in the binary range curve to account for our results. Since sources (2) and (3) cannot be completely discounted, the yields we report should be regarded as upper limits for production of long-range fission fragments.

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Mass-Spectrometric Measurements of ^{3}H, ^{3}He, and ^{4}He Produced in Thermal-Neutron Ternary Fission of ^{235}U: Evidence for Short-Range ^{4}He

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The mass-spectrometric technique has been used to obtain relative and absolute yields for ^{3}H, ^{3}He, and ^{4}He and energy distributions for ^{3}H and ^{4}He emitted in ternary fission of $^{236}U^*$. Evidence has been obtained for a short-range (<7.7-MeV) component of ^{4}He. The $^{3}He/^{4}He$ ratio is lower by 2 to 4 orders of magnitude than the values previously found in studies of other fissile nuclides. $^{3}H/^{4}He$ is lower by about a factor of 2 than values obtained for $^{236}U^*$ using other experimental techniques. Energy distributions obtained for long-range ^{3}H and ^{4}He particles are in agreement with results from other experiments.

I. INTRODUCTION

Recent experimental studies on light charged-particle emission in ternary fission have been conducted almost exclusively with semiconductor counter telescopes measuring dE/dx and E for each particle registered. Particle identification is obtained from range-energy characteristics, and using such techniques energy and angular distributions as well as correlations between these two have been measured. Absolute and relative yields have also been determined. To date, ^{1}H, ^{2}H, ^{3}He, ^{4}He, ^{6}He, ^{8}He, and Li, Be, B, C, N, and O ions have been observed to be emitted in both $^{236}U^*$ and ^{252}Cf fission.[1]

The low-energy region (below ~6 MeV for ^{4}He) of the energy spectrum of these particles is not seen in experiments using semiconductor detectors. This is due to necessary shielding of the detectors from natural α particles, heavy fission fragments, or other possible effects. Early experiments using proportional counters in coincidence, and nuclear emulsions, on the other hand, were able to observe the energy spectrum in this lower region and suggested the existence of a "short-range" component of light charged particles, emitted with relatively high yield.[2-7] The obvious experimental difficulties involved in identification and measurement of energy and frequency of emission have, however, left the question of the existence of these "short-range" particles in doubt. In the present work a technique was developed which enables the detection and positive identification of at least some possible short-range light ternary-fission products.

The yield of ^{3}He in ternary fission is also considered of some interest. According to a model suggested by Halpern,[8] a correlation between relative yields and release energies of the light particles is expected. Whetstone and Thomas[9] have measured relative yields and estimated release energies for several light particles emitted in ^{252}Cf fission and have found a rough dependence of measured yield according to $e^{-E_r/T}$, where E_r is the release energy, and T corresponds to a nuclear temperature. Using their estimates of release energies for ^{3}He, ^{4}He, and Li and Be isotopes (and assuming the $e^{-E_r/T}$ dependence), a ratio of ^{3}He/^{4}He of $\sim 4\times10^{-4}$ is predicted. A precise determination of the ^{3}He/^{4}He ratio would therefore serve to test the yield–release-energy correlation over several orders of magnitude. Also, ^{3}He is estimated to be energetically favored over any Li or Be isotopes and is therefore expected to have higher yield. For ^{252}Cf, Cosper, Cerny, and Gatti[10] have shown, however, that Li and Be ions are emitted in greater abundance than ^{3}He (measured ^{3}He/^{4}He $\leq 7.5\times10^{-4}$). On the other hand, Cambiaghi, Fossati, and Pinelli[11] have found a much higher yield for ^{3}He (^{3}He/^{4}He $= 1.8 \times 10^{-2}$) than for total Li and Be ions produced in fission of ^{234}U*. The results of Whetstone and Thomas, Cosper, Cerny, and Gatti, and Cambiaghi, Fossati, and Pinelli were obtained using systems of dE/dx and E detectors, and complete discrimination of ^{3}He from the much more abundant ^{4}He is difficult using these methods. In the work described here a technique employing high-sensitivity mass spectrometry was used to measure the relative yields of ^{3}H, ^{3}He, and ^{4}He in thermal-neutron fission of ^{235}U.

