



**AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT
LUCAS HEIGHTS**

**THE ENERGY DEPENDENCE OF $\bar{\nu}_p$ FOR ^{233}U , ^{235}U AND
 ^{239}Pu BELOW 5.0 MeV**

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ABSTRACT

Measurements were made of the energy dependence of $\bar{\nu}_p$ for neutron induced fission of ^{233}U from thermal to 2.0 MeV. An analysis has been made of all existing data below 5 MeV for ^{233}U , ^{235}U and ^{239}Pu . It is observed from this analysis that the energy dependence of $\bar{\nu}_p$ for all three nuclei may be characterized by a two line dependence in which the change of slope occurs at the pairing energy. The nature of the $\bar{\nu}_p(E_n)$ dependence is explained in terms of the double-humped fission barrier and is consistent with the adiabatic assumption of weak coupling of the collective degrees of freedom at the saddle point to the nuclear degrees of freedom at scission.

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CONTENTS

	<u>Page</u>
1. INTRODUCTION	1
2. EXPERIMENTAL METHOD	2
3. RESULTS	2
4. ANALYSIS OF ^{233}U DATA	2
5. ANALYSIS OF ^{235}U AND ^{239}Pu DATA	3
6. DISCUSSION	4
7. ACKNOWLEDGEMENT	6
8. REFERENCES	6

Table 1 Isotopic Abundances of ^{233}U Target

Table 2 Corrections to Data

Table 3 Final $\bar{\nu}_p$ Results for ^{233}U

Table 4 Least Square Fits to ^{233}U , ^{235}U and ^{239}Pu Results

Figure 1 $\bar{\nu}_p$ Versus Incident Neutron Energy - ^{235}U - (Boldeman and Walsh 1970)

Figure 2 The Double-Humped Barrier in the Deformation Energy of a Heavy Nucleus

Figure 3 $\bar{\nu}_p$ Versus Incident Neutron Energy - ^{233}U - This Work

Figure 4 $\bar{\nu}_p$ Versus Incident Neutron Energy - ^{233}U - This Work and Other Measurements

Figure 5 $\bar{\nu}_p$ Versus Incident Neutron Energy - ^{239}Pu - Other Measurements

Figure 6 Average Total Kinetic Energy Data for ^{233}U

1. INTRODUCTION

The precise dependence of $\bar{\nu}_p$, the average number of prompt neutrons produced per fission, on the energy of the incident neutron E_n has been the subject of considerable experimental research. Apart from the obvious importance of such data in reactor physics applications, the exact dependence provides useful information on the later stages of the fission process. In particular the change in the distribution of the increasing compound excitation between kinetic energy of coulomb repulsion of the fragments and fragment excitation at scission can be obtained from data of this type.

In the case of neutron fission of ^{235}U , for which most data are available, conflicting evidence on the $\bar{\nu}_p(E_n)$ dependence has been presented in recent years. Early measurements (e.g. Hopkins and Diven 1963, Mather et al. 1964), showed that the $\bar{\nu}_p(E_n)$ dependence in the region 0-14 MeV above the neutron binding energy was linear, but that a change of slope occurred at between 1.6 and 3 MeV incident neutron energy. Subsequent measurements by Blyumkina et al. (1964) and Meadows and Whalen (1967) in the region 0-1 MeV were claimed to show fine structure in the vicinity of 400 keV neutron energy. The structure was explained assuming weak coupling of the collective saddle point energy at a single-humped fission barrier to the excitation energy at scission. Despite the volume of data available, a clear picture of the $\bar{\nu}_p(E_n)$ dependence for ^{235}U did not emerge because of the large discrepancies between different measurements.

In a previous investigation (Boldeman and Walsh 1970), precise measurements were made of $\bar{\nu}_p(E_n)$ for ^{235}U between 0 and 2 MeV neutron energy. The $\bar{\nu}_p(E_n)$ dependence was found to be linear (Figure 1) and no evidence was found in the data to support the proposals of either Blyumkina et al. (1964) or Meadows and Whalen (1967). Two recent measurements of the correlated parameter, the average total fragment kinetic energy \bar{E}_K , support the $\bar{\nu}_p$ data of Boldeman and Walsh (1970). Dyachenko et al. (1968) and Ajitanand and Boldeman (1970) both found the average total kinetic energy to be constant for neutron induced fission from 0-1 MeV. In terms of a single-humped fission barrier, the $\bar{\nu}_p$ and \bar{E}_K data were consistent only with strong coupling. However, it was pointed out by Boldeman and Walsh (1970) that the existence of the double-humped fission barrier (Strutinsky 1968) (see Figure 2) does not allow any conclusion to be drawn regarding weak or strong coupling from the linear $\bar{\nu}_p$ and \bar{E}_K data without knowledge of the relative heights of the two humps (A and B in Figure 2) of the fission barrier. It is not known at present which of these two humps is higher for the compound nucleus ^{236}U .

For the compound nucleus ^{234}U however, there is evidence to suggest that the second potential barrier (barrier B) is higher (Bjornholm and Strutinsky 1969) and, as the existing $\bar{\nu}_p$ data are poor, an examination of the energy dependence of $\bar{\nu}_p$ for neutron fission of ^{233}U is worth while.

