Velocities of Fragments from Fission of U^{233}, U^{235}, and Pu^{239}†

R. B. LEACHMAN
University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico
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The velocity distributions of fragments from slow neutron induced fission have been measured by a time-of-flight method. When compared with data from fission fragments stopped in ionization chambers filled with argon and carbon dioxide, these velocity data indicate that the kinetic energies of the fragments exceed those reported by ionization chamber measurements by 5.7 Mev for the most probable light fragment and by 6.7 Mev for the most probable heavy fragment. These energy differences, for the most probable light and heavy fragments, can be explained by energy-ionization ratios which exceed by 6 percent and 11 percent, respectively, the alpha-particle energy-ionization ratio on which the energies from ionization chamber measurements are based. The average kinetic energy of fission fragments determined from velocities is shown to be in good agreement with calorimeter results and the energy calculated from recent mass spectrographic data.

The widths of the peaks of the directly measured velocity distribution, for which the resolution is known, are appreciably narrower than those calculated from ionization chamber data. These differences in width are used to estimate the resolution of ionization chamber measurements of fission fragment energies.

I. INTRODUCTION

THROUGH analyses of the ionization data obtained from the careful and extensive measurements of fission fragments by Brunton *et al.*,[1,2] and by Deutsch and Ramsey[3] with double ionization chambers filled with argon and a few percent of carbon dioxide, data have been reported on the average kinetic energy of the fragments, the distribution in the energy, and the distribution in the fragment masses. However, all comparisons of these results with fission fragment data obtained by other means have disagreed by more than the uncertainties in the measurements. One of these disagreements is a lower average kinetic energy of the fragments reported from measurements with ionization chambers than from a calorimetric measurement of the fission energy.[4] Another disagreement which arises from this energy value has been pointed out by Brunton,[5] who showed that the energy distribution from ionization chamber data, combined with mass spectrographic data, leads to a distribution in nuclear charge of the fragments considerably different from the observed[6] charge distribution. Furthermore, a comparison[7] between the mass distributions obtained from radio-chemical measurements and from ionization chamber data has shown that the peaks of the latter distribution are wider and are further separated, in each case by more than the experimental uncertainties.

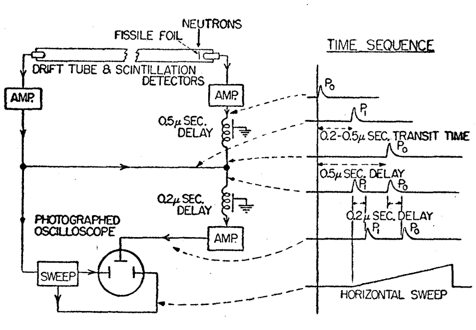

FIG. 1. Schematic diagram of the time-of-flight equipment. The time sequence illustrates that the less frequent pulses P_1 from the fragments which travel the length of the drift tube initiate the oscilloscope display, the pulses P_0 from the complementary fragments are delayed by the maximum transit time, and the mixture of P_1 and P_0 are, in addition, delayed for proper oscilloscope presentation.

An explanation for the differences between the energy values was recently advanced by Knipp and Ling[8] by showing that, because of their larger ionization defects, fission fragments stopped in a gas may be expected to expend larger averages of energy per ion pair, and thus have higher energy-ionization ratios, than alpha-particles stopped in the same gas. Since the energies from ionization chamber data were based on the relative ionization produced by alpha-particles of known energy and fission fragments stopped in the same gas, this difference in energy-ionization ratios leads to apparent values for fission fragment energies that are lower than actual. In addition, Knipp and Ling showed that the energy-ionization ratio of a heavy fragment should exceed that of a light fragment. This difference in the energy-ionization ratios of the heavy and light fragments has been used by the author[7] to explain the different spacings in the mass peaks. In this analysis it was also shown that the greater width of the mass peaks from ionization chamber data can be explained by an 8-Mev half-width (full width at half-maximum) in the

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[5] D. C. Brunton, Phys. Rev. **76**, 1798 (1949).
[6] Glendenin, Coryell, and Edwards, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 52, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[7] R. B. Leachman, Phys. Rev. **83**, 17 (1951).
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resolution of fission fragment energies in ionization chambers.