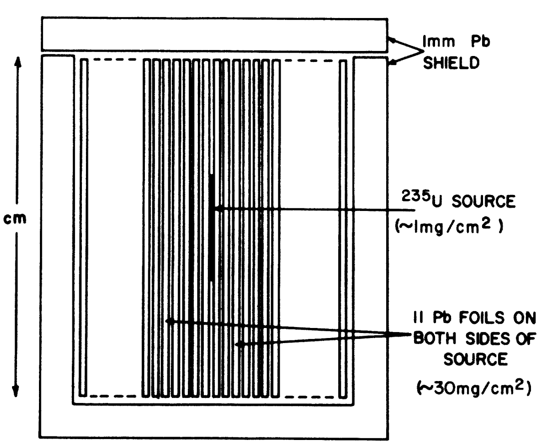

FIG. 1. Assembly of ^{235}U fission source and Pb foil stack.

A review of experimental results[12-20] published on the absolute yield of ^{4}He emitted in ternary fission of ^{236}U* shows that these vary by more than a factor of 2. The present series of experiments were therefore carried out in an attempt to obtain an accurate value for this yield.

II. EXPERIMENTAL

The experiment consisted essentially of measuring mass-spectrometrically the abundances of ^{3}He and ^{4}He present in the Pb catcher foils surrounding a ^{235}U fission source for a given period of time. ^{3}H is detected as ^{3}He, after partial decay. Samples of ^{235}U (93.18%) weighing a few tenths of 1 mg were prepared by evaporation of weak uranyl nitrate solution onto Pb foil. The thickness of the fission source was $\sim$1 mg/cm^2 and permitted recoil of virtually all fission fragments into the Pb catcher foil. Two different arrangements were employed, the thick catcher foil and the stacked foil assembly. The former was used to measure relative and absolute yields of ^{3}H, ^{3}He, and ^{4}He, and the latter was used to obtain information on possible "short-range" components.

A. Thick Catcher Foil Experiment

Pb foils, 30 mg/cm^2 thick, were placed together to give a total thickness of 330 mg/cm^2, sufficient to stop normally incident α particles and tritons of 40 and 14 MeV, respectively. The thick foil assembly was folded around a ^{235}U source (deposited directly on the innermost foil), sealed in a quartz ampul, along with a blank Pb foil, and irradiated for about 10 days in the McMaster reactor (flux $\sim1.5\times10^{13}$ neutrons/cm^2 sec). Some samples were wrapped with Cd ($\sim$1 mm thick) to permit determination of effects due to epithermal neutrons. Further "control" samples consisted of depleted ^{238}U prepared in an identical manner to the ^{235}U samples. Pb foils were chosen because of the relatively low (n,α) and (n,t) cross sections of Pb. Another factor in the choice of Pb, bearing on the method of extraction of He from the foils used, was its relatively low melting and boiling points.

B. Stacked Foil Experiment

In order to detect possible short-range components of the light particles, stacks of Pb foils were arranged in such a way that He contents could be measured for each foil individually. The method is similar to that used by Douthett and Templeton.[21] The geometrical arrangement is shown in Fig. 1. This method, as adapted for the present work, enables measurement of integral-range distributions. Knowledge of range-energy relations can then be used to yield energy distributions. The method has the advantage that it requires no collimator, and hence greatly improved sensitivity is attained. It can be shown[22] that the number of particles found in the ith foil is

$$N_i = A + B\left\{\Delta t_i \int_{E(t_i)}^{\infty} \frac{n(E)dE}{R(E)} + \int_{E(t_{i-1})}^{E(t_i)} \frac{[R(E)-t_{i-1}]n(E)dE}{R(E)}\right\}\dots, \tag{1}$$

where A is the constant background found in each foil, B is a constant depending on source strength, Δt_i is the thickness of the ith foil, t_i is the total foil thickness up to and including the ith foil, $E(t_i)$ is the energy of a particle with range t_i, $n(E)$ is the energy distribution of particles, and $R(E)$ is the range of a particle with energy E. The parameters for $n(E)$ are determined when the measured data points N_i are fitted with Eq. (1). N_i was obtained from mass-spectrometric measurement of the amounts of ^{3}He and ^{4}He present in each foil. t_i and Δt_i were determined by accurately weighing foils of known area. The range-energy data, $R(E)$, were taken from the compilation by Williamson, Boujot, and Picard.[23]

C. Extraction of He from Pb Foils

Helium as well as fission-product Kr and Xe were extracted from the Pb foils by vaporizing these in a vacuum. The Pb foils were dropped into a Mullite tube sealed onto the sample inlet system of the mass spectrometer. The tube was held at 750°C with a resistance furnace. Helium was allowed to expand into the mass-spectrometer volume (^{3}H was removed completely by the use of titanium sponge at room temperature), whereas fission Kr and Xe were condensed on activated charcoal in a sample recovery tube held at −196°C.

