UCRL-10193

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UCRL-10193

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Berkeley, California
Contract No. 7405-eng-48

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ABSTRACT

Average recoil ranges have been measured for five fission products formed by 23 Mev H^2 reactions with U^{238}. The ranges of products from near symmetric fission (Cd^{115} and Ag^{111}) are longer than for neutron fission of U^{235}, while the ranges of asymmetric products (I^{131}, Ba^{140}, and Sr^{89}) are shorter. The kinetic energy deficit for near symmetric fission is 15 to 20 Mev lower for this system than for thermal neutron fission of U^{235}. The magnitude of the kinetic energy deficit for U^{236} fission is reexamined by comparing range data with recent time of flight measurements and neutron emission probabilities. This comparison leads to a kinetic energy deficit of approx 23 Mev for U^{236} fission.

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I. INTRODUCTION

Many different measurements have been made of velocities, energies and ranges of fission products from various kinds of fission.[1,2] One interesting feature of many of these measurements is that the kinetic energies of near symmetric fission products seem to be significantly smaller than the asymmetric products. We define the term "kinetic energy deficit" as the difference between maximum average kinetic energy release and that for symmetric fission. This kinetic energy deficit has been reported for several fissile nuclei at excitation energies near the threshold.[1-3] A few experiments have been reported at very large excitation energies,[4,5] but there is very little information about this effect at excitation energies a few tens of Mev greater than threshold.[3,4] The quantitative evaluation of this kinetic energy deficit has not, as yet, been established for low energy fission. Coincidence counting techniques have been very successful for measurements of the energies of asymmetric products.[1,2] But various difficulties have prevented these techniques from obtaining unambiguous results for the symmetric fission products of much lower yield. Radiochemical recoil range measurements have perfect resolution, but conversion from range to energy requires assumption.

Several workers have reported range measurements for thermal neutron induced fission of Pu^{239} and U^{235} and spontaneous fission of Cf^{252}.[6-10] The analysis of these data is based on a comparison with velocity measurements of the fragments of high yield.[8,9,10] From the velocities and an assumption of

the number of neutrons emitted per fragment, one can obtain final kinetic energies after neutron emission. Then one obtains range energy relationships for products of asymmetric fission. These range-energy relationships are extrapolated to products of near symmetric fission, and kinetic energies can then be calculated from ranges. An extensive study of this type has been made by Niday who reported range measurements in thick U metal foils for U^{236} fission.[9] Niday used Stein's velocity measurements.[11]

Recently, Milton and Fraser have remeasured the fragment velocities.[12] Also, they have proposed that neutron emission probability is determined by fragment mass, independent of fissile nucleus.[13] We have reexamined the range data in the light of these new developments. Previous analyses of range data gave a kinetic energy deficit of approximately 33 Mev for U^{236} fission;[8,9] Milton and Fraser's velocity measurements give 40 Mev,[12,13] and our analysis of Niday's ranges gives approx 23 Mev.

We have measured ranges of Sr^{89}, Ag^{111}, Cd^{115}, I^{131} and Ba^{140} from 23 Mev H^2 irradiation of U^{238}. These range data indicate a decrease of 15 to 20 Mev in the kinetic energy deficit for this system compared to U^{236} fission.

II. EXPERIMENTAL PROCEDURES AND RESULTS

We have used the thick-target technique for measuring average range values.[5,9] Targets of natural uranium metal (0.001 in. thickness) were sandwiched between Au or Al catcher foils. The target foils were cleaned with 6N HNO_3 for a few minutes until the surface was bright and shiny. Irradiations were performed at the University of California 60 inch cyclotron less than 36 hours after the uranium was cleaned.

After irradiation the catcher and target foils were dissolved separately and various products were chemically separated. Activation of the catcher foils was checked by blank determinations for each experiment. The activation was considerably less than 1% of the recoil activity in each case, and no correction was necessary. Counting of β and γ activity was performed as described previously.[8]

Previous studies have shown that 3 to 5% more recoil atoms are deposited in Al catcher foils than in Pb catchers.[9] This effect has been attributed to differences in scattering of the recoils by heavy and light stopping atoms.[8,9] We have used both Au and Al catching foils in this study in order to check the magnitude of this scattering effect.