The present investigation was undertaken to determine quantitatively these ionization properties of fission fragments by comparing directly measured distributions in velocity with the velocity distributions calculated from the data from ionization chambers. In this comparison, the displacements in velocity between the peaks in the two distributions can be evaluated in terms of the energy-ionization ratios of fission fragments, while the relative widths of the velocity peaks can be used to estimate the resolution in the energy measurements of fission fragments in ionization chambers.

II. EQUIPMENT

As shown schematically in Fig. 1, velocities of fission fragments were measured by their time of flight through an evacuated drift tube. The time origin of each measurement is provided by the pulse P_0 from the fission fragment traveling the 1-cm distance from the fission source to the nearer detector. The time of flight of the complementary fragment through the 343-cm drift distance is the time until the occurrence of P_1, the pulse from the remote detector. Fission was induced by a beam of thermal neutrons from a reactor. Because the nearer detector subtended the larger solid angle from the fission source and because this detector was nearer the beam emitted by the reactor, its counting rate of the pulses from the source alpha-particles, reactor gamma-rays, and fission fragments was much greater than that of the remote detector. Since the scintillation detectors gave pulses not greatly different in amplitude for all these particles, no attempt was made to discriminate between the fission pulses and the other pulses. In order to decrease the number of the recorded data, the less frequent pulses P_1 from the remote detector were used to initiate the oscilloscope displays of the pulses. By means of a projector, photographs of these sweeps were analyzed for the distribution in times between pulses.

In order to provide good resolution, the equipment was designed to give rise times of pulses small compared to the flight time of the fragments. The pulses obtained from mosaics of anthracene crystal shavings on 5819 photomultipliers were amplified by Model 460A and 460B Hewlett-Packard amplifiers and delayed by lengths of RG 7/U cable. The resulting pulse rise times of $\sim 10^{-8}$ sec were short compared with the 0.2 μsec to 0.5 μsec flight time of fragments through the 343-cm drift distance. An accurate determination of the time resolution, as well as of the difference in the delays in the detectors, amplifiers, and cables of the two pulses, was made from the distribution in time between fragments having traveled equal 1-cm distances in a short drift tube. With these pulses on the same $1.6(10^7)$ cm/sec oscilloscope sweep and measured in the same manner as for pulses from the longer drift tube, the resolution was found to be Gaussian with a $1.0(10^{-8})$ sec half-width. The time scale was provided by photographs of a 50-Mc signal from an oscillator at frequent intervals on the film containing flight time data.