D. Mass Spectrometry

The mass spectrometer is a 10-in. radius first-order direction-focusing instrument with a resolving power of 620. The sensitivity and background are such that measurements with a few percent precision can be made on ~10^{12} atoms of ^{4}He and ~2×10^6 atoms of ^{3}He. At these levels the ^{4}He background signal is about 15% of the sample ^{4}He signal. The ^{3}He background was barely measurable, and any sample ^{3}He was completely resolved from HD and H_3 in the spectrometer (the HD-H_3 peak was typically about 5 times that of ^{3}He). Samples analyzed contained about 5×10^{12} and 5×10^7 atoms of ^{4}He and ^{3}He, respectively.

Calibration of the mass spectrometer for peak-height response and mass discrimination was achieved by analyzing atmospheric He prepared from aliquots of air of known volume introduced immediately after each fission He sample. A concentration for He in the atmosphere of 5.24 ppm,[24] and a ratio of $(^3\text{He}/^4\text{He})_{\text{atm}} = 1.37\times10^{-6}$ were assumed.[25] The number of fissions in each sample was determined by absolute measurements of the fission ^{86}Kr content. A yield for ^{86}Kr in thermal-neutron fission of ^{236}U* of 2.04% was assumed.[26]

III. RESULTS

A. Relative Yields of ^{3}H, ^{3}He, and ^{4}He

It was anticipated originally that measurement of ^{3}He/^{4}He as a function of cooling period (time elapsed between end of irradiation of ^{235}U and extraction of He from catcher foils) should give the contributions to ^{3}He due to independent formation of ^{3}He in fission and due to β decay of ^{3}H produced in fission. Seven identically prepared samples were irradiated in the reactor for 10 days, but were permitted to cool for different periods of time, ranging from 7.5 to 39 days. Owing to the relatively long half-life of ^{3}H (12.26 yr), the ratio ^{3}He/^{4}He is expected to increase linearly over these short cooling periods. (^{3}H will decay *in situ* in the Pb catcher foils and is extracted and detected in the mass spectrometer as ^{3}He.)

Figure 2 shows the results of measurements of ^{3}He/^{4}He as a function of cooling period. The least-squares fit to the data points produces a straight line with intercept and slope of $(1.38\pm0.29)\times10^{-5}$ and $(0.0103\pm0.0007)\times10^{-5}$/h, respectively. The slope of the straight line is determined by the ^{3}H/^{4}He ratio as well as $T_{1/2}$ for ^{3}H. The intercept, on the other hand, depends on $(^3\text{H}/^4\text{He})_{\text{fission}}$, $T_{1/2}$ of ^{3}H, and a possible component of $(^3\text{He}/^4\text{He})_{\text{fission}}$ due to direct formation of ^{3}He. Using the experimentally obtained slope (solid line in Fig. 2) and $T_{1/2}$ (^{3}H) = 12.262 yr,[27] one calculates $(^3\text{H}/^4\text{He})_{\text{fission}} = 1.60\times10^{-2}$, which in turn yields an intercept at $t=0$ of 1.27×10^{-5} (dotted line in Fig. 2). The difference between this value and the experimental intercept may be taken as the component of ^{3}He/^{4}He due to direct formation of ^{3}He, that is (^{3}He/

$^4He)_{fission} = (1^{+3}_{-1}) \times 10^{-6}$. Alternately, the observed ^3He may be entirely due to ^3H decay. Assuming that this is the case, the dashed line in Fig. 2 was obtained by correcting each datum point for growth of ^3He from ^3H, calculating a $t = 0$ intercept from the weighted mean, and generating a slope using the known half-life of ^3H. It is seen that within errors this line coincides with the least-squares straight line through the experimental points.