The experimental results are given in Table I. The first column shows the nuclide studied; the second column the ratio of recoils forward to backward. In the third column we give the product $2W(F_F + F_B)$ where W denotes the target thickness and F_F and F_B denote the fractions of the total activity observed in the forward and backward catcher foils respectively. Finally, we give the number of independent determinations.

III. ANALYSIS OF EXPERIMENTAL RESULTS

It is convenient to think of nuclear fission as a two step process. In the first step the projectile strikes the target atom and imparts an impact velocity, denoted by $\underset{\sim}{v}$. Then the fission event takes place giving rise to an additional velocity, which we will denote by $\underset{\sim}{V}$, in the reference system of the fissile nucleus. In the laboratory the resultant velocity is the vector sum of $\underset{\sim}{v}$ and $\underset{\sim}{V}$. The objective of these experiments is to obtain measurements of the average magnitude of $\underset{\sim}{V}$ for fission products of different mass. From these average velocities we calculate the average kinetic energy release in the fission process as a function of the mass ratio of the products.

Quite a body of experimental data are available for reactions of U^{238} with H^2. These data give strong evidence that most fission events proceed through compound nucleus formation.[14-16] Therefore, the direction of $\underset{\sim}{v}$ must be along the beam direction and ratio v/V, denoted by η, can be calculated to be about 0.02 to 0.04. Angular distribution measurements for several products indicate that a function of the form $a \pm b \cos^2\theta$ gives a very good representation of the angular distribution of the fragments in the center of mass system (or fissile nucleus system).[17]

Using this information, we can express the range of the products in the target material by the following equation:

$$R = 2W(F_F + F_B) \left[1 + (b/3a)\right] \left[1 + (b/2a)\right]^{-1} \qquad (1)$$

The range R is a hypothetical range that the product would have if the lab system velocity were V and if the product moved along straight paths. Terms of order η^2 have been neglected in Eq. (1).

Cloud chamber studies have shown that the fission products suffer a significant amount of scattering away from a straight path.[18] However, if the scattering in the target and catcher foils is equivalent, then Eq. (1) will still give (to a very good approximation) the average distance from origin to final resting place. We assume that this condition is satisfied for U metal targets and catchers of Au or Pb. It has been shown that scattering effects are different for stopping in Al compared to Au or U.[8,9] Niday found that apparent ranges using Pb catchers were 3 to 5% shorter than those obtained by Al catchers. As shown in Table I we find this difference to be 2. ± 1.% for Sr^{89} and Ba^{140}. Considering the results of Niday and this work we have chosen the multiplicative factor 0.970 to correct all apparent ranges measured using Al catchers. In our work this correction was applied only for the I^{131}

measurements. In order to compare our ranges to those for U^{236} fission this correction was applied to all the measurements of Niday.[9]

Table II shows the range values resulting from this work and compares them to the measurements for U^{236} fission. In the first column we give the nuclide observed. Then we give the anisotropy parameters from reference 17. The third column shows ranges in U calculated from Eq. 1. Finally, we list the ratio of these ranges to the corresponding range for U^{236} fission. Considering the various sources of error, we estimate standard deviations of about 1.5% for these ratios. This leads to an error of about 2.3% for the relative kinetic energies as discussed in the next section.

The forward-backward ratio F_F/F_B gives a measure of η (or v/V) for each product if the variation of cross section with beam energy is known accurately. Values of η were calculated from the data in Table I assuming that the cross sections were constant throughout the target foil. These η values were only about 1/2 as large as calculated for total momentum transfer. We attribute this discrepancy to a cross section decrease of about 8% as the beam energy is degraded approx 3/4 Mev by the 0.001 U foil. Accurate excitation functions are not available and therefore it is not possible to make a correction for this effect.

