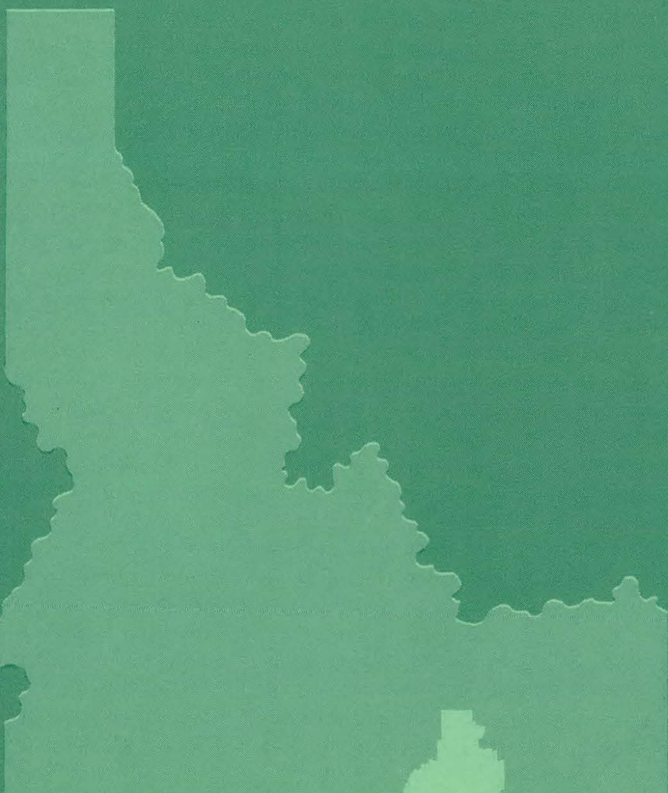
IDO-16797

MASTER

RADIOCHEMICALLY DETERMINED RATIOS OF ASYMMETRIC TO SYMMETRIC FISSION OF U^{233}, Pu^{239}, and U^{235} AS A FUNCTION OF NEUTRON ENERGY

W. H. Burgus



PHILLIPS PETROLEUM COMPANY

ATOMIC ENERGY DIVISION

NATIONAL REACTOR TESTING STATION

US ATOMIC ENERGY COMMISSION

DISCLAIMER

PRICE $ .50

Available from the
Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

Printed in USA

IDO-16797
AEC Research and Development Report
Physics
TID-4500 (17th Ed.)
Issued: July 30, 1962

RADIOCHEMICALLY DETERMINED RATIOS OF ASYMMETRIC TO SYMMETRIC FISSION OF U^{233}, Pu^{239}, and U^{235} AS A FUNCTION OF NEUTRON ENERGY

by

W. H. Burgus

This report was prepared for presentation as a fifteen minute invited paper at a Conference on Nuclear Chemistry - Fission and Other Low Energy Nuclear Processes, sponsored by the U. K. Atomic Energy Research Establishment, Harwell, and held at Oxford University, September 18-20, 1962.

PHILLIPS
PETROLEUM
COMPANY

Atomic Energy Division
Contract AT(10-1)-205
Idaho Operations Office
U. S. ATOMIC ENERGY COMMISSION

TABLE OF CONTENTS

RADIOCHEMICALLY DETERMINED RATIOS OF ASYMMETRIC TO SYMMETRIC FISSION OF U^{233}, Pu^{239}, and U^{235} AS A FUNCTION OF NEUTRON ENERGY

by

W. H. Burgus

I. INTRODUCTION

After the announcement of the Bohr model[1] of fission, a number of detailed implications of this model were published. Among these was the suggestion[2] of J. A. Wheeler that the amount of symmetric fission (i.e., fission giving two fragments of equal mass) should be different for the two possible spin states of the compound system formed upon the addition of a low energy neutron to the target nucleus. A straight-forward way to test this prediction seemed to be the radiochemical measurement of the asymmetric/symmetric ratio for fission induced by neutrons of energies corresponding to various fission resonances. There is evidence based on theoretical analyses of the fission cross sections, that certain of these resonances are associated with one or the other of the two possible spin states. The purpose of this paper is to describe the results of these studies carried out with monoenergetic neutrons provided principally by the Materials Testing Reactor (MTR) Crystal Spectrometer, but also in a few experiments with very low-energy neutrons provided by the MTR Cold Neutron Facility.

II. EXPERIMENTAL

We chose to begin these experiments with U^{233}. A multilevel analysis[3] of the U^{233} cross section (Figure 1) indicated that a very large fraction of the fission cross section at thermal energies arises from nuclei of one spin state of the compound system, while the prominent resonances of 1.8 and 2.3 ev are associated with the other spin state. In addition the broad resonance at 4.7 ev was thought to belong to the same spin state as that which predominates at thermal energies. The experimental approach then was to irradiate U^{233} samples with thermal neutrons and with monoenergetic neutrons of 1.8, 2.3, and 4.7 ev. After irradiation the samples were radiochemically analyzed for fission products such as Mo^{99} (an indicator of asymmetric fission) and Ag^{111}, Ag^{113}, and Cd^{115} (representatives of symmetric fission). Since it was only necessary to compare the peak-to-valley ratio for thermal fission with the same ratio for resonance fission, determinations of absolute yields were not required. It was sufficient to compare the saturated activities of the various fission products (which are, of course, directly proportional to the absolute fission yields), each product counted in a standard reproducible manner.