Assuming two components for ^3He production in fission, however, the relative yields (to be corrected for the short-range ^4He component later) were measured to be

$$^3H/^4He = (1.60 \pm 0.37) \times 10^{-2},$$

$$^3He/^4He = (1^{+3}_{-1}) \times 10^{-6}.$$

B. Frequency of Formation of ^4He

The ratio ^4He/BF (BF = binary fission) was determined from the absolute measurements of ^4He and ^{86}Kr contents. A mean value of $(4.27 \pm 0.12) \times 10^{-3}$ was obtained for ^4He/BF from 12 separate irradiations (the indicated error is 1 standard deviation of the mean). The value 4.27×10^{-3} represents the total ^4He produced.

C. Energy Distributions of ^3H and ^4He

^3H and ^4He contents were measured in each of 11 Pb foils stacked against a ^{235}U fission source (see Fig. 1). The relative amounts of ^3H in each foil were obtained from measurements of the ^3He found in each foil. For the cooling times used in this experiment it was possible to assume that all observed ^3He was due to *in situ* decay of ^3H. Three foil stack assemblies were analyzed, two having been placed on opposite sides of the same fission source. Results of measurements on ^4He and ^3H from one foil stack are shown in Figs. 3 and 4, respectively.

Earlier work on energy spectra of light charged particles from fission[28,29] has shown that these distributions can be fitted adequately with a Gaussian function. Accordingly, the data points in Figs. 3 and 4 were fitted with Eq. (1), where

$$n(E) \sim \exp[-(E - C_1)^2/2C_2^2].$$

The solid curves in Figs. 3 and 4 represent a least-squares fit to the data points when the first points are omitted in each case. The first point of the ^4He spectrum reflects an obvious departure from the assumed Gaussian energy distribution and is considered evidence for a short-range component. The ^3H spectrum also suggests the presence of a short-range component, although less convincingly. The constants C_1 and C_2 of the assumed Gaussian energy distributions were obtained from the best least-squares fit to the measured integral-range distributions. In Figs. 5 and 6