IV. KINETIC ENERGIES

The usefulness of range measurements for measuring kinetic energies depends on our knowledge of range energy relationships. The theory of stopping for fission products is complicated by electron capture and loss processes and by energy transfers to nuclei (or atoms) as well as to electrons of the stopping medium. N. Bohr has estimated that ranges R of fission products vary almost linearly with velocity V and that values of dV/dR are approximately proportional

to $Z^{1/2}/A$ of the product.[19] Niday has fitted his range data for U^{236} fission to such a relationship.[9] Niday used initial energies from Stein's measurements of 1957 and the assumption that 1.25 neutrons were emitted from each fragment.[11] More recent time of flight measurements and neutron emission probabilities make it advisable to reinvestigate the initial kinetic energies of products from U^{236} fission.[12,13,20,21]

The basic assumption that we make is that the range-energy relationship is a smooth function of Z and A of the products. In other words, we assume that there are no violent changes in stopping powers due to closing of electron shells or other considerations. This assumption is tested to some extent by the range measurements for various fissile nuclei U^{236}, Pu^{240}, Cf^{252}. Plots of range versus mass number have significantly different shapes for these three systems.[7-10] The ranges of Cd and Ag products are shorter than those of neighboring products for U^{236} and Pu^{240} fission but this effect is not observed for Cf^{252} fission.[7-10] These differences imply that the range data reflect mainly the differences in the fission processes rather than changing stopping powers.

We use two functional forms for the relationship of range to velocity V or energy E

$$R = k_1 (V-V_c) \qquad (2)$$

$$R = k_2 E^{2/3} \qquad (3)$$

where k_1 and k_2 are functions of Z and A of the products and of the stopping material. Eq. 2 follows Niday's formulation of the Bohr theoretical treatment with V_c taken as a constant.[9] Niday's approach gives values of 0.13 ± 0.02 $(Mev/amu)^{1/2}$ for V_c. Eq. 3, range proportional to energy to the 2/3 power, is from an empirical fit to velocity loss measurements.[8] The use of two different

functions gives a feeling for the sensitivity of the final results to the form of the range-energy relationship.

Milton and Fraser give values of ν[20,21] as a function of fragment mass along with new time of flight measurements.[12] These ν values are shown in Fig. 1. The existing data[20,21] are sufficient to determine ν values rather well for masses of approx 80-106 and 130-150. In the mass region of approx 106-130 there is essentially no information. Milton and Fraser suggest that the ν values for U^{236} follow the same trend as those of Cf^{252}.[13] This is indicated as curve A in Fig. 1. More recently it has been suggested that the maximum value of ν may occur for masses complementary to the shell closure region of $Z \approx 50$ and $N \approx 82$.[22] This would lead to a maximum ν value for A $\approx$ 106. If this is the case ν values would fall near curve B in Fig. 1.

We have used time of flight data from reference 12 and ν values from Fig. 1 to calculate energies and velocities of the U^{236} fission products after neutron emission. Using these initial energies and the ranges of Niday we have calculated values of k_1 and k_2. These values are shown in Figs. 2 and 3. We have designated the points in various ways to indicate their reliability. Solid circles are for high yield products with well known ν values. Open circles are for products with well known ν values but having lower yields. The data for products of mass 107-129 are shown with final energies calculated from both curve A(x) and curve B(+) of Fig. 1. These points come from a region of very low yield and the velocity measurements cannot be considered reliable. (Range data for Sr^{90} and Mo^{99} have been omitted because of their larger errors. Niday's range values have been multiplied by 0.970 to correct for scattering as previously discussed.)

From Figs. 2 and 3 we see that the points for masses 109 to 125 do not fall in line with the trend of the other points. This effect may indicate a

breakdown of the initial assumption of a smooth Z and A dependence of k_1 and k_2. However we consider it more likely that the initial kinetic energies are incorrectly calculated in this region of near-symmetric fission. It is unlikely that the major difficulty is in uncertainties in ν, as indicated by the two alternatives in Figs. 1-3. Incorrect energies probably result from the difficulty of time of flight measurements in the regions of low or rapidly varying yields.

