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REVIEW OF FAST-NEUTRON CAPTURE CROSS SECTIONS OF THE HIGHER PLUTONIUM ISOTOPES AND Am-241*

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Abstract

The fast-neutron capture cross sections of Pu-240, 241, 242 and Am-241 are reviewed. These nuclides are important to core physics of reactors that contain Pu-239. There have been several significant measurements of these cross sections in recent years. These measurements were instigated by the need for these cross sections for reactor calculations involving high burn-up and build-up of the higher actinides. These recent measurements have satisfied the urgent need for these cross sections in the context of the accuracy needed relative to these of the major fissile isotopes. Problems that exist in the experimental measurements and their evaluation are discussed.

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I. Introduction

In recent years there has been emphasis on improved measurement and evaluation[1,2,3] of the capture cross sections of the higher plutonium isotopes and Am-241. This emphasis was brought about because of the production of these nuclides in both thermal and fast reactors. Reactor core physics as well as waste management are effected by these nuclides. Core physics is effected by the build-up of these actinides with high burn-up. The build-up of Pu-240,242 and Am-241 brings about parasitic neutron loss because these nuclides have relatively low fission cross sections. The Pu-241 has a high fission cross section and tends to compensate for the neutron loss to the other nuclides.[4] Waste management is effected because these nuclides are the paths to the build-up of the even higher mass actinides which have high rates of spontaneous fission.

There have been experimental measurements with LINACS as well as Van de Graaff accelerators as neutron sources for the capture cross sections of concern except for Pu-241 where there is only one LINAC measurement.[5] The experimental measurements are in reasonable agreement and their evaluations are converging towards consistency. The capture cross sections of these nuclides in the keV region of neutron energies is no longer a major problem. The foremost problems[6] in these nuclides are the discrepancy between integral and differential measurements of capture in the 1-eV resonance of Pu-240 and the shape of the capture and fission cross sections of Pu-241 in the thermal region of neutron energies.

The status of fast neutron capture in Pu-240,241,242, and Am-241 will be reviewed. The experimental and evaluation problems will be discussed. Emphasis will be given to the remaining difficulties in the evaluation of these cross sections.

II. Pu-240

Figure 1 illustrates the three experimental measurements of the fast neutron capture cross seciton for Pu-240. The data are plotted on an expanded scale and multiplied by the square root of the neutron energy so that the data can be compared in detail. The three measurements on Pu-240 used quite different techniques and they agree within the experimental uncertainties. Hockenbury, Moyer, and Block[7] used a large liquid scintillator tank to detect capture and fission events and a LINAC as a neutron source. Weston and Todd[8] used "total energy detectors" which are small liquid scintillators with pulse-height weighting to detect capture and fission events and a LINAC as a neutron source. Wissak and Kappeler[9] used Moxon-Rae detectors to detect capture and fission events and a Van de Graaff accelerator as a neutron source. The Wisshak and Kappeler data have been normalized downward by 3.4% from the published results in accordance with their recent normalization calculations.[10] The Wisshak and Kappeler data[9] were measured relative to Au-197 with a calculated absolute efficiency ratio. Part of the Wisshak and Kappeler data[9] was averaged and not all of the data are presented in Fig. 1. The ENDF/B-V evaluation of Au-197 capture,[11] which has structure in this neutron energy region, was used to convert the data to capture cross sections, so Fig. 1 should only be considered as representative of the Wisshak and Kappeler data.[9] The Hockenbury et al. data were normalized to their total cross section measurement in the resonance region and the neutron flux measured with a B-10 slab detector. The Weston and Todd data[8] were normalized at thermal and at the 1-eV resonance and the flux was measured with a parallel plate BF_3 ion chamber.

The evaluation of the fast capture data on Pu-240 presents the major problem since the experimental measurements are in reasonable agreement. The ENDF/B-V evaluation[12] is also illustrated in Fig. 1. This evaluation gave most weight to the Weston and Todd data since these data were in agreement with the average resonance parameters from the resonance region and the data of Wisshak and Kappeler[9] were preliminary at the time. The resolved resonance region extended to 3.9 keV and the shape from 1 to 3.9 keV was an attempt to reconcile the resonance parameters with the average capture cross section measurements.