2. EXPERIMENTAL METHOD

The $\bar{\nu}_p$ measurements were made using the large liquid scintillator method. Experimental details have been reported by Boldeman and Walsh (1970) and Boldeman and Dalton (1967). The neutron detection efficiency of the liquid scintillator was obtained by comparing the mean neutron count per spontaneous fission of ^{252}Cf with the assumed $\bar{\nu}_p$ value of 3.782 for this process. Details of the ^{233}U fission target are summarised in Table 1. Neutrons were produced via the $^7\text{Li}(p,n)^7\text{Be}$ and $^3\text{T}(p,n)^3\text{He}$ reactions using analysed proton beams from a 3 MeV Van de Graaff accelerator. Table 2 summarises the corrections made to the raw $\bar{\nu}_p$ data and the estimated accuracy of each correction (Boldeman and Dalton 1967, Boldeman and Walsh 1970).

3. RESULTS

Table 3 and Figure 3 show the final ^{233}U $\bar{\nu}_p$ data for incident neutron energies between 0 and 2 MeV relative to $\bar{\nu}_p = 3.782$ for ^{252}Cf . The thermal value shown is that of Boldeman and Dalton (1967). Figure 3 also shows the two straight lines:

$$\bar{\nu}_p(E_n) = 2.491 + 0.035 E_n \text{ for } E_n < 0.44 \text{ MeV} \quad \dots(1)$$

and
$$\bar{\nu}_p(E_n) = 2.453 + 0.122 E_n \text{ for } E_n > 0.44 \text{ MeV} \quad , \quad \dots(2)$$

which are considered the best representation of the presently available ^{233}U $\bar{\nu}_p(E_n)$ data (see Section 4).

4. ANALYSIS OF ^{233}U DATA

Previous $\bar{\nu}_p$ data for ^{233}U together with the present data have been plotted in Figure 4. The plotted data have been limited to measurements below 5 MeV. Above 5 MeV, Soleilhac et al. (1969) have observed a change in slope of the $\bar{\nu}_p(E_n)$ dependences for ^{235}U , ^{238}U and ^{239}Pu , which they attribute to the onset of $(n,n'f)$ processes. A similar effect can be expected to occur in the ^{233}U $\bar{\nu}_p(E_n)$ dependence above 5 MeV. The agreement between the different data sets is good.

A preliminary inspection of the data in Figures 3 and 4 suggests that a change of slope probably occurs between 300 and 500 keV neutron energy. This was borne out by a linear fit to the combined data in Figure 4. The linear fit was poor - the probability that the data set belonged to a linear dependence was approximately 3 per cent. Closer inspection of the systematics of this linear fit indicated that the principal difficulty lay with the relatively high value of the thermal data. If the thermal data are excluded, the fit improves considerably. Furthermore, a weighted thermal average (Hanna et al. 1969) lies approximately five standard deviations above the thermal intercept of the second fit. In an endeavour

to determine the most likely energy at which the $\bar{v}_p(E_n)$ dependence changes slope, linear fits were made above and below a particular energy which was varied between 300 and 600 keV. The goodness of the two line fit as measured by χ^2 was significantly better than for a single line fit. χ^2 for the composite two line fit had its minimum for data groupings above and below 400 keV. We submit therefore the two straight lines (1) and (2) (also shown in Figures 3 and 4) as the best representation of the data. The two lines (1) and (2) intersect at 440 keV.

5. ANALYSIS OF ^{235}U AND ^{239}Pu DATA

In an evaluation of existing \bar{v}_p data for ^{235}U for E_n between 0 and 2 MeV (Boldeman and Walsh 1970), the best representation of the data in this energy region was shown to be the straight line

$$\bar{v}_p = 2.416 + 0.107 E_n \quad \dots(3)$$

The accurate \bar{v}_p data from Soleilhac et al. (1969) between 1.36 and 5 MeV give the dependence there as

$$\bar{v}_p = 2.373 + 0.129 E_n \quad \dots(4)$$

Combination of the lines (3) and (4) gives a change of slope at 1.95 MeV.

All available \bar{v}_p data for ^{239}Pu between 0 and 5 MeV are plotted in Figure 5. A least squares linear fit has been made to this data and the dependence (5) obtained

$$\bar{v}_p = (2.870 \pm 0.003) + (0.147 \pm 0.002) E_n \quad \dots(5)$$

The quality of the linear fit (5) as measured by χ^2 was reasonable. However it is observed that the thermal and low energy data appear to be systematically higher than the fitted line. It is also apparent that (5) does not intersect the \bar{v}_p value for the spontaneous fission of ^{240}Pu at - 6.3 MeV (Holmberg and Conde 1965). The fact that a two line dependence is a good representation of both the ^{235}U and ^{239}Pu \bar{v}_p data suggested that a two line dependence be tried for the ^{239}Pu data. As for ^{235}U , linear fits were made to the data above and below a specific energy which was varied from 600 keV to 1400 keV. χ^2 for the two line fits were fractionally but not significantly better than for a single line fit. However, χ^2 is not a particularly sensitive indicator of the relative probability of a single or two line dependence. Alternatively, if the systematics of the linear fits are closely examined, some evidence appears that supports a two line

dependence. For example, the fits for the data above and below 700 keV (shown in Figure 5) are

$$\bar{v}_p = 2.889 \pm 0.007 + (0.108 \pm 0.014) E_n \quad 0 \leq E_n \leq 700 \text{ keV} \quad \dots(6)$$

$$\bar{v}_p = 2.861 \pm 0.006 + (0.152 \pm 0.003) E_n \quad 700 \text{ keV} < E_n \leq 5 \text{ MeV} \quad \dots(7)$$

These two lines are sufficiently different within the errors to provide some evidence that the two data sets may belong to different populations. The various two line fits are all fairly similar and they favour a point of intersection at about 600 keV. The two line dependence (6) and (7) is therefore offered as a reasonable alternative to a single line dependence. These two lines intersect at 640 keV. It is observed that (6) is in satisfactory agreement with the ^{240}Pu spontaneous fission \bar{v}_p value.