The comparisons of the ratio of asymmetric to symmetric fission were made in terms of a quantity "R" defined as follows:

$$R = \frac{A' \text{ asy}/A' \text{ sym}}{A \text{ asy}/A \text{ sym}} \qquad \text{(Figure 2)}$$

where A is a saturated counting rate for any nuclide, subscript asy denotes an asymmetric (peak) fission product such as Mo^{99}, and subscript sym a symmetric (valley) fission product such as Ag^{111}. The primed activities refer to resonance fission and the unprimed to thermal fission. Briefly, R is the peak-to-valley ratio for resonance fission divided by the peak-to-valley ratio for thermal fission.

Before discussing the data it may be well to mention a few of the difficulties involved in these experiments. First of all, one should mention the low fission-product activities to be measured. During spectrometer runs the fission rates were only about 10^6 to 10^7 fissions per minute. Figure 3 shows the fission rates and neutron energy resolutions associated with these experiments. In order to obtain measurable fission product samples it was necessary to sacrifice neutron energy resolution in favor of obtaining higher neutron intensities. Irradiations ran from about 75 to 100 hours with spectrometer neutrons. Typical valley element samples only counted of the order of 10^2 counts per minute and usually decay curves had to be resolved. Therefore, low background counting was necessary. The unirradiated U^{233} contained some U^{232} so that unusually good radiochemical purification procedures were required since the U^{232} decay products were several orders of magnitude more active than the fission products. We turn now to the data (Figure 4).

Despite the low counting rates and other difficulties, we were, for the first time, able to demonstrate that there were observable differences between the peak-to-valley ratios at the 1.8-and 2.3-ev resonances (where one spin state is involved) and thermal energy (where the analysis had indicated that the other spin state was involved). We would, of course, have preferred to investigate several of the higher energy resonances but with our present spectrometer we had neither the required intensity nor resolution to do this.

Although the effects observed with U^{233} were well established, one had the feeling when talking with physicists that they had rather hoped for a larger effect. By this time, also, the first simple experiments with U^{235} using epi-cadmium neutrons had been published by the Los Alamos group[4]. The effects noted were again in the 20% range. Furthermore a contradictory experiment employing direct measurement with a double-sided ionization chamber had been reported by Roeland, Bollinger, and Thomas[5]. Also, U^{235} radiochemical experiments, similar to ours with U^{233} had been conducted by Nasuhoglu, et al[6] and no experimentally significant effects were noted. Similar radiochemical experiments with U^{235} had also been undertaken at Brookhaven but these had been abandoned because of insufficient intensities of spectrometer neutrons. By this time we had also learned that the Los Alamos spinning-wheel time-of-flight experiment had been planned for U^{235}. So, believing that there was sufficient effort being devoted to U^{235}, we turned our attention then to Pu^{239}. This turned out to be a fortunate choice as we will presently see.

Figure 5 shows a portion of a plot of the Pu^{239} fission cross section versus energy. Note that there is a prominent resonance peaking at 0.297 ev and that the next resonance is out at nearly 8 ev. Our monoenergetic neutron experiments were limited to the region of the first resonance and below. Again as in the U^{233} experiments, resonance and thermal fission were compared in terms of the value "R".

In these experiments, however, the "valley" nuclides examined were Cd^{115}, Sn^{121}, and Sn^{125}. The position of these on the Pu^{239} fission yield curve are shown in Figure 6. Figure 7 shows the very low energy portion of the fission cross section curve on an expanded scale with the points indicating energies in which our data were taken. Slide 8 presents the "R" value data.

In Figure 8, note first of all that at 0.297 ev (peak of the resonance), the "R" values for Cd^{115} and Sn^{121} are on the order of three. This compares with values of 1.2 and 1.4 observed with U^{233} and U^{235}. In other words, in Pu^{239} fission, the change in the peak-to-valley ratio is an order of magnitude greater than with U^{233} and U^{235}. Note also that the effect as measured with Sn^{125} is smaller than with the other two isotopes. This is perhaps understandable because Sn^{125} is several mass numbers removed from completely symmetric fission while Cd^{115} and Sn^{121} are produced in fission events which may be described as "close to completely symmetric fission." The Sn^{125} is therefore a less sensitive indicator of change in the peak-to-valley ratio.

Returning to Figure 7 for a moment, one can see that the fission cross section of Pu^{239} is quite high in the thermal region and this cannot be accounted for by contributions from the first (or higher energy) resonances. In the multilevel analysis of Vogt[(7)] a negative energy resonance (at -1.200 ev) is invoked to account for the high thermal cross section and also to account for other features of the cross-section curve. Furthermore, Vogt has deduced that this bound level is not required to interfere with and thus presumably does <u>not</u> have the same spin as the 0.297-ev level. Bearing this in mind we have calculated the contribution of the 0.297-ev level to the fission cross section at five different energies at which "R" values were measured. With the respective contributions to fission cross section as weighting factors, with the peak-to-valley ratio normalized to 1.00 for fission occurring in the bound level, and with an "R" value of 3.2 taken from the data at the peak of the 0.297-ev resonance, we calculate that the asymmetric/symmetric ratio characteristic of the 0.297-ev level is 5.3 times that ratio characteristic of the bound level! This indeed is a sizeable effect and is the largest yet observed.