Table 1 gives the average resonance parameters evaluated by Weigmann et al.[13] and recent unpublished results by Gwin.[14] The average resonance parameters of ENDF/B-V are (back in) those of Weigmann.[13] The unpublished results of Gwin[14] are from thick sample transmission measurements. The higher s-wave strength function of Gwin is in better agreement with the Weston and Todd[8] and

Hockenbury et al.[7] capture measurements in the 1- to 10-keV neutron energy region (see the dotted line in Fig. 1), however, the difference is within the uncertainties of the capture measurements.

The evaluation problem is that the high values of the capture cross sections at around 30 keV cannot be reproduced with the average resonance parameters from the resonance region with the normal accepted assumption.s It is usually assumed in fitting capture cross sections that the radiation width divided by the spacing is the same for p-wave neutrons as for s-wave neutrons. The dotted line in Fig. 1 is a good representation of the experimental data, however, to achieve this fit it had to be assumed that $\langle \Gamma_\gamma/D \rangle$ was ~40% higher for p-wave neutrons than for s-wave. This calculation was done with the program, UR, by Pennington[15] which is frequently used in ENDF calculations.

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There is no theoretical reason why the average radiation width, Γ_γ, in particular could not be higher for p-wave neutrons than for s-wave neutrons, however, there has been no clear case in other nuclides where there was a major difference. It is unlikely that the evaluated s-wave $\langle \Gamma_\gamma/D \rangle$ could be in error by 40% because the experimental uncertainties from the determination of this quantity in the resonance region is much smaller (~4%).

Both the Wisshak and Kappeler[9] and the Weston and Todd data are indicating a rapid decline in the capture cross section above 50 keV which would be due to competition with inelastic scattering from the first 2+ level in Pu-240 at 42.8 keV. The ENDF/B-V evaluation did not properly take this effect into account.

III. Pu-241

There has been only one reported measurement of the differential fast capture cross section for Pu-241 by Weston and Todd.[5] This was a measurement of the ratio of capture-to-fission in Pu-241 using "total energy detectors," fast neutron detectors and a LINAC as a neutron source. The data were normalized at thermal neutron energies. These results and the ENDF/B-V evaluation[16] are illustrated in Fig. 2. Since the ENDF Evaluation was based on these experimental data, the agreement yields no insight. The experimental data and evaluation are reasonable, however, it is unfortunate that there is not an additional experimental measurement. The measurement of this cross

Kappeler measurements [22] and the Gather and Thomas measurements [23] have a higher average cross section than the Weston and Todd measurements.[24] The Gather and Thomas measurements [23] have a preliminary normalization to the Weston and Todd measurements [24] between 1 and 2 keV.

The gather and Thomas measurements [23] used a large liquid scintillator tank to detect capture, a Li-glass scintillator at 30 keV and a U-235 fission detector at >30 keV to measure the shape of the neutron flux, and a LINAC as a neutron source. The normalization is preliminary. The Wissak and Kappeler measurements [22] used the same techniques as discussed for their Pu-240 measurements. Their data shown in Fig. 4 have been normalized downward by 2.7% as suggested by Wisshak and Kappeler in a normalization calculation to be published.[10] All of the Wisshak and Kappeler data is not shown in Fig. 4 and part of their data are averaged so that the plot should be regarded as only indicative of the data. Also the ENDF/B-V Au-197 capture evaluation [11] which has structure in this neutron energy region was used to convert the ratio data to the capture cross section of Am-241.

The ENDF evaluation [25] by F. M. Mann *et al.* was based on the average resonance parameters from the resolved resonance region and the Weston and Todd [24] measurements. The other measurements were not complete at the time of the ENDF evaluation. It has been shown by Derrian *et al.* the higher average capture cross section indicated by the other measurements can be reproduced by evaluated average resonance parameters from the resonance region and a reasonable p-wave strength function of (2.54 x 10^{-4}).

