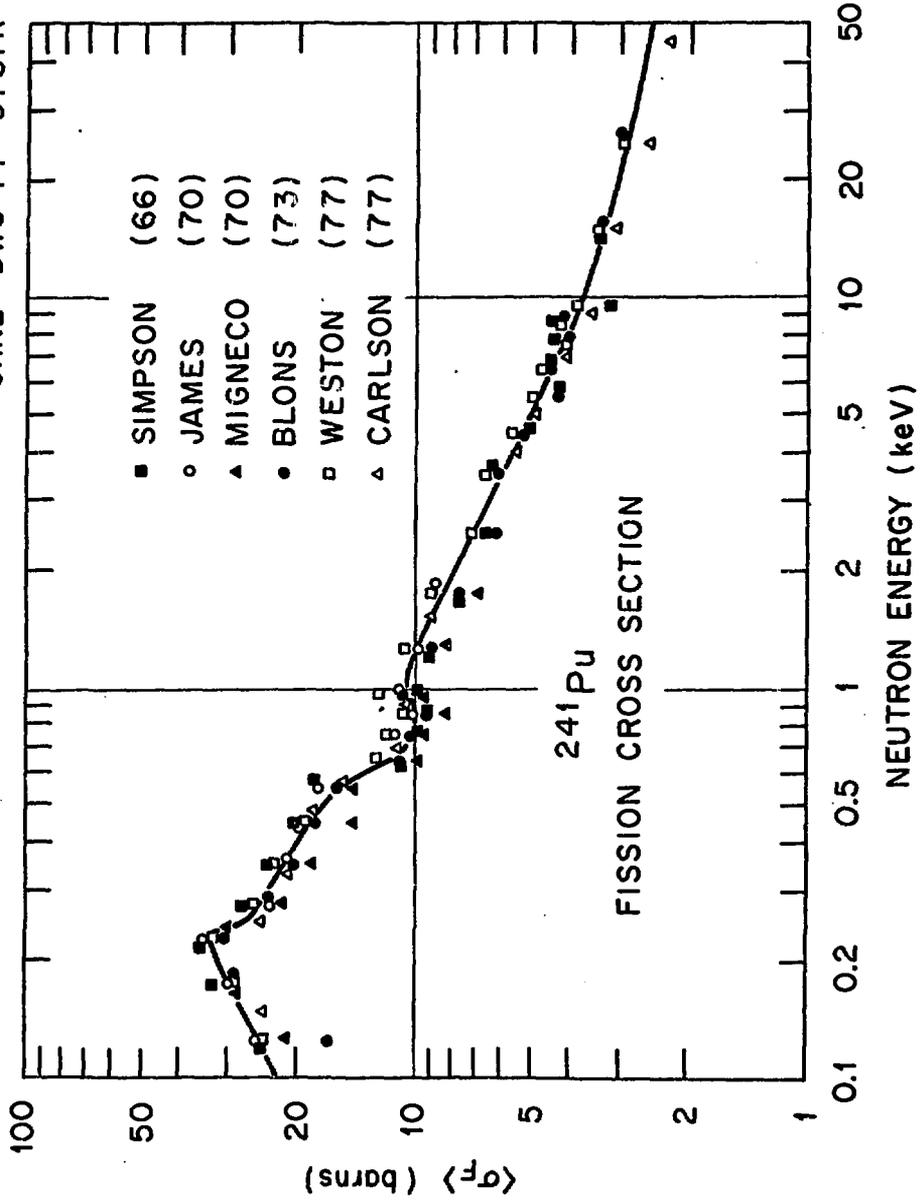


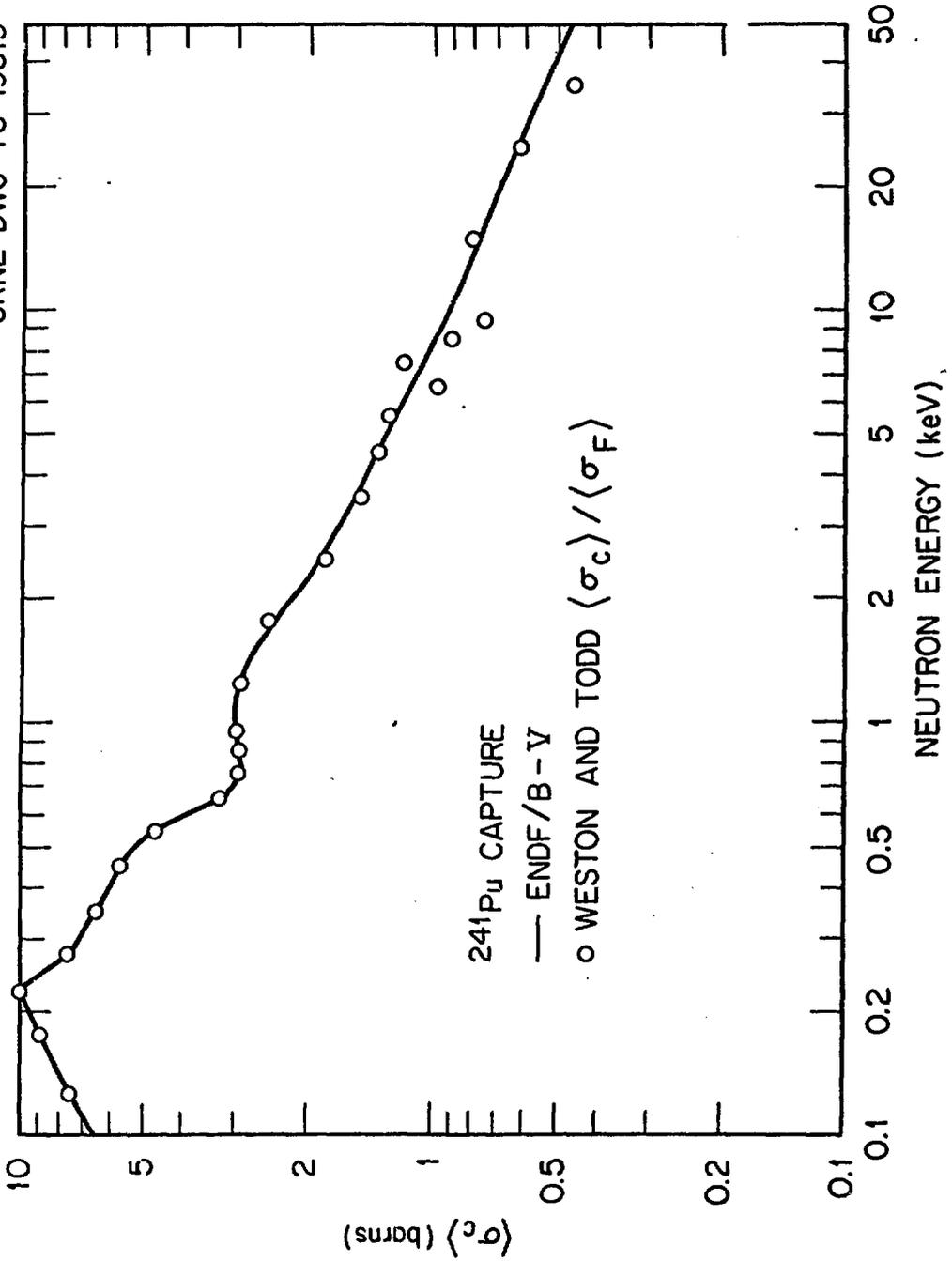