With this information concerning the peak-to-valley ratios assumed characteristic of each of the two different symmetry classes of Pu^{239} fission, we calculated "R" values at the various energies at which experiments were run. These are compared with the measured values in Figure 9. The particular data to which I wish to call your attention is that for 0.06 ev. Here the calculated "R" value is 1.21. The measured values for Mo^{99}/Cd^{115} and Mo^{99}/Sn^{121} ratios are 1.33 ± 0.06 and 1.26 ± 0.05 respectively. All three values are the same within the errors of the assumptions and the experiments, and demonstrate the consistency of this approach. The important thing to remember about Pu^{239} is that the peak-to-valley ratios for the two classes of fission differ by a factor of more than 5.

Turning now to the case of U^{235}, there are two very significant experiments that must be mentioned in any discussion of radiochemically-determined U^{235} peak-to-trough ratios. The first of these is the well-publicized "spinning wheel" experiment of the Los Alamos group(8) in which a nuclear explosion and the time-of-flight technique were used for resonance activation of U^{235}. In this work a sizeable number (29) of resonances were definitely assigned to two groups, one having a smaller (R = 0.715) and one having a larger (R = 1.05) peak-to-valley ratio than is observed in thermal fission of U^{235}. For U^{235} this particular behavior had been predicted by Wheeler[2] on the basis of the symmetry properties of the wave function at the saddle point which would permit relatively less symmetric fission for the 3^- spin state than for the 4^- spin state. (We have recently heard indirectly that the Los Alamos measurements have been repeated some four years later with confirming results.) However in another important piece of work with U^{235} (that of Cunninghame, et al[9]), the argument of Wheeler based on symmetry considerations was shown to be incorrect, since it had predicted relatively more symmetric fission for p-wave neutrons than for s-wave neutrons and actually the opposite was observed!

Our own peak-to-valley experiments with U^{235} have been concerned only with the lower energy portion of the cross-section curve. Figure 10 is a plot of U^{235} cross section in the region below 1.0 ev. Only one resonance, peaking at 0.284 ev, is observed. The next resonance is at 1.1 ev. The dash line shows the resonance and background remaining after subtraction of contributions from the other known resonances. To "fit" the cross-section data in this region Shore and Sailor[10] have postulated two negative energy resonances at -0.02 ev and -1.45 ev which do not interfere with the 0.284-ev resonance and may therefore be of the opposite symmetry type. In another analysis, that of Vogt[7], this background cross section is taken care of by assuming a -0.95-ev resonance which does interfere with the first positive (0.284 ev) energy resonance and therefore is presumably of the same symmetry type.

Figure 11 shows the U^{235} fission cross-section curve along with our experimentally determined "R" values of several energies. A decided increase in symmetric fission is seen in the region of the 0.284-ev resonance where "R" falls to 0.74. Since only about 42% of the fission cross section at this energy is due to this particular resonance, the actual increase in symmetric fission (decrease in "R") could be much greater if contributions from the opposite symmetry group were not involved.

The "R" value increases as symmetric fission becomes less probable near thermal energy and then unexpectedly decreases again! The latter trend is not explainable on the basis of the assumptions of Vogt or of those of Shore and Sailor concerning the occurrence of negative energy levels. Thus another anomaly has been introduced in the U^{235} peak-to-valley ratio data.

One further group of experiments must be mentioned in discussing peak-to-valley ratios although these experiments were not done radiochemically. The work to which reference is made is that of Gibson, Thomas, and Miller(11) and that of Milton and Fraser(12). By direct physical measurements, each of these groups has unquestionably demonstrated that in low-energy fission there is a dramatic drop in the kinetic energy of the symmetric fission products (as compared to asymmetric fission events) with presumably a corresponding increase in the total excitation energy of the two symmetric fragments. One interpretation of these observations is that the final division into fission fragments is almost entirely dependent upon the excitation energy available to the two fragments at the "scission point" which time-wise is somewhat later than the "saddle point."

III. SUMMARY

In summary, the following observations may be made concerning fission symmetry in the low-energy region:

1. The asymmetric to symmetric fission ratio for neutron-induced fission in the low-energy resonance region does change from resonance to resonance.

2. The ratios appear to fall into two symmetry classes each of which has a characteristic peak-to-valley mass yield ratio.

3. The largest observed difference between the two symmetry classes is found in Pu^{239}.

4. There is very probably a spin dependence involved in the mass distribution as predicted by the Bohr model. The exact nature of this dependence is not clearly understood. This is in part because many of the resonances that have been examined have not been unambiguously assigned spins. Also the apparent conflict of the Cunninghame p-wave neutron data with theoretical predictions has not been resolved.

5. Determinations of peak-to-valley ratios are useful from the practical standpoint since reactors operating at very high temperatures will have "thermal" energies in the resonance energy region. From the theoretical standpoint these determinations are also important since they may be used to support analyses of cross-section data and also may provide to some extent, at least, confirmatory evidence of the validity of the fission models.

IV. REFERENCES

1. A. Bohr, "Proceedings of the International Conference on the Peaceful Uses of Atomic Energy", United Nations, New York, 1956, Vol. 2, p. 151.
2. J. A. Wheeler, Physica 22, 1103 (1956).
3. M. S. Moore and C. W. Reich, Phys. Rev. 118, 718 (1960).
4. Los Alamos Radiochemistry Group, Phys. Rev. 107, 325 (1957).
5. Roeland, Bollinger, and Thomas, Bull. Am. Phys. Soc. Ser. II, 2, 352 (1957).
6. Nasuhoglu, Raboy, Ringo, Glendenin, and Steinberg, Phys. Rev. 108, 1522 (1957).
7. E. Vogt, Phys. Rev. 118, 724 (1960).
8. Cowan, Turkevich, Brown, et al, Phys. Rev. 122, 1286 (1961).
9. Cunninghame, Kitt and Rae, Nuc. Phys. 27, 154 (1961).
10. F. J. Shore and V. L. Sailor, Phys. Rev. 112, 1 (1958).
11. Gibson, Thomas, and Miller, Phys. Rev. Letters 7, 65 (1961).
12. J. C. D. Milton and J. S. Fraser, Phys. Rev. Letters 7, 67 (1961).