Recent unpublished results by Anderl, Schroeder, and Harker [26] concerning an integral measurement in CFRMF, which has a peak sensitivity at about 300 keV, indicate that Am-241 capture should be about 16% higher than the ENDF evaluation. As can be seen in Fig. 4 the results of Gather and Thomas [23] would be quite consistent with these results which would also be within the experimental uncertainties of the Weston and Todd measurements.[24] The recent results of Anderl, Schroeder, and Harker,[26] thus, represent no discrepancy with the previously measured results.

For reactor physics, it is important whether Am-241 capture leads to the ground state of Am-242 or the excited state, Am-242m. The ground state has a half-life for beta decay to Cm-244 of only 16 hours whereas Am-242m has a half life of 152 years. There are recent measurements and theoretical calculations of this ratio by Wisshak, Wickenhauser, and Kappeler.[27] these theoretical calculations and those by others [28] are within experimental uncertainties, however, the experimental measurement [27] indicates an appreciably stronger neutron energy dependance.

section is difficult because of the relatively short half-life of Pu-241 (14.8 y), the build up of Am-241 by beta decay, and the associated gamma-ray activity.

Since Pu-241 has a large fission cross section and exhibits intermediate structure, describing the average capture cross section with average resonance parameters is complex. The available capture cross section data can be fitted with reasonable average resonance parameters as was done for ENDF/B-V as well as other evaluations.[17,18] Because the average capture can be fit with reasonable resonance parameters, capture is small compared to fission, and the experimental difficulty of this capture measurement, the need for an additional measurement is doubtful.

IV. Pu-242

Figure 3 illustrates the two experimental measurements of the fast capture cross section of Pu-242. The agreement of the Hockenbury et al.[19] and the Wisshak and Kappeler data[5,10] is well within the experimental uncertainties. The ENDF evaluation[20] by F. Mann and others was based on the average resonance parameters from the resonance region and the measurements shown in Fig. 3. Other evaluations have described the experimental data quite well.[21] Since there is consistency between the experimental measurements of the capture cross sections of Pu-242, there appears to be no outstanding problems in the fast capture cross section of Pu-242.

V. Am-241

The experimental measurements and the ENDF evaluation for fast neutron capture by Am-241 are illustrated in Fig. 4. The experimental measurements are in agreement within experimental uncertainties, however, the Wisshak and

The fast neutron capture cross section of Am-241 is reasonably well known in the context of the uncertainty weighted by the occurrence in fuel elements as compared to the capture cross section of U-235 and Pu-239. The agreement of the experimental data is marginally within experimental uncertainties and the ENDF/B-V evaluation probably needs revision, however, these difficulties probably do not warrant additional experimental measurements.

VI. Conclusions

The status of fast neutron capture measurements for Pu-240,241,242, and Am-241 has been reviewed. There appear to be no outstanding discrepancies to warrant a recommendation for additional measurements at this time unless an appreciably more accurate experimental technique is developed. If such a technique is developed, it should be first applied to Pu-239 rather than the higher nuclides.

The ENDF/B-V evaluations are not very consistent with the presently available data for Pu-240 and Am-241, however, this is an evaluation problem rather than an experimental measurement problem. Thus the status of the fast capture cross sections of the higher actinides which are important to core physics as well as waste disposal which were in alarmingly poor condition a number of years ago are now reasonably well known as compared to the cross sections of the major fissile nuclei.

Table 1. Average Resonance Parameters for Pu-240.

	$\Gamma_N^o/D(x10^{-4})$	$\bar{D}$ (eV)	Γ_γ (eV)	$\Gamma_n^1/D(x10^{-4})$
Weigman et al.	$1.04^{+0.14}_{-0.12}$	12.7 ± 0.3	30.8 ± 1	2.2 ± 0.2
Gwin et al.	1.20 ± 0.04			2.2 ± 0.11

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$\Gamma_n/D = 1.2$ [illegible]
$\Gamma_n/D = 2.2$ Γ_γ/D + 40%