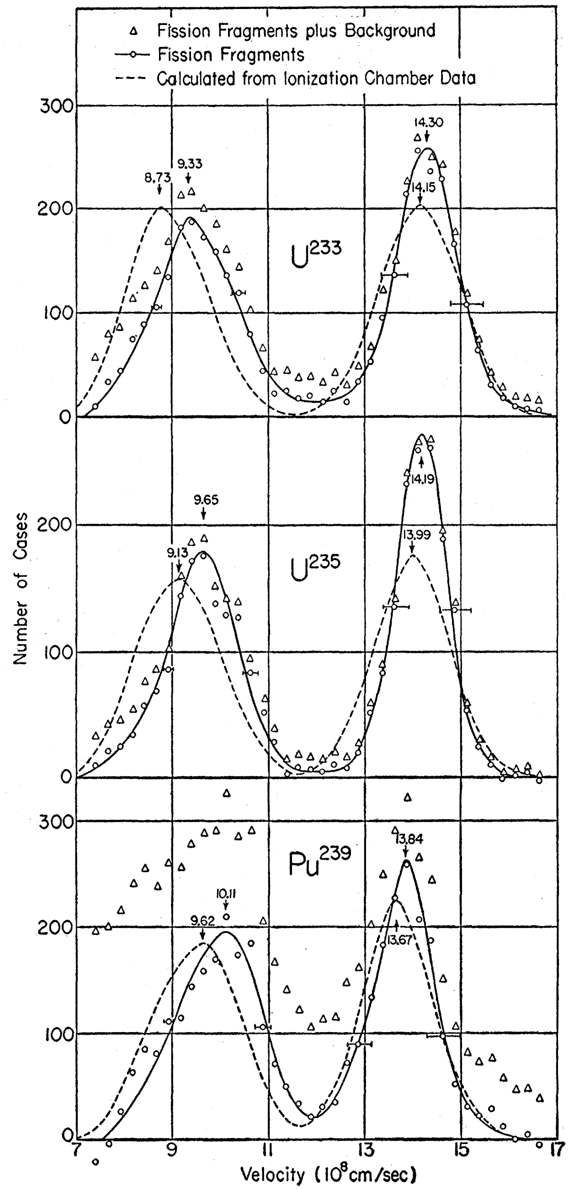

FIG. 2. Comparison of the velocity distribution of fission fragments with the distribution inferred from ionization chamber data. All distributions are normalized. Horizontal lines through the data represent half-widths of the resolutions of the velocity measurements. The large background in the Pu^{239} data is due to the large alpha-activity of Pu^{239}.

The fission sources consisted of UO_3 or PuO_2 on 1.1 mg/cm^2 nickel backings placed so that the backing faced the nearer detector. Estimates of the combined energy loss of each fragment in the 20-μg/cm^2 to 50 μg/cm^2 sources and the residual air in the drift tube are 0.9 Mev for U^{233}, 0.6 Mev for U^{235}, and 0.8 Mev for Pu^{239}.

TABLE I. Fission fragment quantities determined from a comparison between the present data and data from ionization chambers. On the basis of the present data, the energy differentials ΔE_L and ΔE_H should be added to the respective energies reported by ionization chamber experiments.[a] These corrected energies would be obtained from ionization by the use of the most probable energy-ionization ratios w_L and w_H, rather than by w_α of alpha-particles.

	U^{233}	U^{235}	Pu^{239}
ΔE_L	6.1 Mev	5.7 Mev	5.2 Mev
ΔE_H	7.3 Mev	6.5 Mev	6.4 Mev
w_L	1.06 w_α	1.06 w_α	1.05 w_α
w_H	1.13 w_α	1.11 w_α	1.10 w_α

[a] See references 1 and 2.

III. RESULTS

Shown by triangles in Fig. 2 are the velocity data including the background, which is mainly due to the occurrence of alpha-particle pulses P_1 from 0.2 μsec to 0.5 μsec after the occurrence of alpha-particle or gamma-ray pulses P_0. Since the probability of such events is constant with time, the background varies as v^{-2}. That this v^{-2} relation applies for the background from alpha-particles was confirmed by the use of the Pu^{239} source in the absence of neutrons. When the backgrounds are subtracted from the data, the fission fragment velocity distributions of the solid lines through circles in Fig. 2 are obtained. It is required of the total background that the resulting probabilities of the highest and lowest velocities in Fig. 2 approach zero.

Since these velocity data are of individual fragments and not of fragment pairs, a correlation with fragment mass as required for energy determinations is not possible without additional data. For this reason, comparisons with the fragment pair data from ionization chambers are used for energy determinations. The data used for these comparisons are those of Brunton and Hanna[1] for U^{233} and U^{235} and those of Brunton and Thompson[2] for Pu^{239}, all of which data are based on the energy-ionization ratio w_α of alpha-particles. Shown as broken lines in Fig. 2 are the velocity distributions calculated from these data from double ionization chambers. Included in these calculations are small mass corrections made for the neutrons emitted per fission and energy corrections for these authors' estimates of source and collimator losses.