We believe that the most likely source of this discrepancy is indeed the time of flight data. Therefore we have recalculated the energies of products of near symmetric fission from the range data. We assume that the ν values for U^{236} fission (curve A, Fig. 1) are correct and that the smooth curves in Figs. 2 and 3 are correct. Of course, the kinetic energies after neutron emission depend only on the latter assumption. Primary kinetic energies require both assumptions. The final energies for the products studied in this work are listed in Table III. The two different functional forms lead to slightly different energies for the near symmetric products. Kinetic energies for the products from deuteron fission of U^{238} are also listed in Table III. These values were obtained from the range data given in Table II. The values of k_1' and k_2' for deuteron fission of U^{238} were taken to be 1/2% smaller than the smooth curves for U^{236} fission (see Figs. 1 and 2). This small correction is for the slightly greater average Z expected for fission of U^{238} by 23 Mev H^2.[15,23]

The primary total kinetic energy release E_T in the fission process depends on the number ν of neutrons emitted from each fragment,

$$E_T = E\left(\frac{A+\nu}{A}\right)\left(\frac{A_c}{A_c - A - \nu}\right) \qquad (4)$$

where A_c denotes the mass of the fissile nucleus, A the mass of the observed product and E the average energy of the observed product after neutron emission. It can be shown that the value of $dE_T/d\nu$ is 2.5 to 2.8 Mev per neutron. In Fig. 3 we show results for E_T as a function of mass ratio. The solid curve for U^{236} fission is from the time of flight data of Milton and Fraser.[12] The open triangles and the dashed curve are from (a) Niday's range values[9] (b) ν values from curve A of Fig. 1 (c) the smooth curve for $R = k_1 E^{2/3}$ from Fig. 1. The solid circles are for U^{238} reactions with 23 Mev H^2 assuming all ν values to be 3.

The kinetic energy deficits from Fig. 3 are approx 19 and 6 Mev for U^{236} and Np^{240} fission respectively. The kinetic energy deficits are somewhat dependent on the choice of range-energy parameters and the ν values. In Table IV we show results for several different choices of these quantities. Figure 3 and Table IV lead us to several conclusions. (a) Range data and time of flight data for U^{236} fission lead to quite different values of the kinetic energy release in near symmetric fission. The most important assumption that leads to this difference is that of smooth dependence of stopping power on Z and A. Uncertainty in ν values gives rise to significant uncertainties in E_T values from range measurements but probably cannot account for all the difference between range studies and time of flight studies. (b) The kinetic energy deficit for near symmetric fission is significantly smaller for fission of U^{238} with 23 Mev H^2 than for U^{236} fission. This kinetic energy deficit for H^2 fission depends slightly on the ν values taken, but mainly on the final energies of products in U^{236} fission. (c) From range data we calculate kinetic energy deficits of 19 to 27 Mev and 0 to 7 Mev for U^{236} fission and for deuteron fission of U^{238} respectively. If the 40 Mev kinetic energy deficit is correct for U^{236} fission as reported by Milton and Fraser,[12] then the range data imply a deficit of approx 20 Mev for deuteron fission of U^{238}.

ACKNOWLEDGMENTS

We appreciate helpful discussions with Drs. Earl K. Hyde, Morton Kaplan, J.C.D. Milton, Nathan Sugarman, and W. J. Swiatecki. We thank A. Tripps for assistance with several of the chemical separations.

FOOTNOTES AND REFERENCES

† Work done under the auspices of the U. S. Atomic Energy Commission.

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Table I. Summary of experimental results for U^{238} + 23 Mev H^2

Nuclide	Forward backward ratio (F_F/F_B)	2W (F_F+F_B) (mg/cm^2)	Number of measurements
	Experiments with Au catchers		
Sr^{89}	1.072 ± 0.008[a]	11.51 ± 0.15	5
Ag^{111}	1.066 ± 0.003	10.00 ± 0.04	3
Cd^{115}	1.076 ± 0.009	9.77 ± 0.11	3
Ba^{140}	1.138 ± 0.004	8.83 ± 0.06	6
	Experiments with Al catchers		
Sr^{89}	1.080 ± 0.016	11.76 ± 0.02	2
I^{131}	1.123 ± 0.014	9.55 ± 0.09	4
Ba^{140}	1.139 ± 0.010	9.00 ± 0.07	5

[a] The indicated errors are standard errors or standard deviations of the mean.