The $\bar{v}_p(E_n)$ dependences for ^{233}U , ^{235}U and ^{239}Pu are summarised in Table 4. Also shown is the energy difference between the point where the slope changes and the fission threshold (Northrop et al. 1959) for each nuclide.

6. DISCUSSION

It has been observed that the $\bar{v}_p(E_n)$ dependence for each of the three nuclides ^{233}U , ^{235}U and ^{239}Pu may be characterized by a two line dependence. The energy difference between the intersection of the two line dependence and the fission threshold is similar for all three nuclides and sufficiently close to the pairing energy (Britt et al. 1963, Huizenga et al. 1968) to suggest that pairing is an influential factor. The change in slope for ^{233}U is far larger than for either ^{235}U or ^{239}Pu . This suggests that below the pairing energy some additional effect is involved in the determination of the $\bar{v}_p(E_n)$ dependence for ^{233}U .

The important properties of the fissioning nucleus, determining the form of the $\bar{v}_p(E_n)$ dependence, are the relative heights of the two humps of the fission barrier and the strength of the coupling of the collective energy at the second barrier to the nuclear degrees of freedom at scission. Elyumkina et al. (1964) have shown that weak coupling of the collective energy at a single-humped fission barrier should produce non-linear $\bar{v}_p(E_n)$ behaviour for ^{235}U between 100 and 500 keV neutron energy. With a double-humped fission barrier, weak coupling will not necessarily produce fine structure unless barrier B in Figure 2 is higher than barrier A.

In our earlier measurements of $\bar{v}_p(E_n)$ for ^{235}U (Boldeman and Walsh 1970) no evidence whatsoever was found of any fine structure of the type mentioned above. Related measurements of \bar{E}_K (Ajitanand and Boldeman 1970), between thermal and 1 MeV showed this parameter to be constant. In terms of a single-humped fission

barrier, the ^{235}U data are consistent with strong coupling only, but viewed in the light of a double-humped fission barrier, the linear ^{235}U data are consistent with either of the alternatives:

- (a) Strong coupling if barrier B is higher.
- (b) Weak or strong coupling if barrier A is higher.

In case (b), the linearity of the $\bar{v}_p(E_n)$ data allows no clear inference to be drawn regarding the strength of the coupling. Two possibilities allow the coupling to be weak but do not result in fine structure:

- (1) The nucleus spends sufficient time in the potential well between the two humps to 'forget' the properties it had when passing over the first barrier.
- (2) Alternatively, even if these properties are preserved, only a small difference is required between the relative heights of the two barriers such that for a particular entry channel at the first hump there are a number of channels available at the second: averaging occurs and the fine structure effects are smeared out.

Whereas weak coupling of the collective energy at the second potential hump is expected to produce non-linear behaviour in $\bar{v}_p(E_n)$ for ^{235}U if hump B is higher, the effect for ^{233}U should be somewhat different. For ^{235}U , because the fission barrier is only slightly lower than the neutron binding energy, it is possible to obtain \bar{v}_p data for neutron fission in an energy region in which there are effectively only the transition channels of two collective bands available, i.e. the negative and positive parity bands with $K = 0$. In other words the collective energy at the saddle point can have effectively only two discrete values and the preferential selection of either of these bands can, with weak coupling, produce the fine structure or non-linear behaviour in $\bar{v}_p(E_n)$. For ^{233}U however, the fission threshold is considerably lower than the neutron binding energy and, as a consequence, measurements can be made in neutron fission only in an energy region well above threshold where a considerable number of collective channels are available. Consequently, the fine structure that might be expected from the discreteness of the collective states is averaged out by their large number. The behaviour to be expected will then be an average increase in the mean collective saddle point energy which, if weak coupling is effective, will give rise to a reasonably smooth increase in \bar{E}_K and a slope for $\bar{v}_p(E_n)$ significantly lower than that expected if all additional compound excitation were to appear as fragment excitation. Above the pairing energy the availability of a large number of single particle transition states substantially reduces the effect of weak coupling. All additional compound excitation above the pairing energy is bound, even at the saddle point, to the intrinsic degrees of freedom. Thus, above the pairing energy, \bar{E}_K

should remain constant and the slope of $\bar{v}_p(E_n)$ should increase correspondingly. This type of behaviour has been observed (Figure 4). In Figure 6, existing ^{233}U \bar{E}_K data have been plotted together with the \bar{E}_K dependence estimated from the $\bar{v}_p(E_n)$ data (i.e. the linear relationships (1) and (2)). The dependence is in good agreement with the experimental data, particularly that of Dyachenko et al. (1968). Their data have the smallest errors.

It may be concluded that the ^{233}U data show the influence of weak coupling, whereas the ^{235}U data do not show any significant effect. This situation can occur if, between the compound nuclei $A = 234$ and $A = 236$, the higher of the two humps of the fission barrier changes from the second to the first i.e. the second barrier is higher for ^{234}U whereas the first barrier is higher for ^{236}U . Weak coupling then would have a sizeable effect on the $\bar{v}_p(E_n)$ dependence for ^{233}U without seriously affecting the dependence for either ^{235}U or ^{239}Pu .