Some of the experimental work carried out at the MTR has been published in the following references:

R. B. Regier, W. H. Burgus, R. L. Tromp, and B. H. Sorensen, Phys. Rev. 119, 2017 (1960)

R. B. Regier, W. H. Burgus, and R. L. Tromp, Phys. Rev. 113, 1589 (1959)

K. T. Faler and R. L. Tromp, Trans. of the Amer. Nuc. Soc. 5, 22 (1962)

V. FIGURES

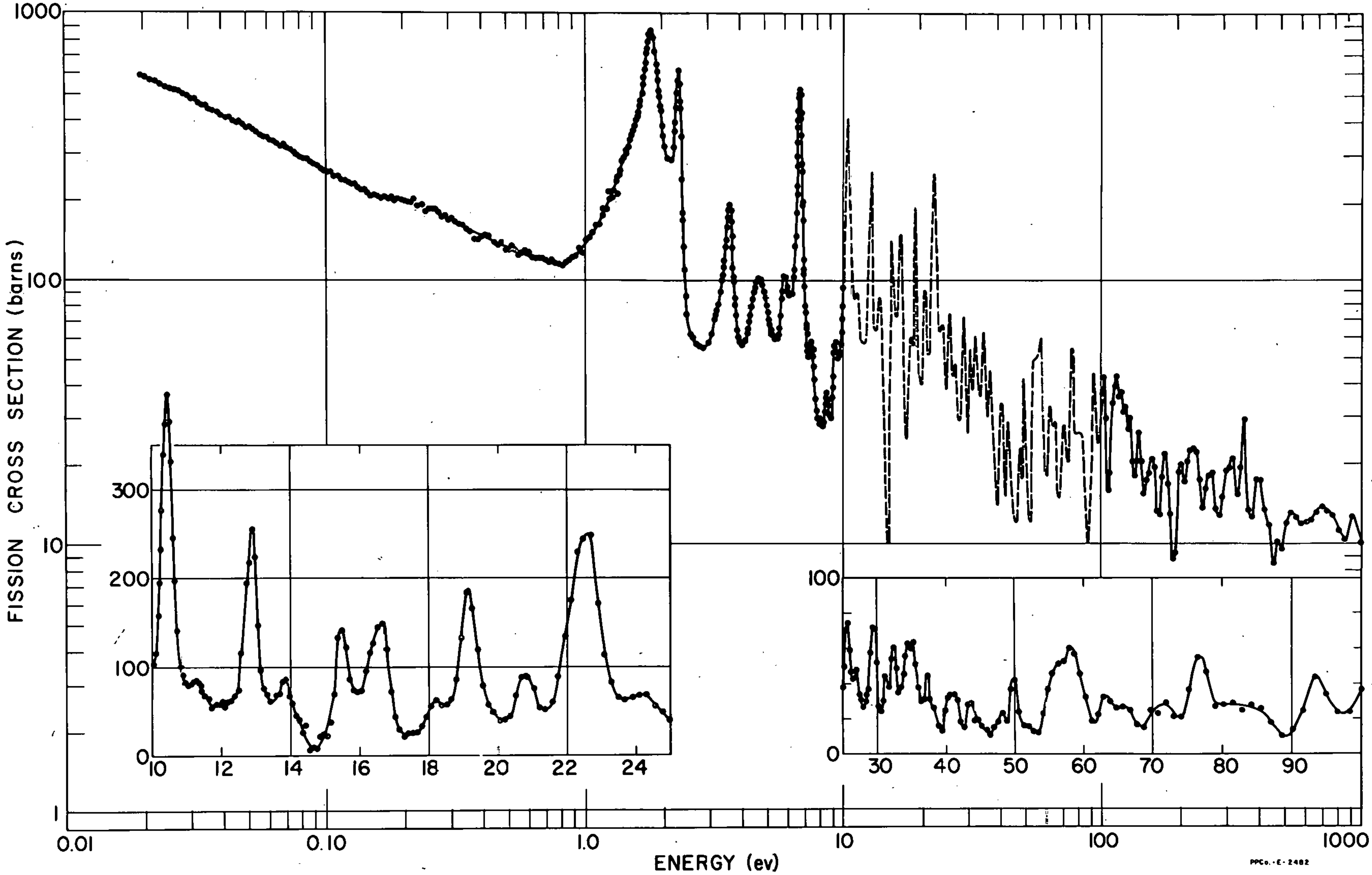


Fig. 1 U^{233} Fission Cross Section

$$R = \frac{A'_{asy} / A'_{sym}}{A_{asy} / A_{sym}}$$

P P Co.-B-4290

Fig. 2 Definition of the Quantity "R"

FLUX AND RESOLUTION

Energy	Fissions /sec / gm U-233	Resolution ($\Delta E/E$)
0.0253 ev	6×10^5	~ 2%
1.8	1.7×10^5	~ 16%
2.3	6.7×10^4	~ 18%
4.7	1.8×10^4	~ 25%