Table II. Average ranges in U for U^{238} + 23 Mev H^2

Nuclide	Anisotropy[a] (b/a)	Average range in U (mg/cm^2)	$\frac{R_{(U^{238}+H^2)}}{R_{(U^{235}+n)}}$
Sr^{89}	0.24	11.10	0.991[b]
Ag^{111}	0.17	9.74	1.031[b]
Cd^{115}	0.17	9.52	1.031[b]
I^{131}	0.21	8.97[b]	0.954[b]
Ba^{140}	0.28	8.47	0.996[b]

[a] These values are from reference 17. The standard errors are $\lesssim 0.04$.

[b] A factor 0.970 was used to correct the range values for scattering into the Al catchers.

Table III. Kinetic energies after neutron emission.

Nuclide	U^{235} + thermal n[a]		U^{238} + 23 Mev H^2 [b]	
	$R=k_1 E^{2/3}$	$R=k_2 (V-V_c)$	$R=k_1' E^{2/3}$	$R=k_2' (V-V_c)$
Sr^{89}	98.5	98.4	97.9	97.7
Ag^{111}	80.2	78.2	84.5	83.3
Cd^{115}	77.2	75.4	81.5	80.4
I^{131}	78.2	78.5	73.5	73.0
Ba^{140}	66.0	65.9	66.1	66.0

[a] The values of k_1 and k_2 were taken from the smooth curves in Figs. 2 and 3 respectively.

[b] The values of k_1' and k_2' were taken to be 1/2% smaller than k_1 and k_2.

Table IV. The kinetic energy deficit for U^{236} fission and Np^{240} fission calculated from range data.

	U^{235} + n		U^{238} + 23 Mev H^2	
Range-energy relationship[a]	ν values[b]	Kinetic energy deficit	ν values[b]	Kinetic energy deficit
$R=k_1 E^{2/3}$	A	19	3.0	6
$R=k_2 (V-V_c)$	A	23	3.0	7
$R=0.52 E^{2/3}$	A	26		
$R=k_1 E^{2/3}$	B	24	A+2	0
$R=k_2 (V-V_c)$	B	27	A+2	0

[a] The values of k_1 and k_2 for U^{236} fission were taken from the smooth curves in Figs. 2 and 3. Values of k_1' and k_2' for H^2 fission were taken to be 0.995 times k_1 and k_2 respectively.

[b] Symbols A and B denote curves A and B in Fig. 1.

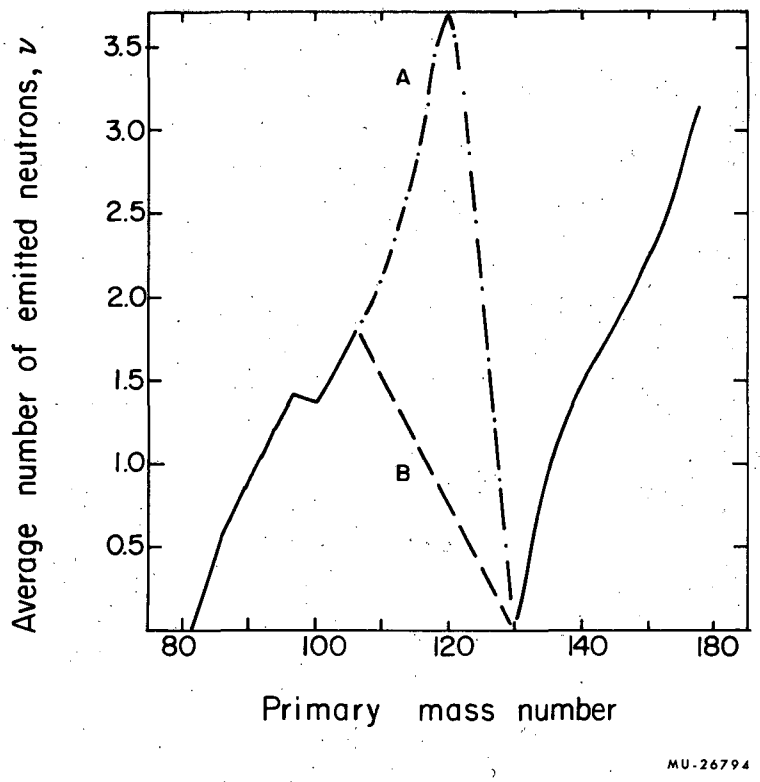

Fig. 1. Number ν of emitted neutrons versus mass number of the primary fragments. Curve A is from reference 12 and is based on Cf^{252} data. Curve B is an alternate proposal based on shell structure considerations.