An explanation may now be presented for the slight changes in slope of the $\bar{v}_p(E_n)$ dependences of ^{235}U and ^{239}Pu at the pairing energy. Boldeman (1970) has shown from the data of Thomas and Grover (1967) that gamma ray competition below the pairing energy accounts for approximately 8 per cent of additional fragment excitation and that this proportion is probably very much smaller above the pairing energy. Gamma ray competition of this kind would therefore produce a change in slope of $\bar{v}_p(E_n)$ at the pairing energy. The magnitude of the change produced in this manner is approximately 50 per cent of that actually observed. It is possible that the remainder of the observed change is caused by weak coupling effects which have not been entirely smeared out.

The type of behaviour observed here in the $\bar{v}_p(E_n)$ dependences has been reported in other properties of the fission process. Bjornholm and Strutinsky (1969) attributed the change in the magnitude of the anisotropy of the fission fragment angular distributions to a change in the relative heights of the two humps of the fission barrier between $A = 234$ and $A = 239$.

7. ACKNOWLEDGEMENT

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TABLE 1

ISOTOPIC ABUNDANCES OF ²³³U TARGET

Isotope	Per Cent Abundance
²³³ U	99.27
²³⁴ U	0.07
²³⁵ U	0.04
²³⁶ U	0.07
²³⁸ U	0.53

TABLE 2
CORRECTIONS TO DATA

	Percentage Correction Applied	Error in Percentage Correction
1. Statistical Accuracy *	-	0.50
2. Fission Spectra Differences	- 0.53 to - 0.45	0.24
3. Dead Time Correction (Relative to Cf ²⁵² correction) * [†]	- 0.20	0.04
4. Delayed Gamma Rays	0.00	0.10
5. Thermal Contamination *	+ 0.12	0.03
6. Second Neutron Group from ⁷ Li(p,n) ⁷ Be †	+ 0.06	0.01
7. Preferential Detection of Fragments	+ 0.05	0.05
8. Fragment Anisotropy	0.00	0.00
9. False Gates *	+ 0.05	0.02
10. Impurities in ²³³ U sample	0.00	0.00
11. Electronics Errors	0.00	0.03

* Average over all runs

† Average for 700 and 917 keV results

TABLE 3

FINAL $\bar{\nu}_p$ RESULTS FOR ^{235}U Relative to $\bar{\nu}_p (^{252}\text{Cf}) = 3.782$

Neutron Energy (keV)	$\bar{\nu}_p$
thermal	2.491 ± 0.008
300 ± 25	2.502 ± 0.014
485 ± 31	2.508 ± 0.010
600 ± 32	2.546 ± 0.012
700 ± 25	2.546 ± 0.011
917 ± 33	2.564 ± 0.012
1500 ± 50	2.645 ± 0.019
1870 ± 50	2.684 ± 0.022

TABLE 4

LEAST SQUARES FITS TO ^{233}U , ^{235}U AND ^{239}Pu RESULTS

$$\bar{\nu}_p(E_n) = A + BE_n$$

	A	B(MeV ⁻¹)	E _f (MeV)	E _{DIFF} [*] (MeV)
^{233}U	E _n < 0.44 MeV	2.491 ± 0.008	0.035 ± 0.026	
	E _n > 0.44 MeV	2.458 ± 0.013	0.123 ± 0.014	- 1.47 1.91
^{235}U	E _n < 1.95 MeV	2.416 ± 0.004	0.107 ± 0.004	
	E _n > 1.95 MeV	2.373 ± 0.014	0.129 ± 0.004	- 0.60 2.55
^{239}Pu	E _n < 0.64 MeV	2.889 ± 0.007	0.108 ± 0.014	
	E _n > 0.64 MeV	2.861 ± 0.006	0.152 ± 0.003	- 1.61 2.25

E_f is the fission threshold (Northrop et al. 1959)

* E_{DIFF} is the energy difference between the point where the slope changes and the fission threshold.