P P CO.-A-2927

Fig. 3 Fission Rates and Neutron Energy Resolutions in U^{233} Experiments

COMPARISON OF THERMAL AND RESONANCE FISSION OF U-233

Neutron Energy	R, Indicated By	
	Mo-99/Ag-111	Mo-99/Cd-115
1.8 ev	1.217 ± 0.038	—
2.3	1.200 ± 0.058	1.415 ± 0.103
4.7	0.957 ± 0.022	1.031 ± 0.168
All epi-Cd	1.088 ± 0.023	1.200 ± 0.052

PP CO.-A-2925

Fig. 4 U^{233} "R" Value Data

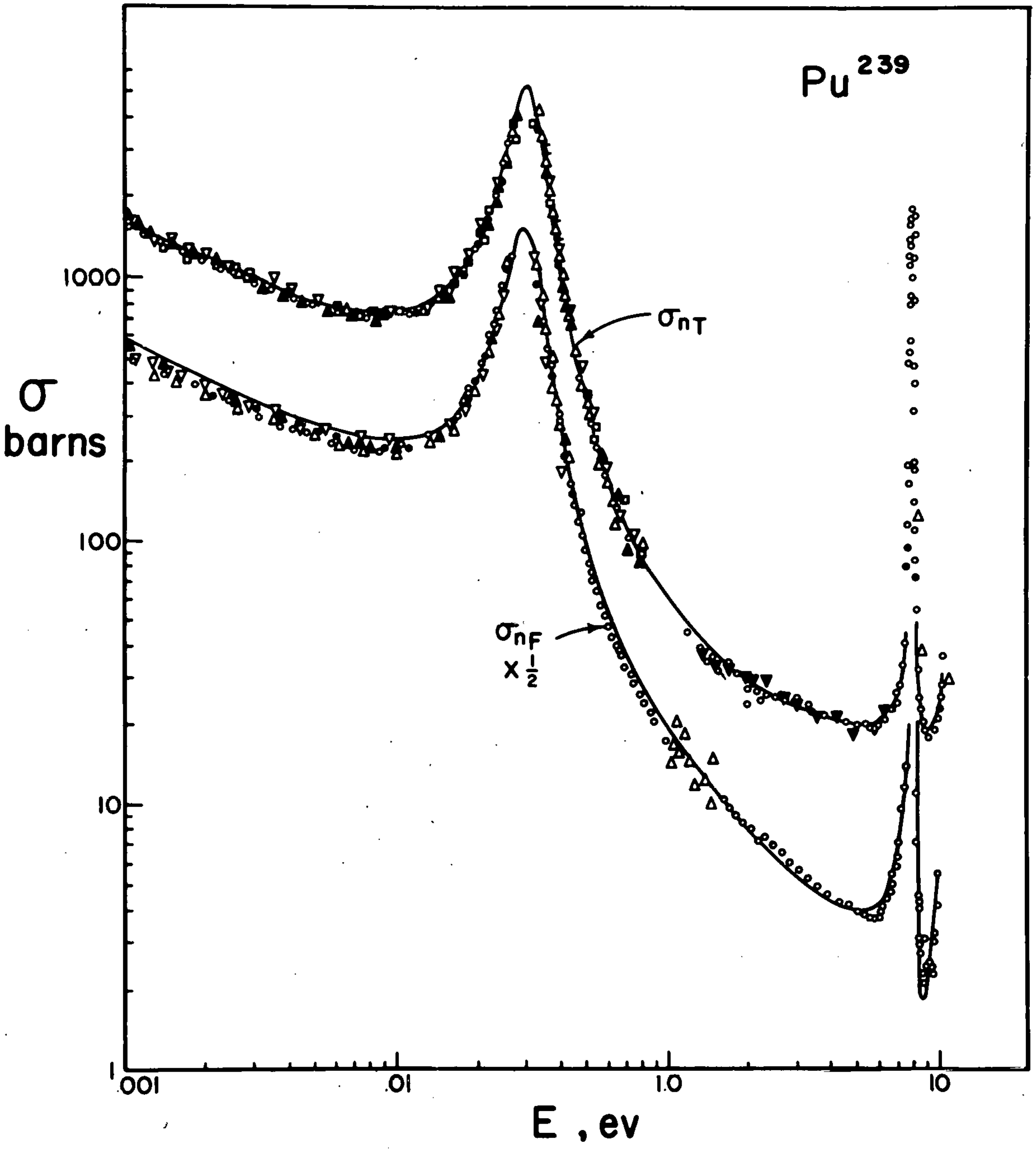


Fig. 5 Pu^{239} Fission Cross Section Curve

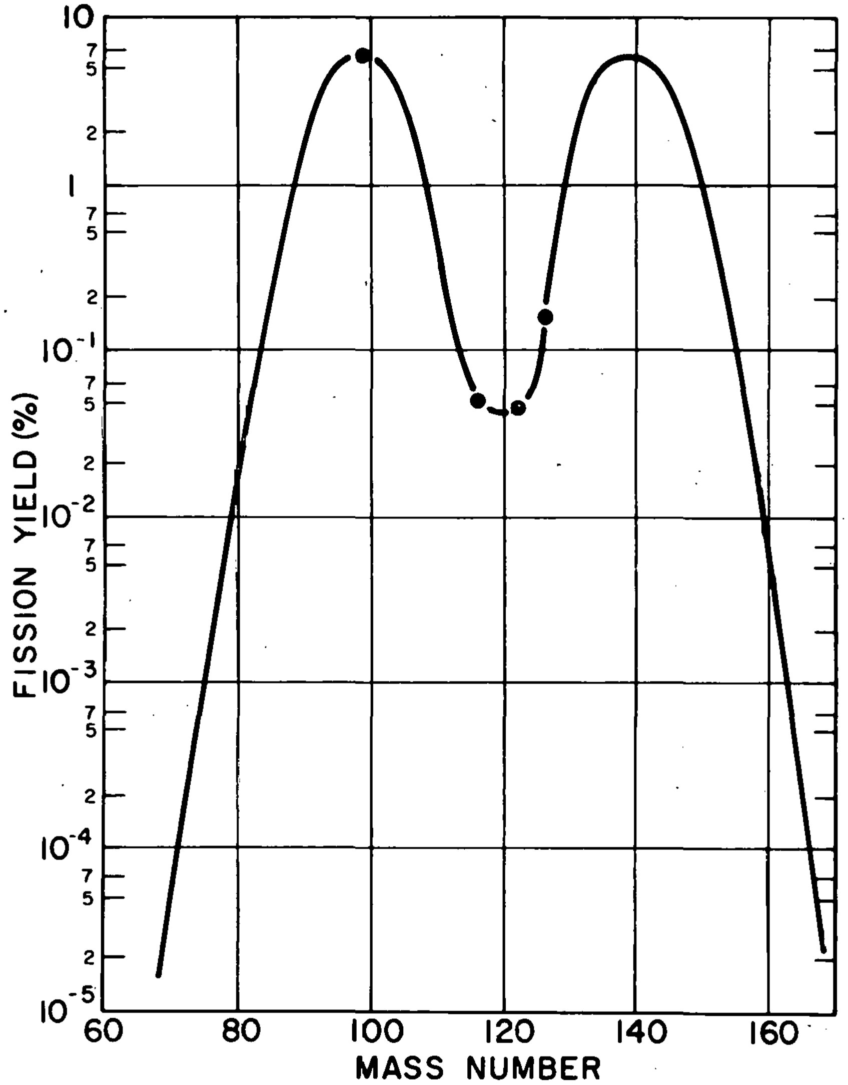


Fig. 6 Pu^{239} Fission Yield Curve Showing Positions of Valley Nuclides Investigated