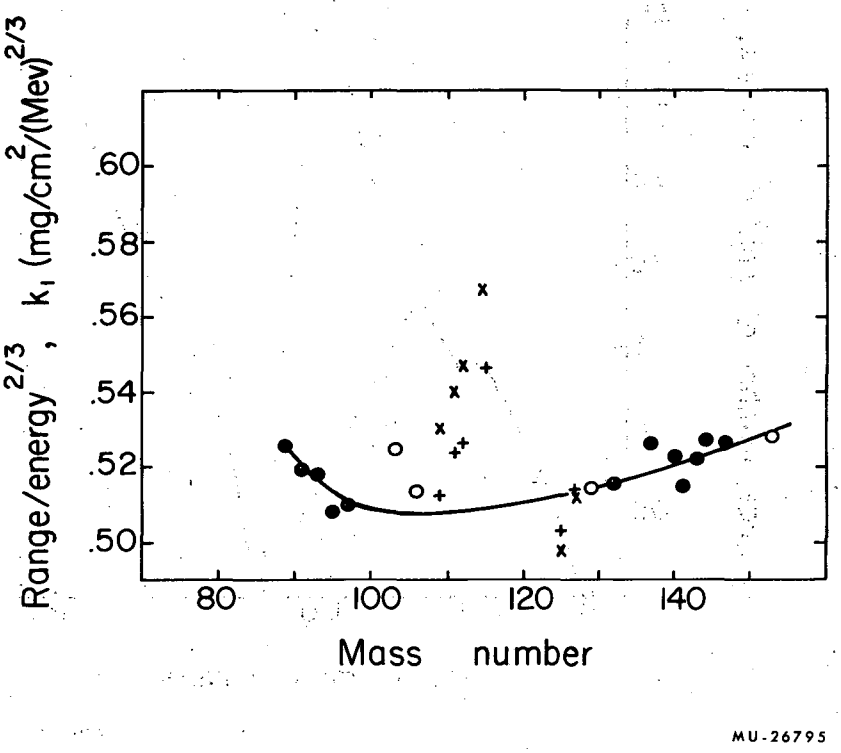

Fig. 2. Average range in U divided by kinetic energy to the 2/3 power. The range data are from reference 9 multiplied by the factor 0.970 to correct for scattering. The final kinetic energies were obtained from primary kinetic energies in reference 12. Fig. 1 shows ν values. Symbols give the reliability of the points as follows: ● product of high yield with measured ν; o product of low yield with measured ν; x product of very low yield with ν taken from curve A; + product of very low yield with ν taken from curve B.