Use the subscripts L and H to refer to the light and heavy fragments, respectively, and we now write the relations between the differences in the velocity distributions in Fig. 2 and the corresponding energy and mass differences. For the light fragments this relation is

$$\Delta E_L/E_L = \Delta m_L/m_L + 2\Delta v_L/v_L, \qquad (1)$$

where E_L, m_L, and v_L are the most probable energy, mass, and velocity, respectively, from the ionization data and ΔE_L, Δm_L, and Δv_L are the differences between these values and the most probable values from the present data. An equation relating the corresponding values for the heavy fragments is obtained by their substitution in Eq. (1). The mass differences Δm_L and Δm_H in these relations are obtained from the momentum relations

$$m_L v_L = m_H v_H,$$
$$(m_L+\Delta m_L)(v_L+\Delta v_L) = (m_H+\Delta m_H)(v_H+\Delta v_H), \qquad (2)$$

where $\Delta m_L = -\Delta m_H$ and for U^{235} fission $m_L+m_H=236$.

The most probable velocities indicated in Fig. 2 are first used in Eq. (2) to solve for the mass differentials for the three cases of U^{233}, U^{235}, and Pu^{239}. It can be shown that when these mass differentials are applied to the masses from ionization data the corrected masses are in reasonable agreement with the masses from radiochemical data. When the mass and velocity data are combined in Eq. (1) and the corresponding equation for the heavy fragments, the energy differentials ΔE_L and ΔE_H are obtained. These energy differentials applied to the ionization data would result in calculated velocity distributions with the same most probable velocities as the present data.

In Table I are these results corrected for the source and residual air losses discussed above. Also in Table I are w_L and w_H values calculated from

$$w_L = w_\alpha(E_L+\Delta E_L)/E_L$$

and the similar relation for w_H. Since the mass and energy distributions of the fragments from U^{233}, U^{235}, and Pu^{239} fission are not greatly different, the values in Table I should be nearly the same for the three cases. On this basis, the variation in values between the three cases is a good indication of the accuracy of the results.

Although less direct and less sensitive than the comparison[7] between ionization and mass distributions, comparisons of the widths in Fig. 2 of the velocity peaks with the usually wider peaks inferred from ionization data can be used to estimate the energy resolution of the latter data. The difference in widths is greater for the higher velocity peaks because dispersion in the measurements with ionization chambers broadens the narrower peaks of the light fragment energies more than that of the heavy fragments.

Comparison of the velocity widths to obtain the energy resolution of the ionization chamber measurements is complicated by two other resolutions that are involved. One is the velocity resolutions of the present data which are calculated to be $0.59(10^8)$ cm/sec and $0.24(10^8)$ cm/sec half-width for the most probable light and heavy fragments, respectively. In addition, due to the use of a finite number of velocity intervals in the conversion of the ionization data to velocities, a dispersion of $0.25(10^8)$ cm/sec half-width is present in the velocity data from ionization data. With allowance made for these dispersions, the half-widths of the energy resolutions of the ionization chamber data estimated by this velocity-ionization analysis are about 9 Mev and so are in rough agreement with the 8-Mev half-width determined from the previous mass-ionization analysis.

IV. DISCUSSION

When the energy differentials found from the present data are applied to the ionization data, all the above disagreements with other data are considerably reduced. To illustrate these corrections the averages $\langle\Delta E_L\rangle_{\text{Av}}=5.7$ Mev, $\langle\Delta E_H\rangle_{\text{Av}}=6.7$ Mev, and $\langle w_L/w_H\rangle_{\text{Av}}=0.95$ of the quantities in Table I are used.

With this total energy differential of $\langle\Delta E_L\rangle_{\text{Av}}+\langle\Delta E_H\rangle_{\text{Av}}=12.4$ Mev added to the 154.7 Mev reported[1] for the average total kinetic energy of the fragments from U^{235}, the corrected value of 167.1 Mev is in excellent agreement with the calorimetric value[4] of 165±8 Mev.