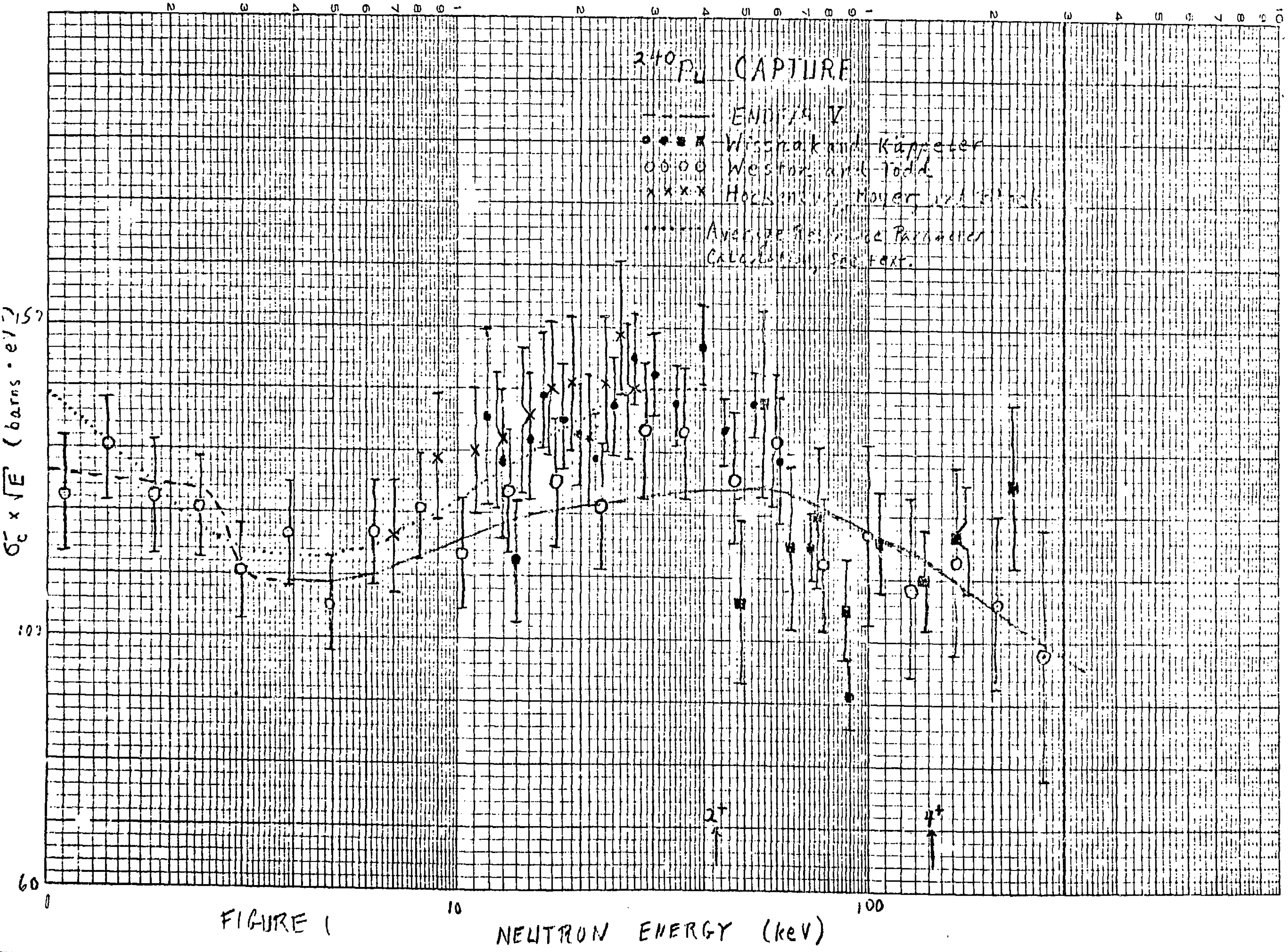

FIGURE 1

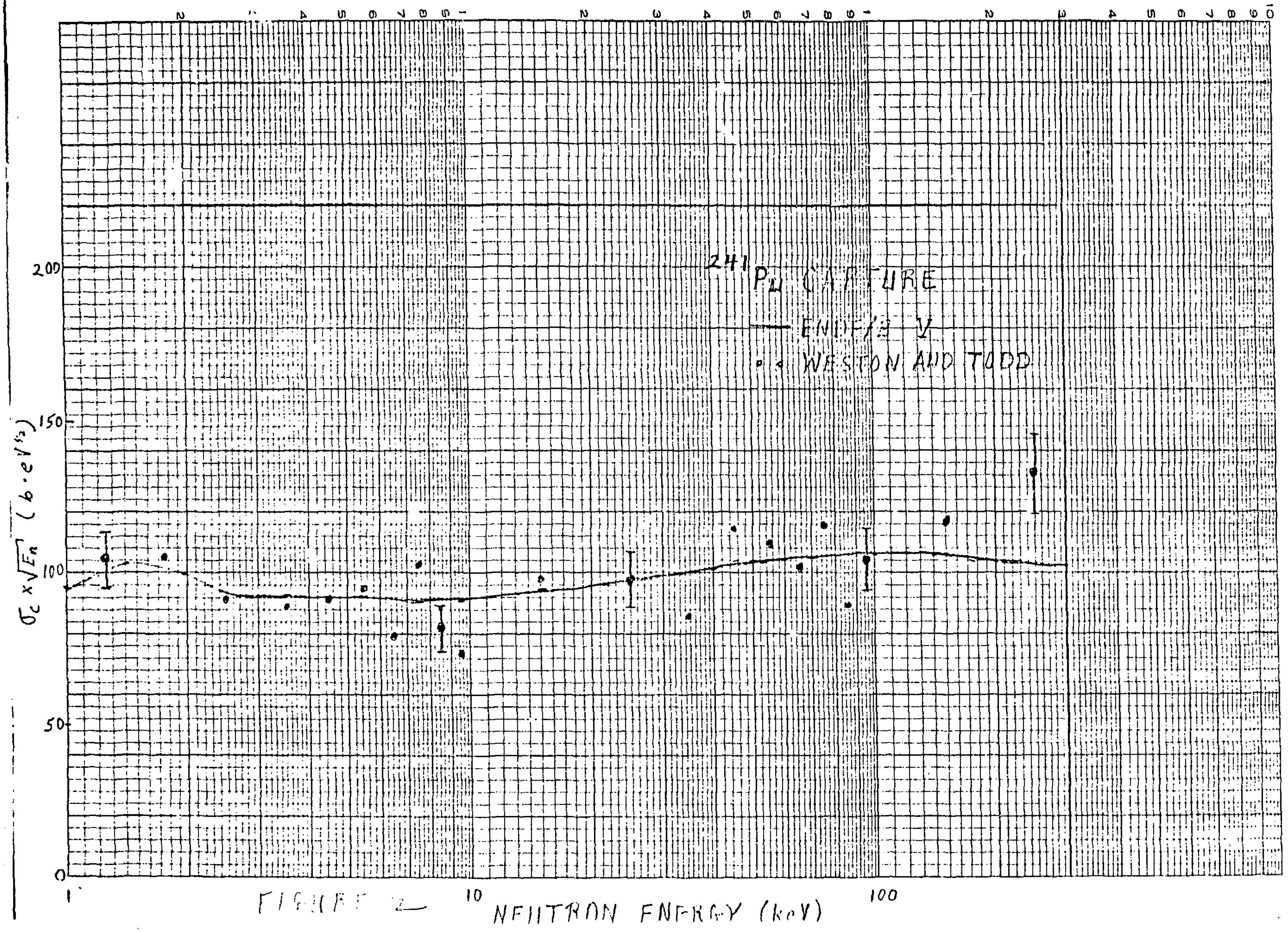
241Pu CAPTURE
— ENDF/B V
• WESTON AND TODD
200
150
100
50
0
σc × √En (b · eV½)
1
10
100
NEUTRON ENERGY (keV)