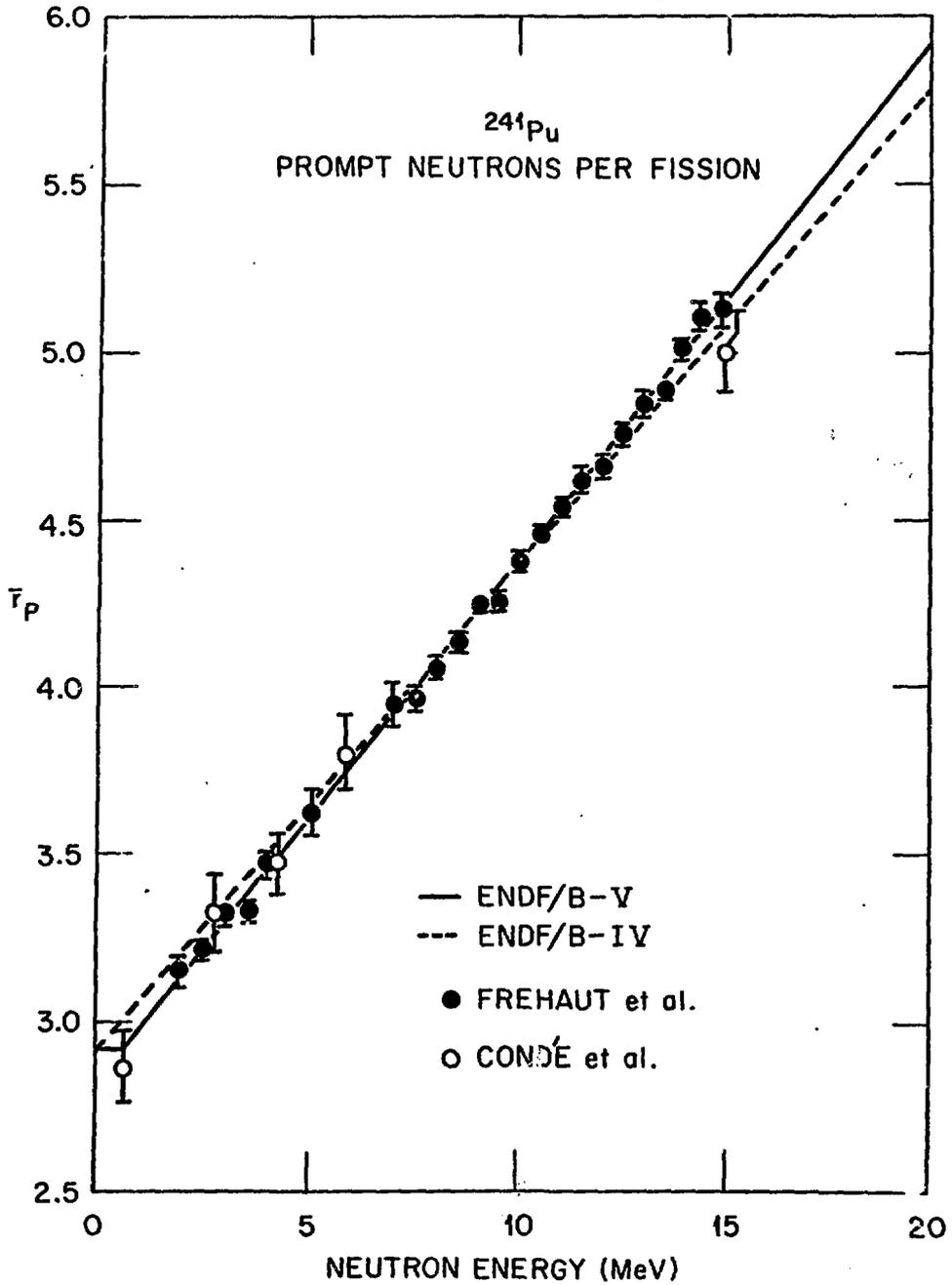


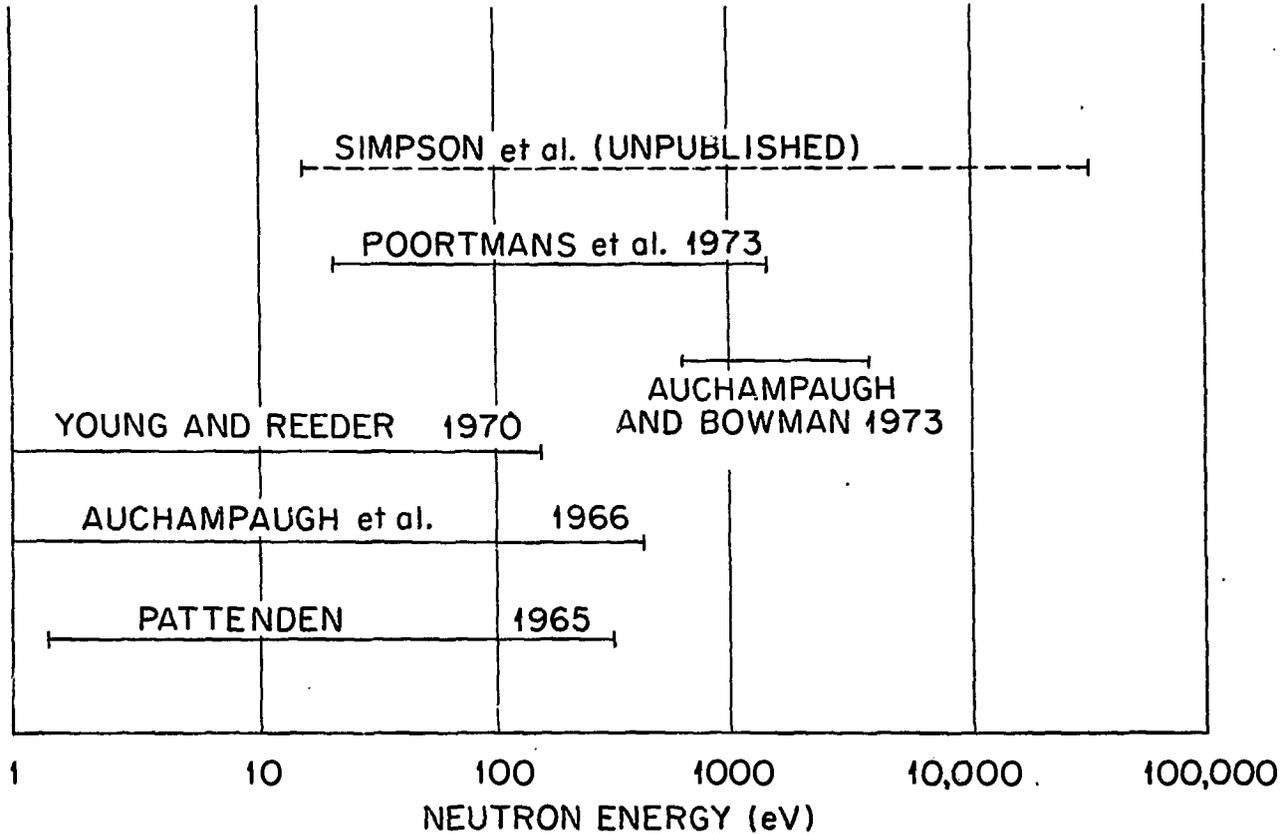


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^{242}Pu Total Cross Section Measurements in the Resolved Resonance Region.