This corrected value for the total kinetic energy of the fragments is also confirmed by a calculation based on mass spectrographic data. For the most probable fission mode the most probable corrected energy is 168 Mev, while this energy calculated from mass spectrographic data is 174 Mev. Based on the method of Brunton,[5] this calculation of energy from spectrographic masses uses the equation

$$E_K=931[m(U^{235})-m(A_L,Z_L)-m(A_H,Z_H)-(\nu-1)m(n^1)]-\nu E_n-E_\gamma \quad (3)$$

to obtain the kinetic energy of the fission fragments E_K as a function of the nuclear charges of the fragments, Z_L and Z_H. The measured distribution[6] in nuclear charges is applied to this function. The resulting 174-Mev value is based on the use of $\nu=2.5\pm0.1$ for the average number of neutrons emitted per fission,[9] $E_n=2.0\pm0.1$ Mev for the average energy of these neutrons,[10] and $E_\gamma=4.6\pm1.0$ Mev[11] for the average energy of the prompt gamma-rays. The masses $m(A_L,Z_L)$ and $m(A_H,Z_H)$ of the unstable fragments which have emitted their neutrons, and are in the ground state, are obtained by an extrapolation[12] from the spectrographic masses of the stable atoms[13] by the parabolic mass-charge relation for isobars. The masses of the stable atoms are known with a probable error of ±1 mMU. The mass of U^{235}, $m(U^{235})$, is known with a probable error of ±2 mMU from the spectrographic mass[14] of Pb^{208} and the $U^{235}-Pb^{208}$ mass difference.[15] The neutron mass is represented in Eq. (3) by $m(n^1)$. In regard to the uncertainties in the charge distribution, it is seen by Brunton's analysis that E_K is not sensitive to reasonable variations from the measured distribution.

Although the 6-Mev difference between the result of $E_K=174$ Mev from Eq. (3) and the 168±2-Mev value from velocities is somewhat large in view of the known errors of the data used in the equation, the 18-Mev difference between 174 Mev and the corresponding 156 Mev reported from ionization data is far greater than these known errors.*

That the results of this analysis of velocities bring the mass and ionization data into agreement is seen by the agreement of these results with those derived from the comparison[7] of mass and ionization data. This latter comparison showed that if $w_L/w_H=0.96$ the separation of the two sets of peaks agreed and if the resolution of the ionization chamber measurements were 8 Mev the widths of the peaks agreed. This is in reasonable agreement with the present results of $\langle w_L/w_H\rangle_{\text{Av}}=0.95$ and a resolution of roughly 9 Mev half-width.

The energy differentials $\langle\Delta E_L\rangle_{\text{Av}}=5.7$ Mev and $\langle\Delta E_H\rangle_{\text{Av}}=6.7$ Mev do not by themselves confirm the respective 2.5-Mev and 4.2-Mev ionization defects calculated by Knipp and Ling[8] from limited data. Instead, the energy differentials may be wholly explained by linear energy-ionization relations for fission fragments with different slopes from that for alpha-particles, rather than by a nonlinear energy-ionization relation as required by the ionization defect theory. In addition, it is possible that a large part, if not the whole, of the energy differentials are due to such difficulties with ionization measurements as recombination and negative ion formation.

However, in view of the close agreement in the results of the many[16] ionization chamber measurements of fission fragments, experimental errors of this magnitude are unlikely. Because of this agreement in ionization data, it also is unlikely that an appreciable part of the estimated 8-Mev resolution in these data is due to instrumental difficulties.

These considerations make reasonable the belief that the energy differentials and the large dispersion are inherent in the ionization process. Furthermore, the fact that the dispersion is far greater than that anticipated by the theory[17] of the fluctuations in the number of ions produced by charged fragments indicates that a large part of the energy differentials can be attributed to ionization defects. On the basis of the ionization defect theory, this dispersion is explained by each fragment losing several Mev of energy to recoiling gas atoms which have a reduced ionization efficiency. Fluctuations in the number of recoiling atoms and in their ionization efficiency would result in relatively large fluctuations in the number of ions produced.

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* *Note added in proof.*—If the mass of U^{235} determined by Stanford, Duckworth, Hogg, and Geiger, Phys. Rev. **85**, 1039 (1952) is used in Eq. (3), the value $E_k=171$ Mev is obtained. This is in satisfactory agreement with the value from velocities.

16 See reference 1 for a listing of earlier work.

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