FIGURE 2

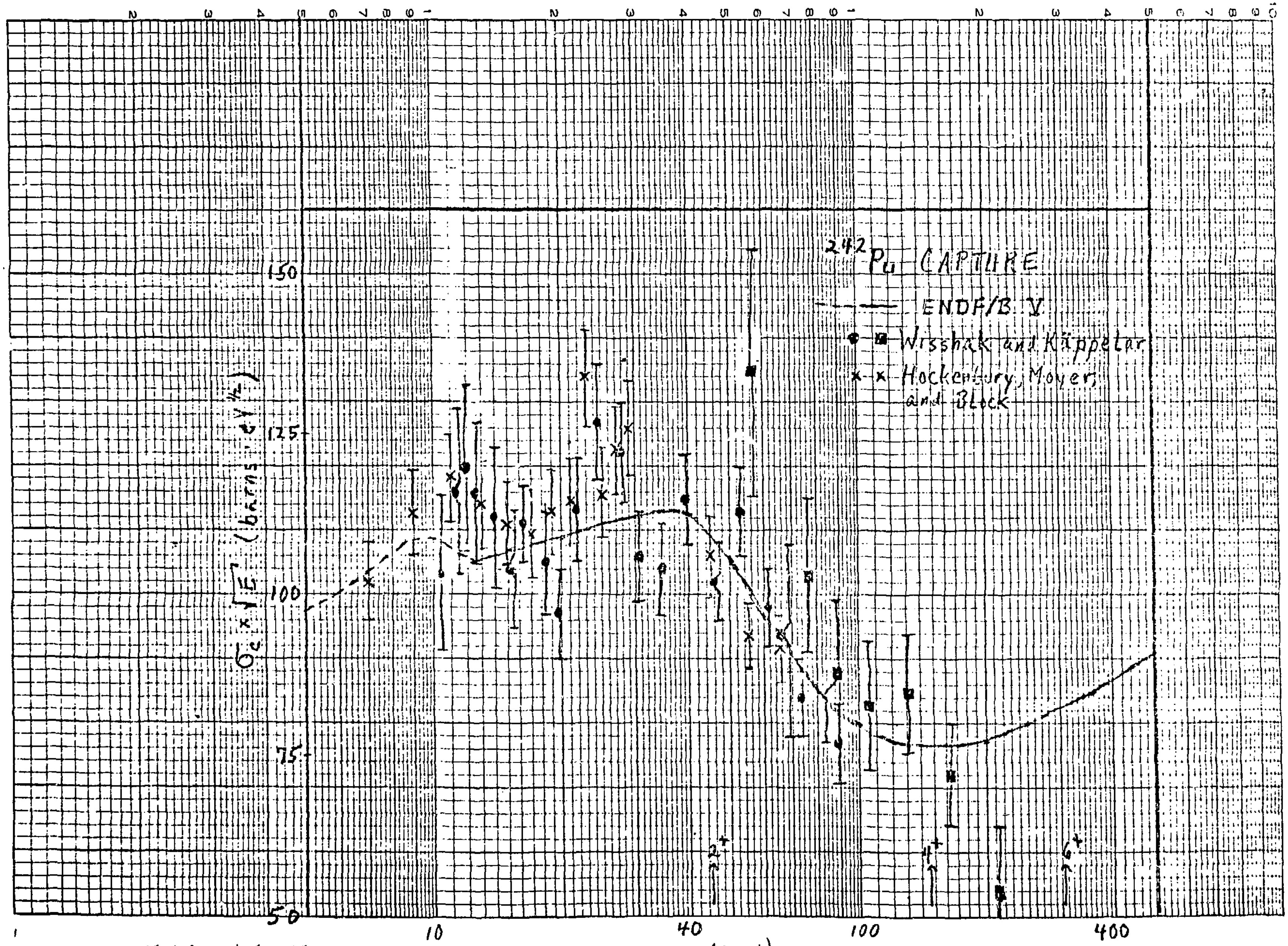
^{242}Pu CAPTURE
ENDF/B V
● ■ Wisshak and Käppelar
× × Hockenbury, Moyer, and Block
$\sigma_c \times \sqrt{E}$ (barns · eV$^{1/2}$)
150
125
100
75
50
1
10
40
100
400
2^+
4^+
6^+
NEUTRON ENERGY (keV)