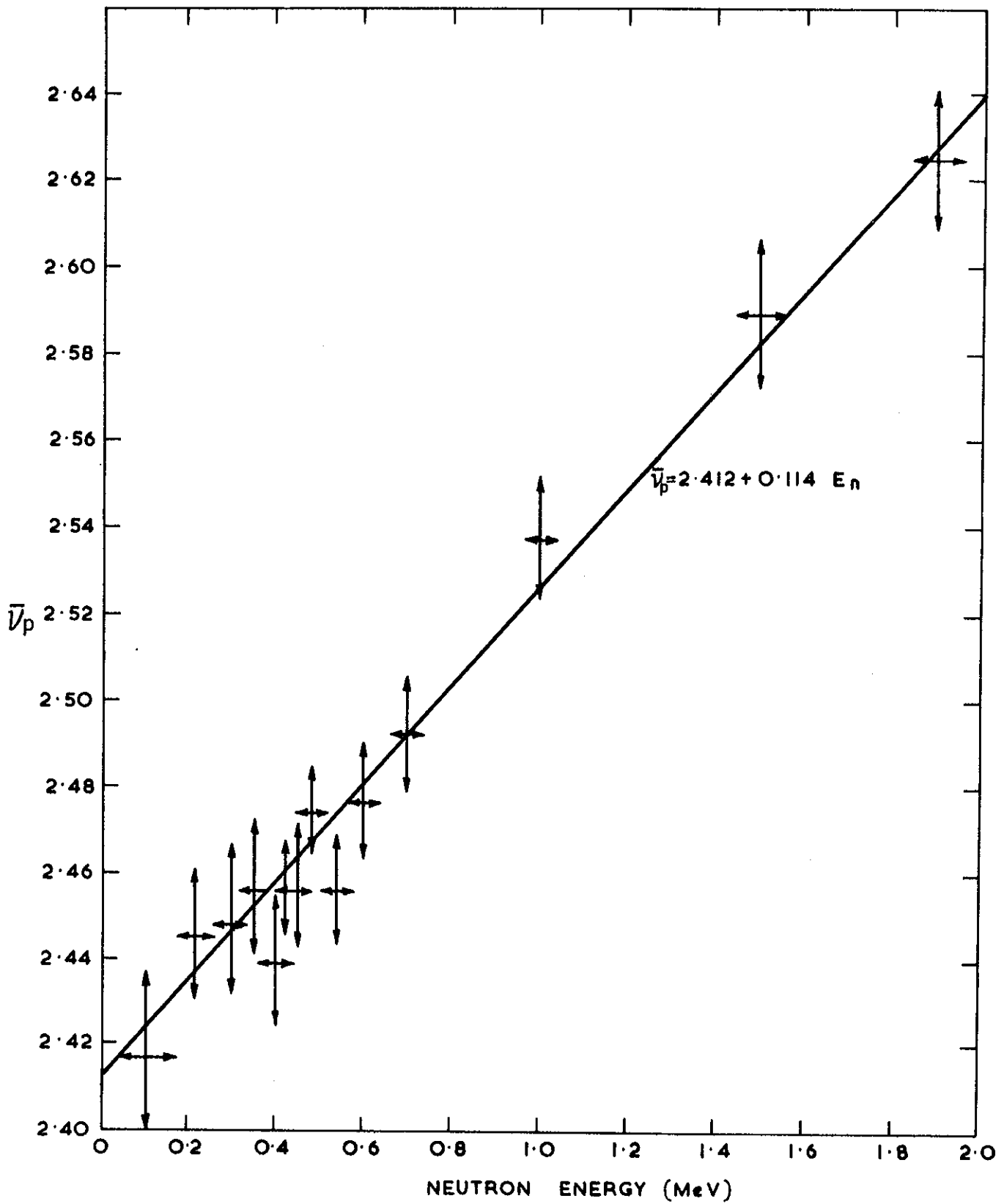


FIGURE 1. $\bar{\nu}_p$ VERSUS INCIDENT NEUTRON ENERGY ^{235}U
 (Boldeman and Walsh 1970)