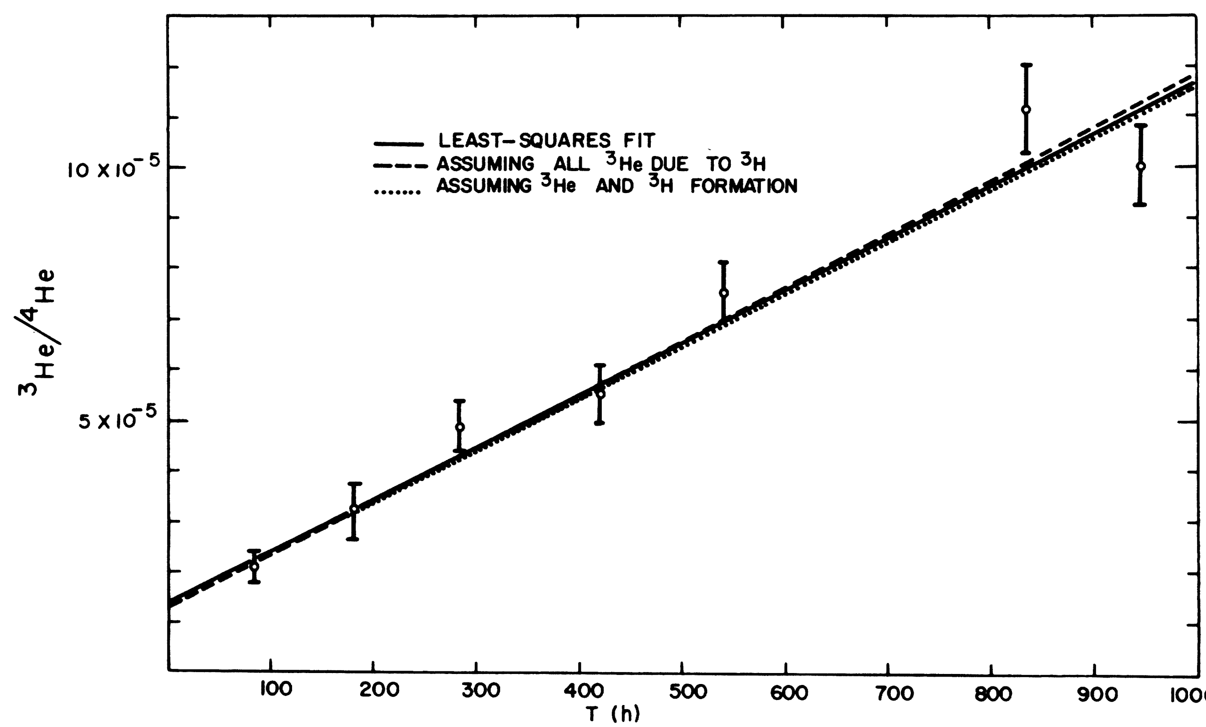

FIG. 2. Variation of $^3He/^4He$ with cooling period. The intercept of the straight line depends on $^3H/^4He$, $T_{1/2}$ of ^3H, and on the component of $^3He/^4He$ due to independent formation of ^3He. The slope depends only on $^3H/^4He$ and $T_{1/2}$ of ^3H.

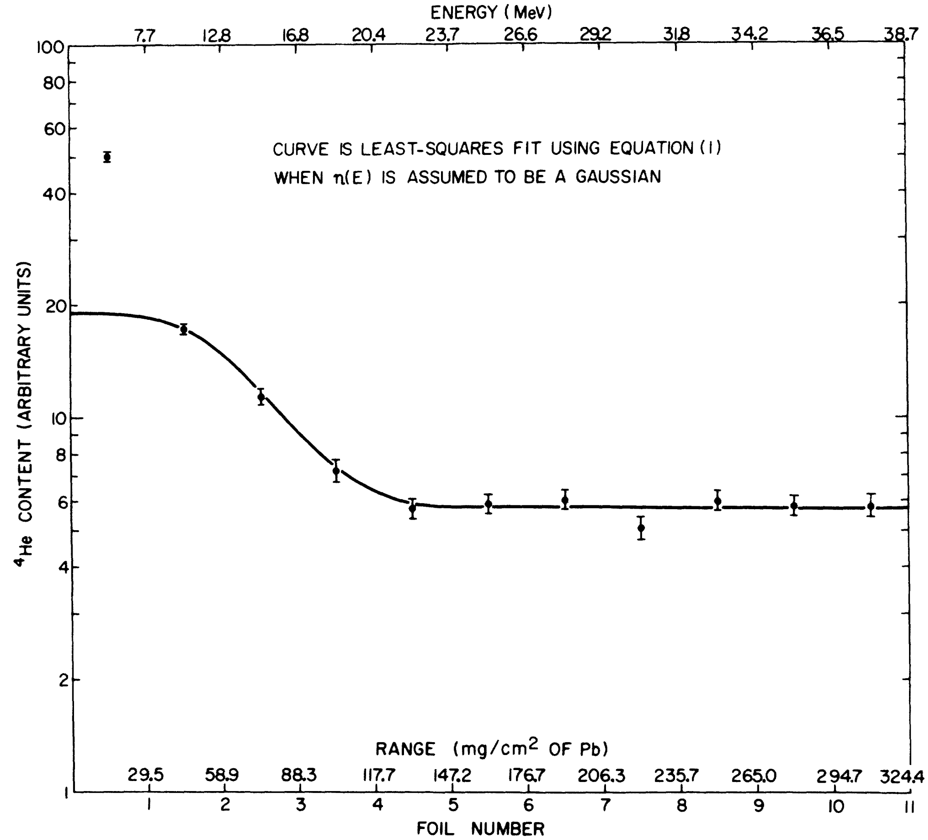

FIG. 3. Integral-range (energy) distribution for ^4He particles. The departure of the first point from the solid line is evidence for a distinct short-range component.