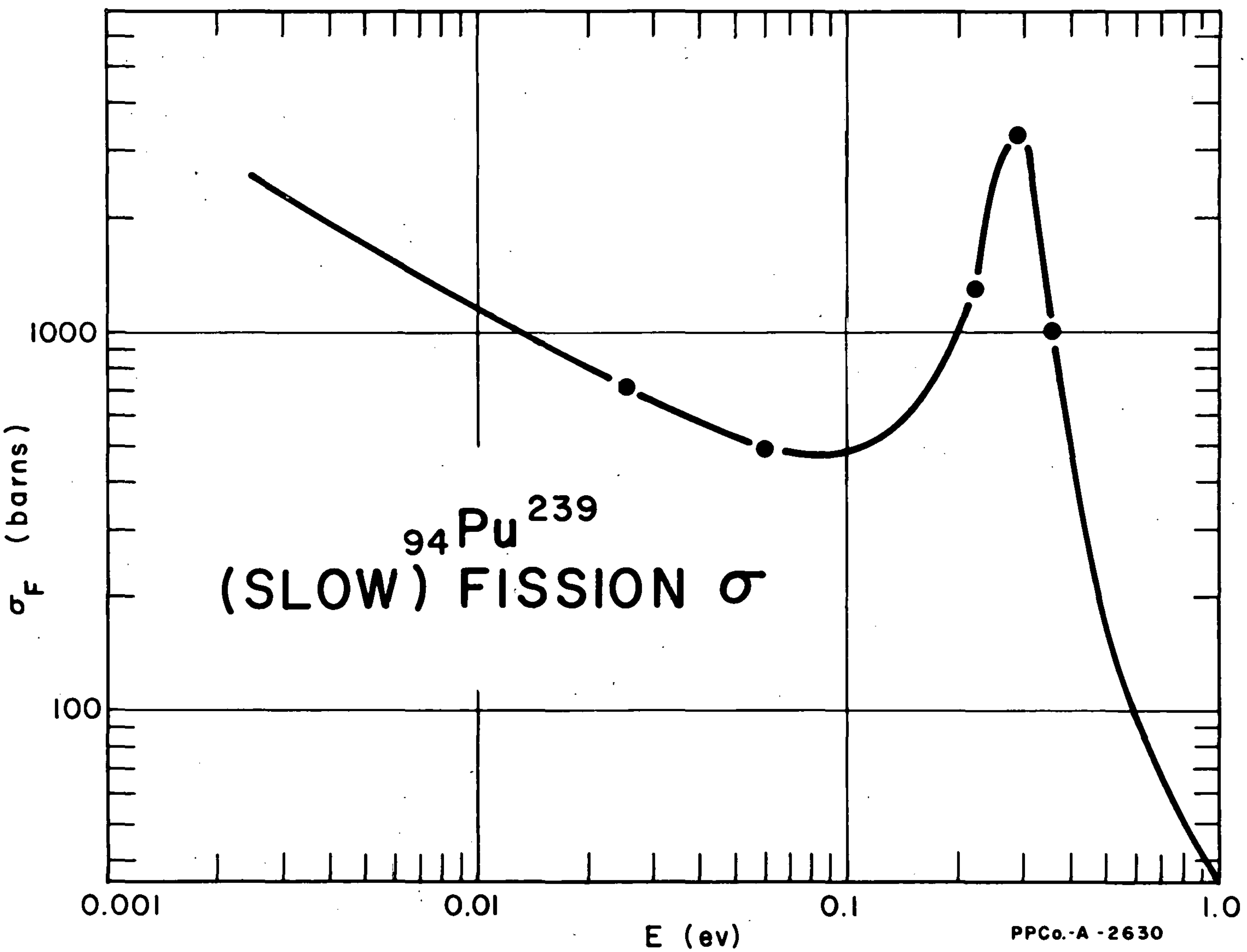


Fig. 7 Low Energy Portion of Pu^{239} Fission Cross Section

COMPARISON OF THERMAL AND RESONANCE FISSION IN Pu-239 AND Pu-241

Pu ISOTOPE	NEUTRON ENERGY	R, INDICATED BY		
		Mo^{99}/Cd^{115}	Mo^{99}/Sn^{121}	Mo^{99}/Sn^{125}
239	0.06 ev	1.33 ± 0.06	1.26 ± 0.05	1.16 ± 0.04
	0.22 ev	2.60 ± 0.15	2.82 ± 0.32	1.85 ± 0.18
	0.297 ev	3.00 ± 0.28	3.28 ± 0.31	2.05 ± 0.07
	0.36 ev	3.24 ± 0.14	3.22 ± 0.25	1.78 ± 0.16
	ALL epi-Sm	2.41 ± 0.14	2.34 ± 0.06	1.79 ± 0.06
241	ALL epi-Sm	1.016 ± 0.031	1.011 ± 0.084	0.995 ± 0.15

PPCo.-A-2631

Fig. 8 Pu^{239} "R" Value Data

COMPARISON OF CALCULATED & MEASURED R VALUES FOR Pu^{239}

Energy	Thermal	0.06 ev	0.22 ev	0.297 ev	0.36 ev
Contribution to $\sigma_{n,f}$ from 0.297-ev level	358 b (48%)	303 b (61%)	1100 b (97.4%)	3265 b (99%)	1120 b (97.4%)
Contribution to $\sigma_{n,f}$ from other spin state	388 b (52%)	192 b (39%)	~30 b (2.6%)	~30 b (~1%)	~30 b (2.6%)
Calculated R value / Observed R value	1.00 (normalized)	1.21	2.97	3.21 (assumed)	2.95
Mo^{99}/Cd^{115}	1.00 (normalized)	1.33 ± 0.06	2.60 ± 0.15	3.00 ± 0.28	3.24 ± 0.14
Mo^{99}/Sn^{121}	1.00 (normalized)	1.26 ± 0.05	2.82 ± 0.32	3.28 ± 0.31	3.22 ± 0.25

PP Co.-B-4289

Fig. 9 Comparison of Calculated and Measured "R" Values for Pu^{239} at Several Energies