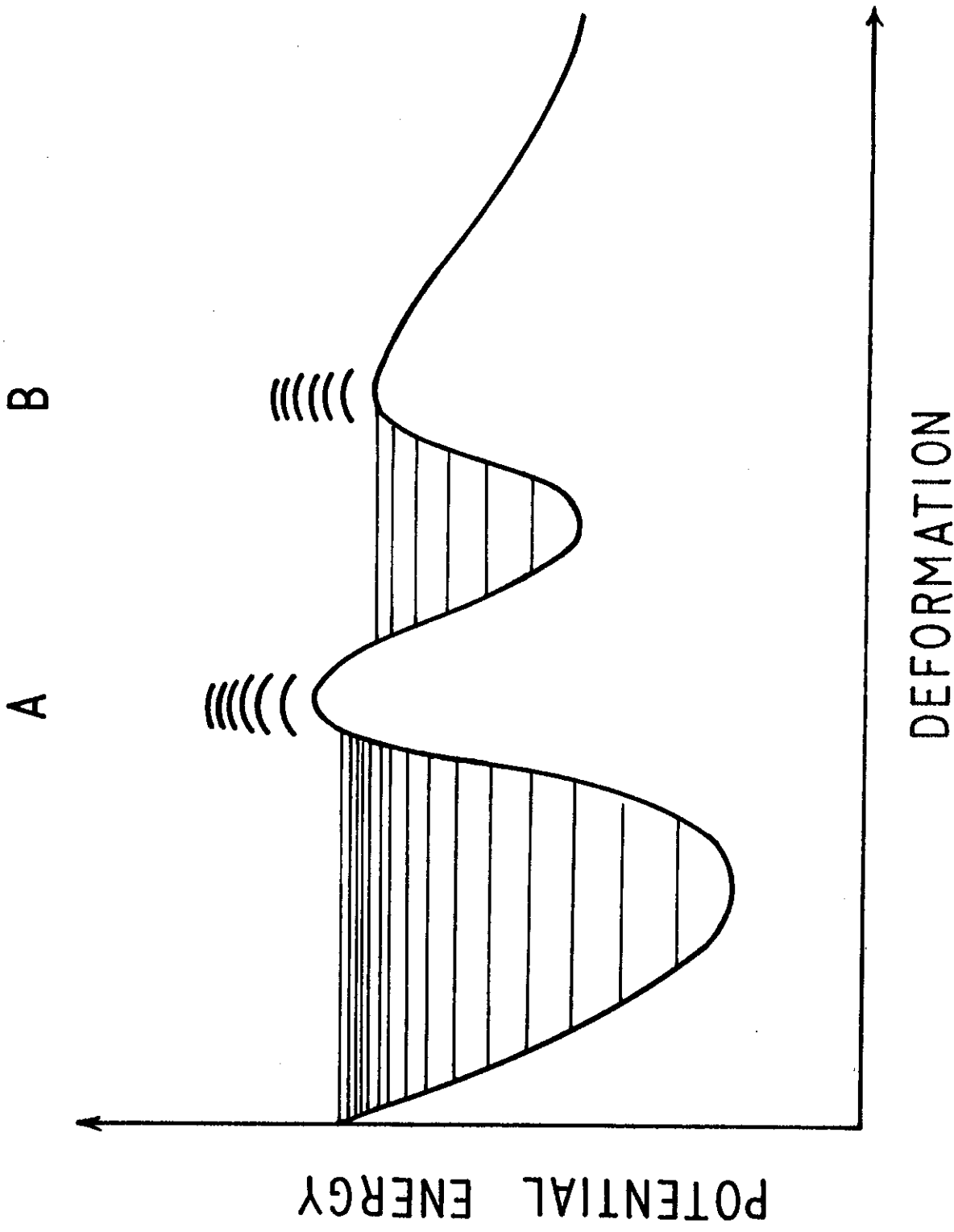


FIGURE 2. THE DOUBLE-HUMPED BARRIER IN THE DEFORMATION ENERGY OF A HEAVY NUCLEUS

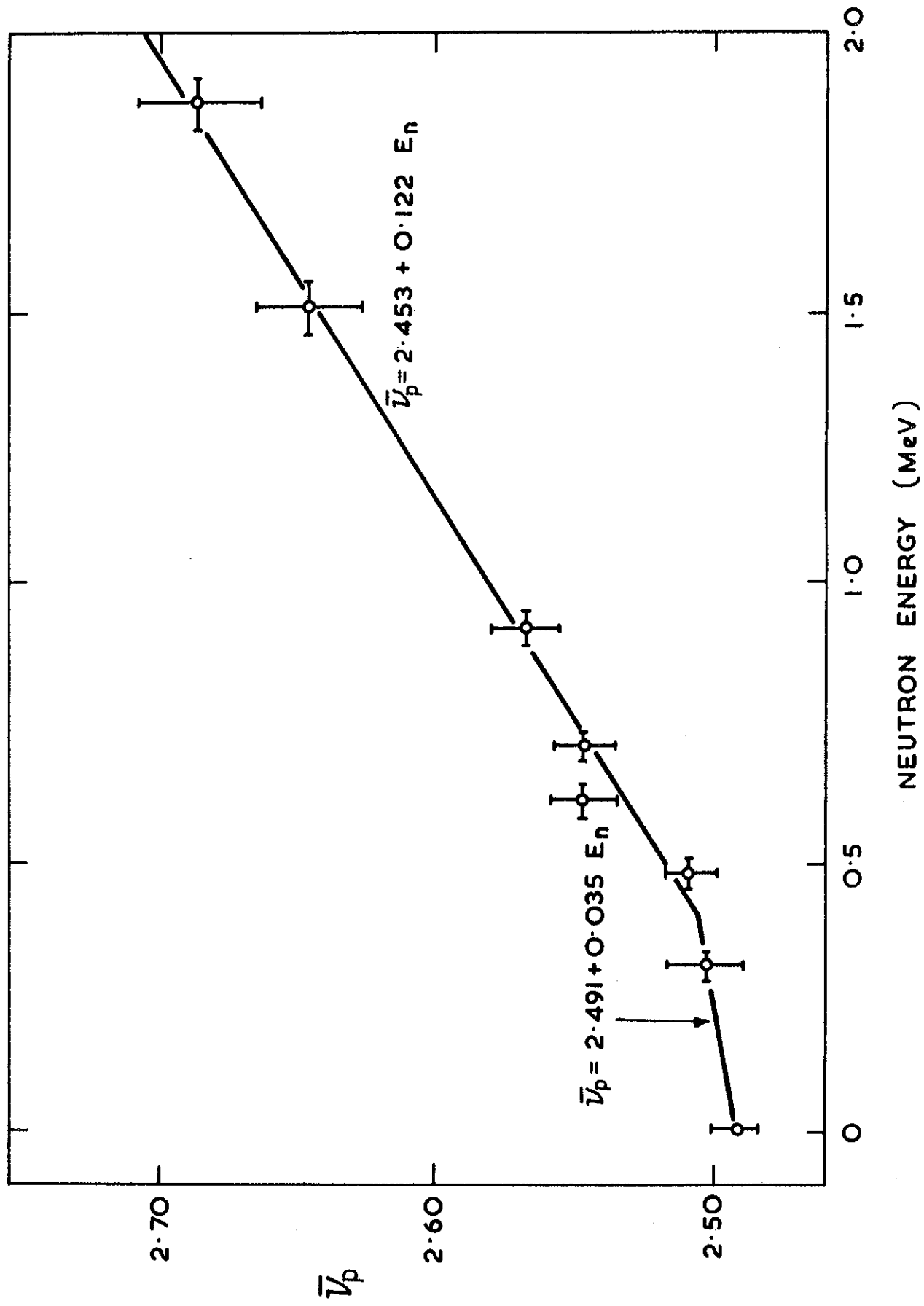