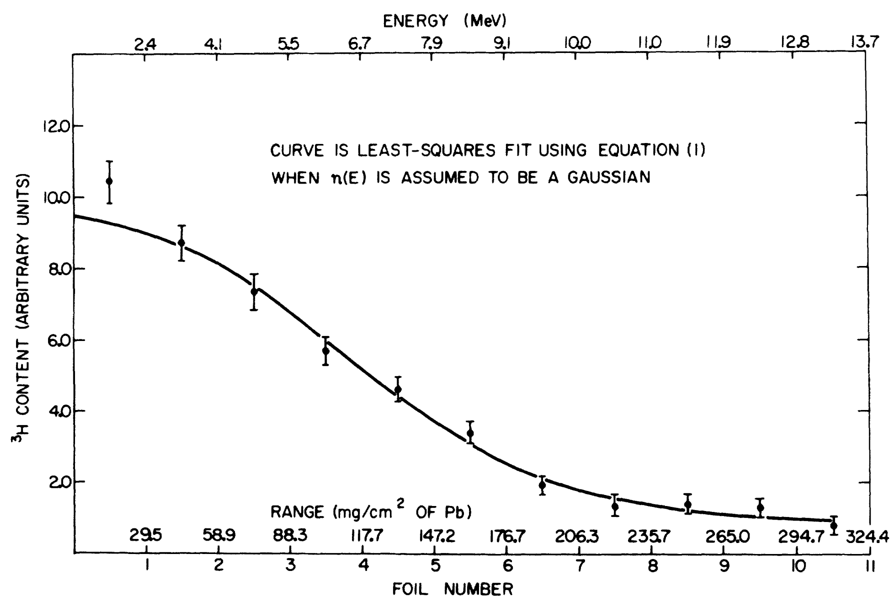

FIG. 4. Integral-range (energy) distribution for ^3H particles. The existence of a short-range component is indicated, but with much less certainty than for ^4He.

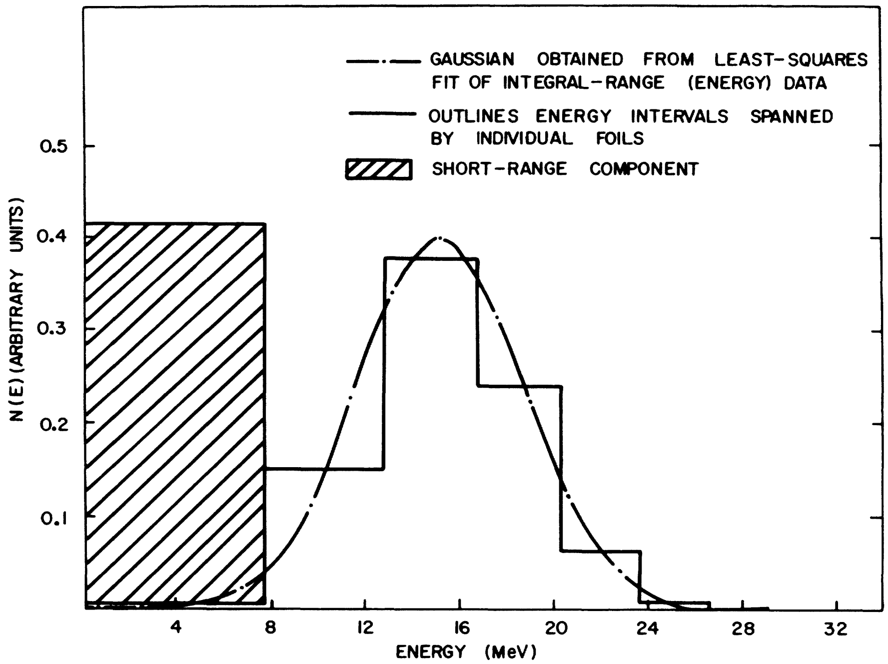

FIG. 5. Energy distribution of observed ^{4}He particles. Relative areas of shaded portion and under Gaussian indicate relative yields of short-range and long-range components.

the Gaussians obtained from the fits for ^{4}He and ^{3}H are shown, respectively. Also indicated are the short-range components. The shaded areas in each figure, when compared with the areas under the respective Gaussians, represent relative abundances of short- and long-range ^{4}He and ^{3}H.

Table I summarizes results obtained for relative and absolute yields of ^{3}H, ^{3}He, and ^{4}He when these are separated according to long-range and short-range components.