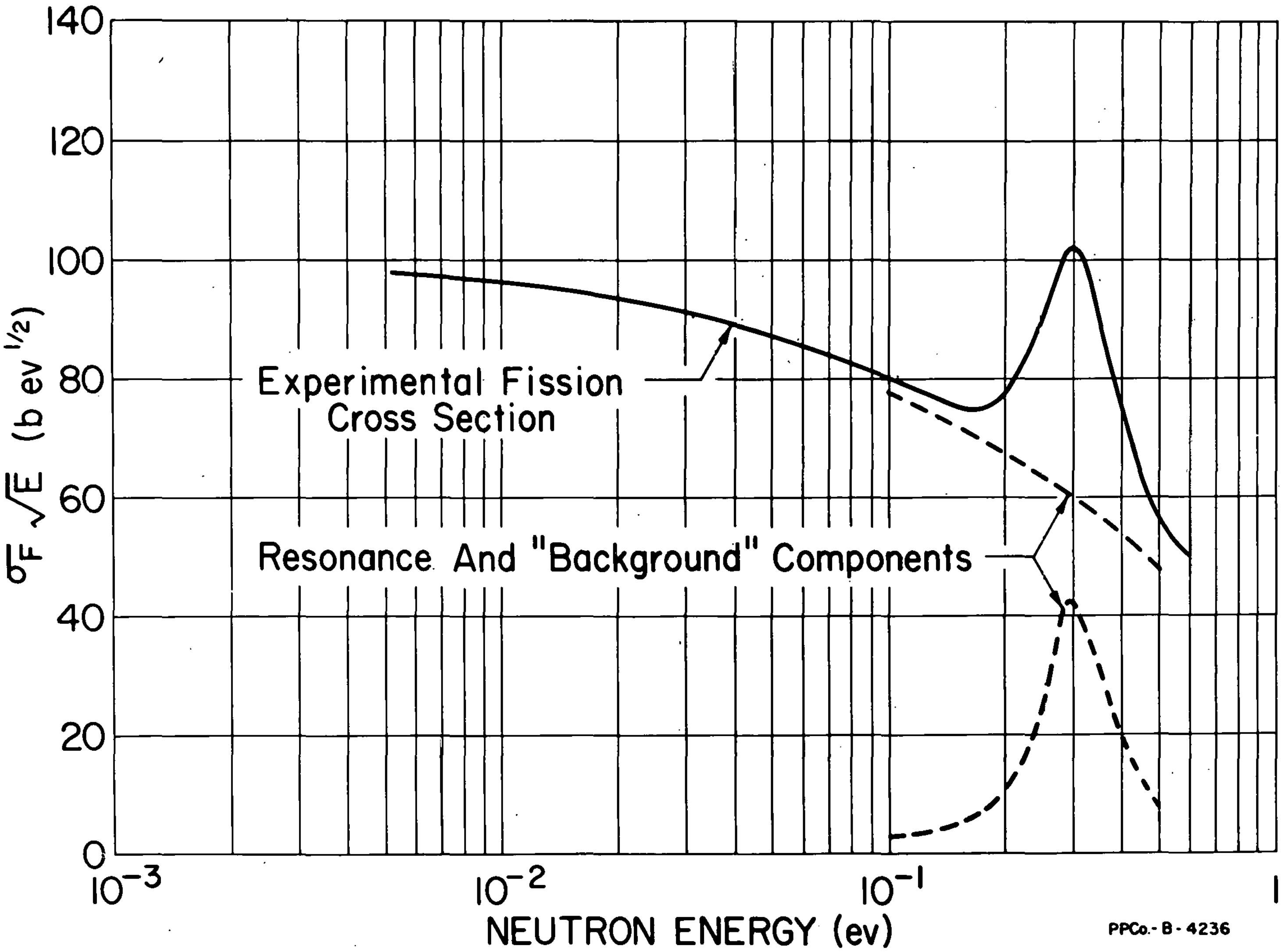


Fig. 10 Low Energy Fission Cross Section of U^{235}

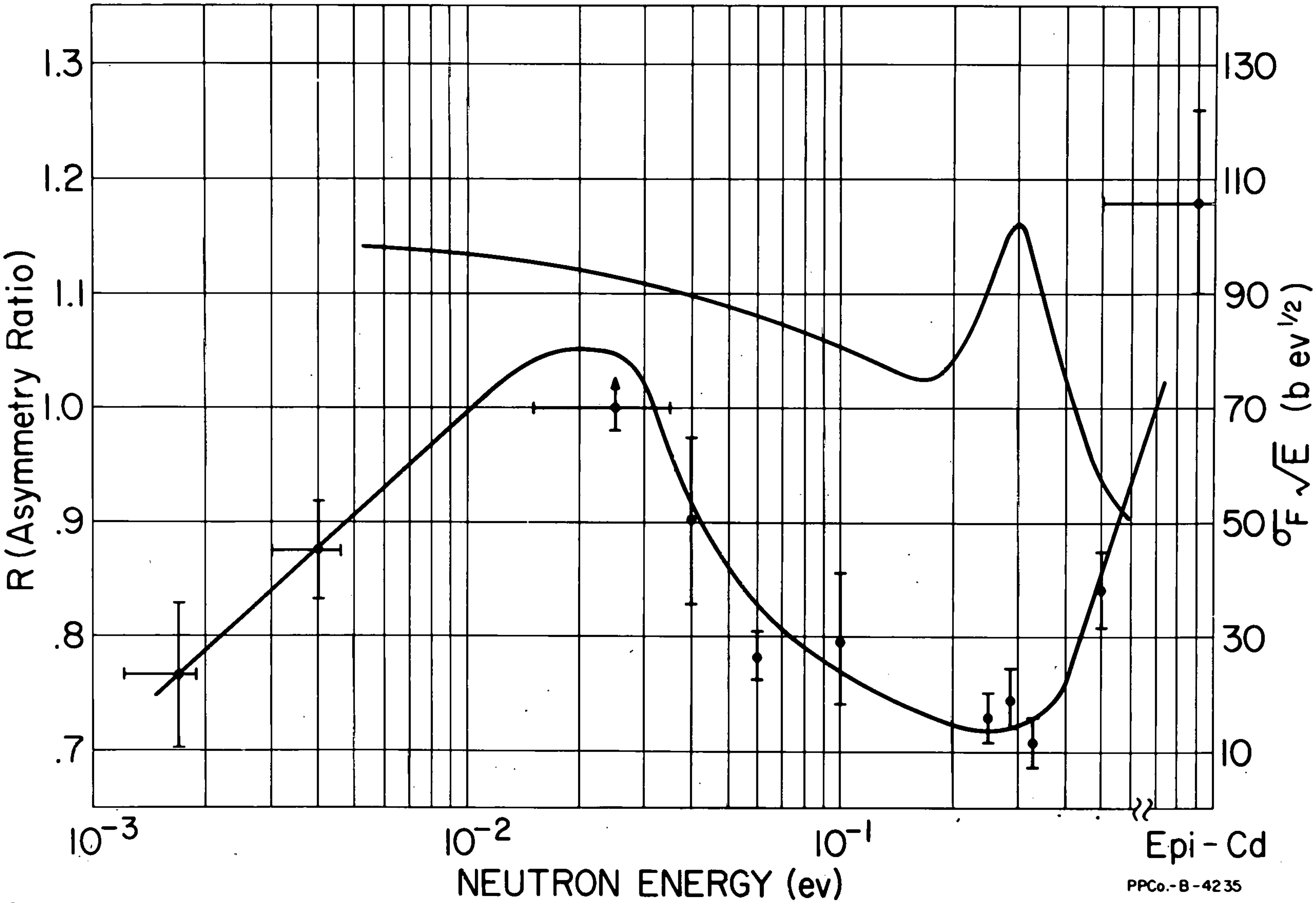


Fig. 11 Low Energy Fission Cross Section of U^{235} and Plot of "R" Values

PHILLIPS
PETROLEUM
COMPANY

PHILLIPS 66

ATOMIC ENERGY DIVISION