FIGURE 3. $\bar{\nu}_p$ VERSUS INCIDENT NEUTRON ENERGY ^{233}U - THIS WORK

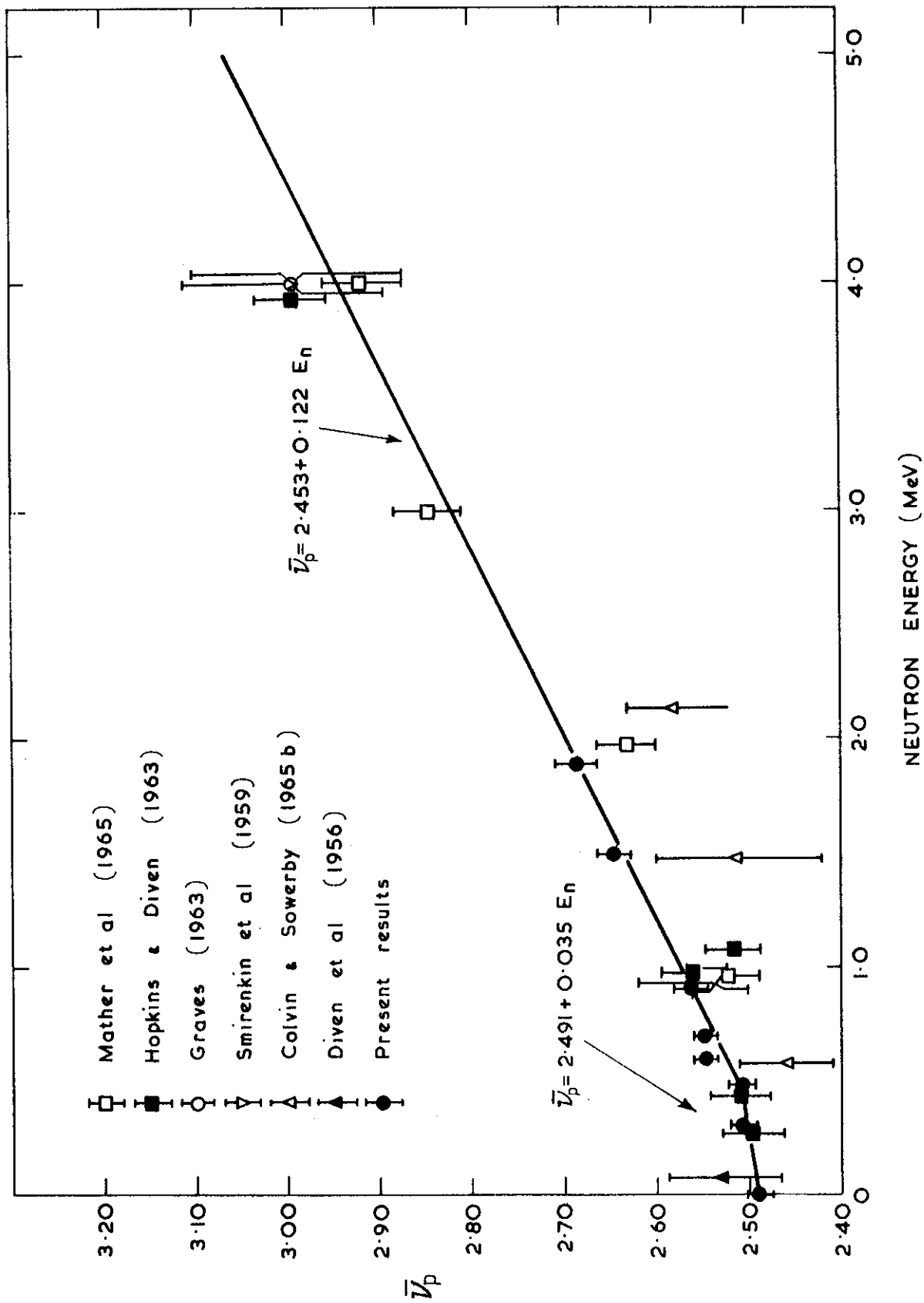


FIGURE 4. \bar{v}_p VERSUS INCIDENT NEUTRON ENERGY ^{233}U - THIS WORK AND OTHER MEASUREMENTS