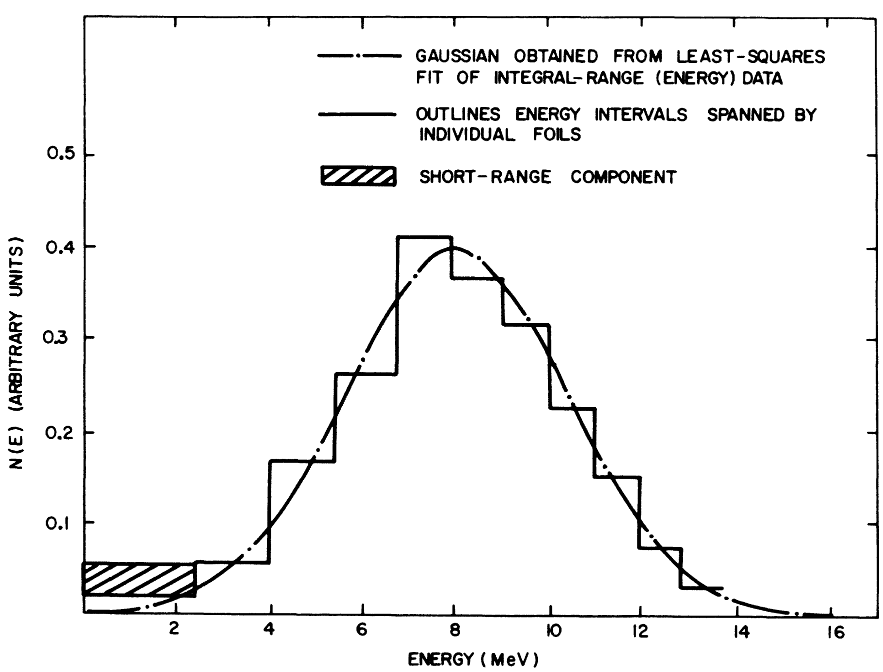

FIG. 6. Energy distribution of observed ^{3}H particles. Relative areas of shaded portion and under Gaussian indicate relative yields of short-range and long-range components.

TABLE I. Frequency of formation of long-range and short-range ^{3}H, ^{3}He, and ^{4}He in thermal-neutron fission of ^{235}U.

	Long-range particles only	Short-range [a] particles only	All particles
$^3H/^4He$	$(3.1 \pm 0.8) \times 10^{-2}$	$(1 \pm 0.6) \times 10^{-3}$	$(1.60 \pm 0.37) \times 10^{-2}$
$^3He/^4He$	$(2^{+6}_{-2}) \times 10^{-6}$	$(7 \pm 4) \times 10^{-6}$	$(1^{+3}_{-1}) \times 10^{-6}$
^4He/BF [b]	$(2.18 \pm 0.11) \times 10^{-3}$ $\equiv (1:459 \pm 22)$	$(2.09 \pm 0.10) \times 10^{-3}$ $\equiv (1:478 \pm 24)$	$(4.27 \pm 0.12) \times 10^{-3}$ $\equiv (1:234 \pm 7)$

[a] The observed short-range ^{3}He particles may be due to ^{3}H decay, or direct formation of ^{3}He, or both. The values of $^3H/^4He$ and $^3He/^4He$ are obtained when the first or second assumption, respectively, is made.

[b] BF = Binary fission.

IV. DISCUSSION

A. Origin of Short-Range Particles

1. *Neutron Reactions*

Possible contributions to short-range ^{4}He particles due to (n, α) reactions on Pb, N, O, ^{238}U, or other nuclei in the fission source assembly were investigated by irradiations under various control conditions. ^{235}U sources wrapped with Cd produced <3% of the ^{4}He observed when ^{235}U was exposed to the full neutron spectrum. The He produced with epithermal neutrons is probably due to fissions induced by resonance or fast neutrons. Contributions to observed He due to (n, α) reactions on Pb or other targets within the foils were corrected for by irradiating blank foils along with each fission assembly. The background of He found in these foils (<10% of fission He in most cases) was subtracted from total observed He in each foil. It should be noted that the results of the Cd experiment also rule out natural α decay of any isotope present in the source as a mechanism for production of short-range ^{4}He particles.

Samples of depleted ^{238}U prepared and irradiated in an identical manner to the ^{235}U samples showed no ^{4}He or ^{3}He produced above the background due to the Pb foils, and hence (n, α) reactions on ^{238}U within the source can be ruled out as a mechanism for producing the short-range ^{4}He particles.