FIGURE 3

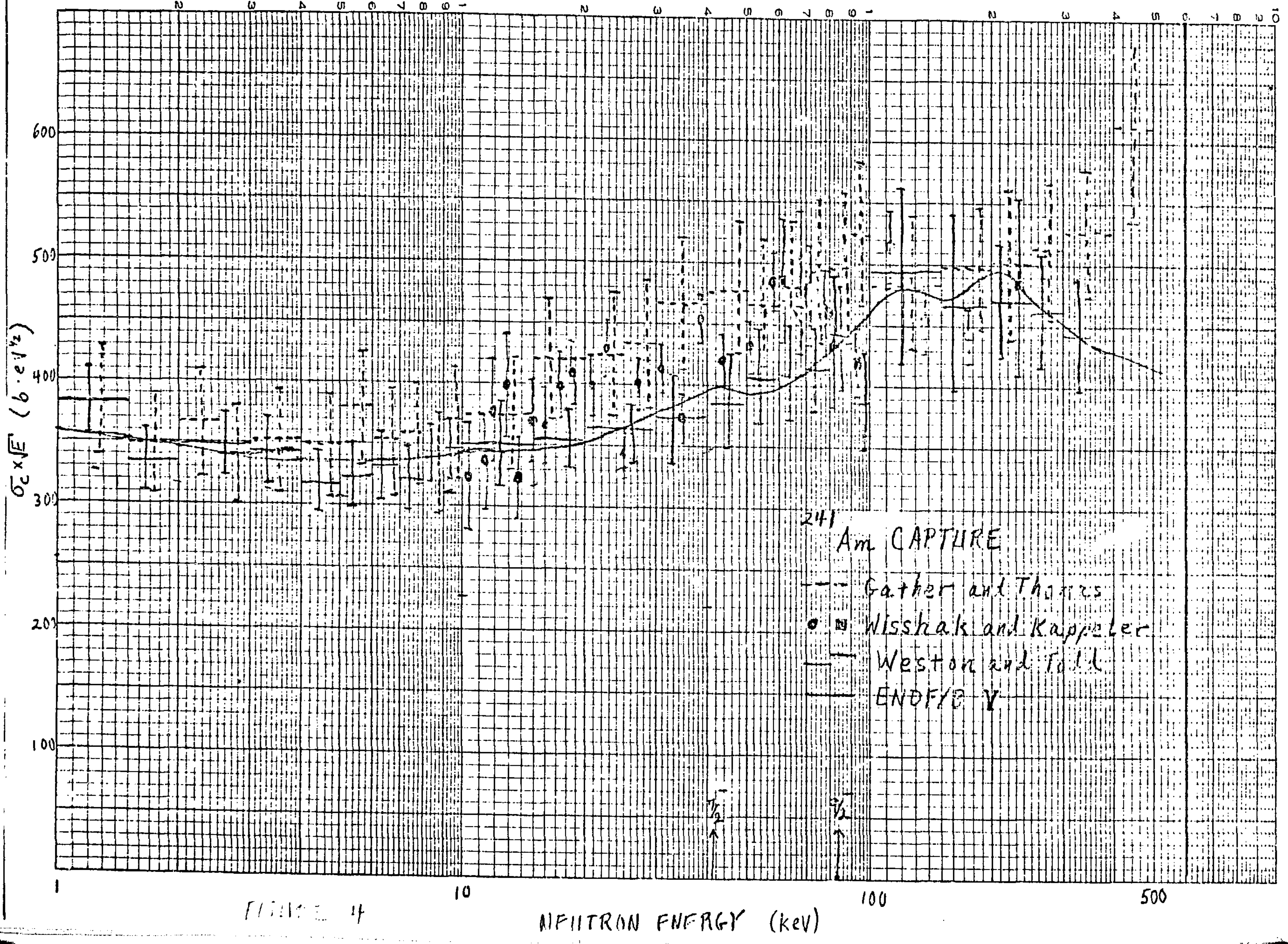
$\sigma_c \times \sqrt{E}$ $(b \cdot eV^{1/2})$
600
500
400
300
200
100
1
10
100
500
NEUTRON ENERGY (keV)
^{241}Am CAPTURE
Gather and Thomas
Wisshak and Kappeler
Weston and Todd
ENDF/B V
$T_{1/2}$
$T_{1/2}$

FIGURE 4