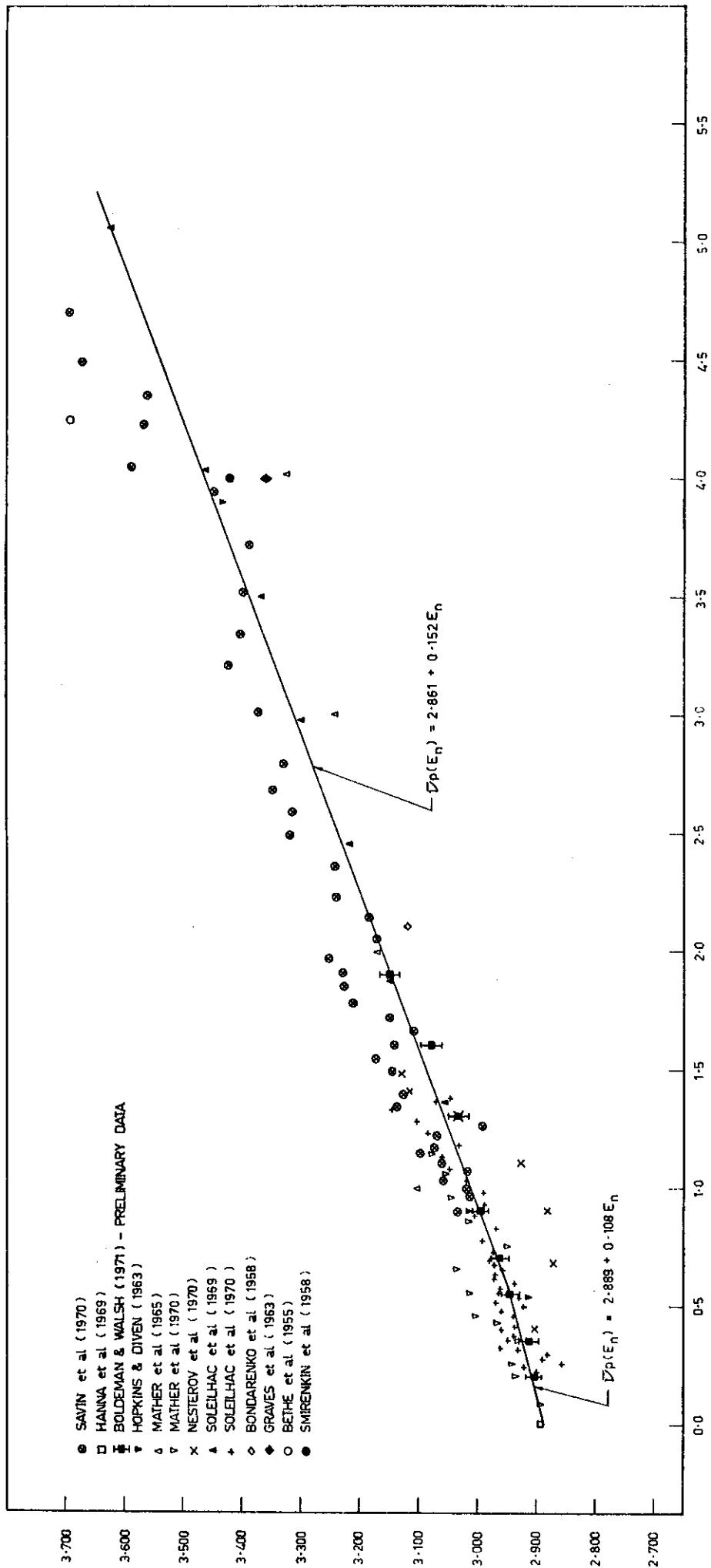


FIGURE 5. $\bar{\nu}_p$ VERSUS INCIDENT NEUTRON ENERGY, ^{239}Pu

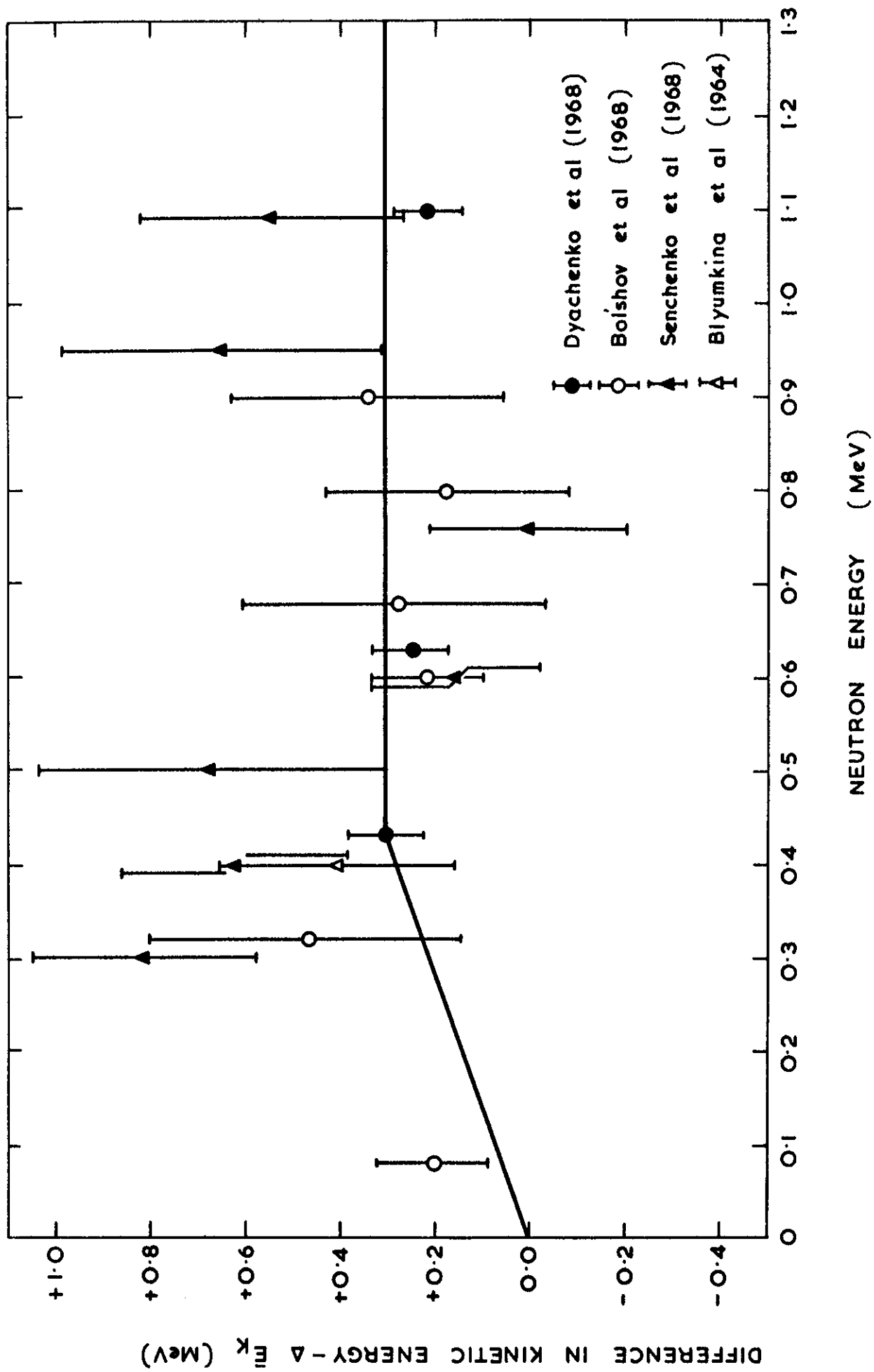


FIGURE 6. AVERAGE TOTAL KINETIC ENERGY DATA FOR ^{233}U