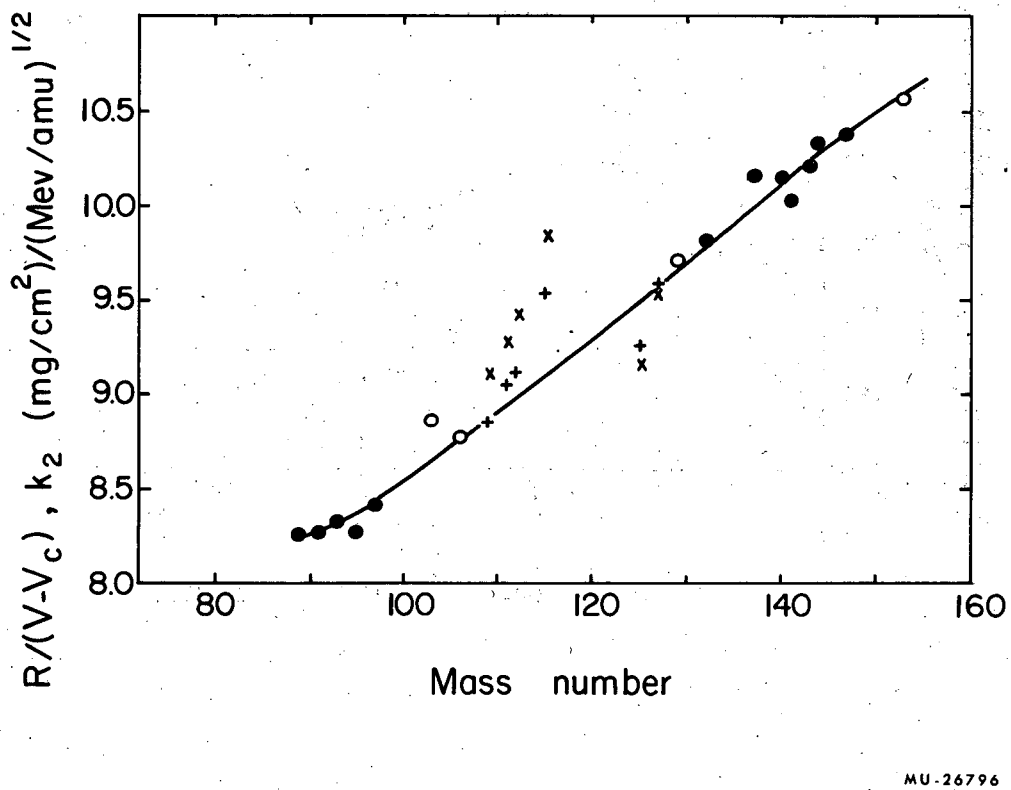

Fig. 3. Average range R in U divided by initial velocity V minus critical velocity V_c. The value of V_c was taken as 0.13 $(\text{Mev/amu})^{1/2}$. Initial velocities were obtained from reference 12. Symbols are as in Fig. 1.

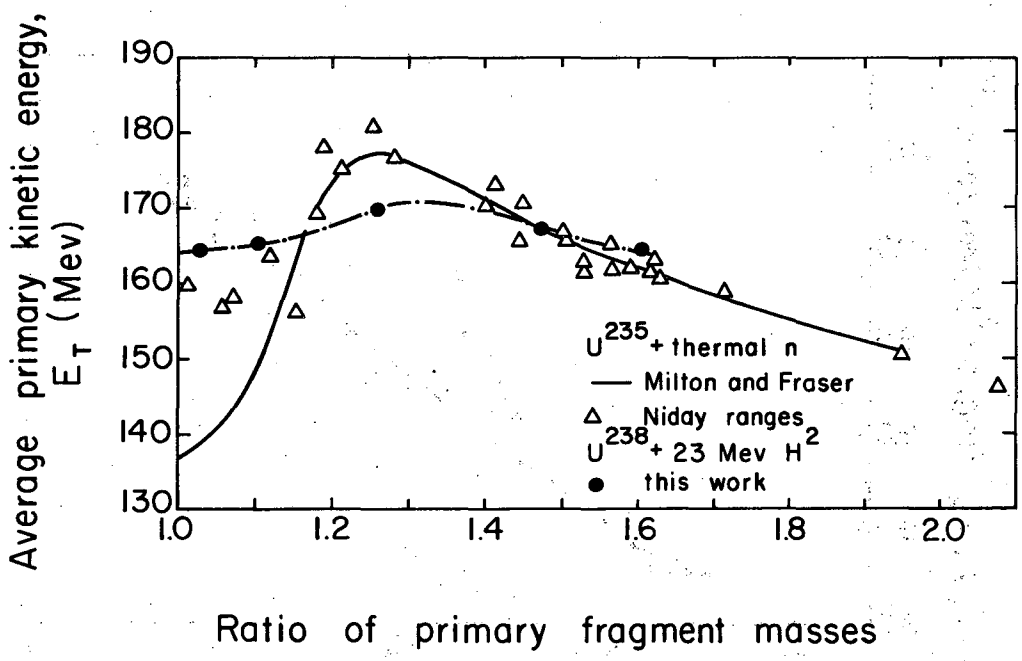

Fig. 4. Average total kinetic energy before neutron emission versus primary mass ratio. Symbols are as follows: U^{235}+n, solid line from reference 12, open triangles from range data of reference 9 and ν values from curve A of Fig. 1; U^{238} + 23 Mev H^2, closed circles and dot dash line, range data from this work and all ν values taken as 3.0. Ranges taken as proportional to $E^{2/3}$.