The reaction ^{235}U(nth, α)^{232}Th remains to be considered. From the observed absolute yield of short-range ^{4}He particles and known fission cross sections of ^{235}U, it is easily calculated that the reaction ^{235}U(nth, α)^{232}Th would require a cross section of 1.2 b to produce the amounts of He observed by us. Total cross sections for this reaction have not been measured but Chwaszczewska *et al.*[28] have set upper limits of 3 and 2 mb for the transitions to the ground state and to any excited state of ^{232}Th up to 5 MeV, respectively [general trends of (n, α) cross sections for the very heavy elements produce order-of-magnitude estimates which are <1 μb]. It therefore seems highly improbable that the observed short-range ^{4}He is due to (n, α) reactions.

Data for the ^{235}U$(n, t)^{233}$Pa and ^{235}U$(n, {}^3He)^{233}$Th

TABLE II. Results from energy distribution measurements for long-range ^{4}He and ^{3}H produced in thermal-neutron fission of ^{235}U.

	^{4}He			^{3}H		
Fissioning nucleus	MPE [a] (MeV)	FWHM [b] (MeV)	Observed interval (MeV)	MPE [a] (MeV)	FWHM [b] (MeV)	Observed interval (MeV)
^{236}U [c]	15.4 ± 0.2	9.8 ± 0.4	0–>26	8.0 ± 0.2	5.5 ± 0.6	0–>14
^{236}U [d]	16.2 ± 0.5	12 ± 1	5–17.5	...	...	...
^{236}U [e]	15.7 ± 0.3	9.8 ± 0.4	12–32	8.6 ± 0.3	6.7 ± 0.6	6–17
^{252}Cf [f]	15	(11)	10–30	...	...	...
^{252}Cf [g]	16.0 ± 0.2	(10.2 ± 0.4)	8.3–37.7	8.0 ± 0.3	(6.2 ± 0.6)	6.5–24.3
^{252}Cf [h]	16 ± 0.5	11.5 ± 0.5	7.8–34.8	8 ± 1	6 ± 1	3.9–23.1
^{234}U [i]	15.6	9.4	12.8–26.7	7.0	3	5.3–11.1
^{240}Pu [j]	16.0 ± 0.1	10.6 ± 0.2	10–29	8.2 ± 0.2	7.6 ± 0.4	5.5–20

[a] MPE means most probable energy.

[b] FWHM means full width at half maximum. Parentheses indicate measurement of half width only.

[c] Present work.

[d] Reference 28.

[e] Reference 29.

[f] Reference 31.

[g] Reference 10.

[h] Reference 9.

[i] Reference 11.

[j] Reference 32.

reactions are lacking, and it is therefore not possible to rule out these reactions as sources for the short-range ^3H observed.

2. *Heavy-Ion Reactions*

Compound-nucleus formation involving binary-fission fragments and nuclei such as O or N, with subsequent evaporation of α particles, must be considered as a potential source for short-range ^4He.

Assuming the fission source to consist entirely of U_3O_8 (the presence of appreciable amounts of C or N in the source will not affect the following argument), and knowing the flux of binary-fission fragments, the reaction cross section required to produce the observed ^4He is calculated to be 75 ± 15 b. Typical total reaction cross sections for 100- and 150-MeV ^{12}C and ^{16}O projectiles, respectively, on targets in the mass region spanned by the fission fragments are ~2 b.[30] The most prominent mode of deexcitation in such reactions is multiple neutron evaporation. Cross sections for reactions leading to α emission are normally a small fraction of the total; for example, ~200 mb for α emission from the system $^{140}Ce + ^{16}O$ (90 MeV).[30] For the slightly lower average energies of binary-fission fragments, still lower cross sections would be expected. Clearly, heavy-ion reactions induced by fission fragments can account for at most 0.4% of the observed ^4He, as a high upper limit.

3. *Scattering of Atmospheric Helium*

Atmospheric He atoms in the space between the fission source and the first Pb foil may be scattered by fission fragments and become embedded in the first foil. To determine the magnitude of this contribution to the observed short-range ^4He an analysis based on Rutherford scattering and trapping probabilities of low-energy He atoms in Pb was carried out.[22] It was found that this mechanism can account for <0.1% of ^4He observed in the first foil.

Supporting evidence for the absence of appreciable contributions from scattering events is derived from an independent experimental study of Kr and Xe fission yields of ^{252}Cf carried out in this laboratory. An Al catcher foil separated by at least 2 mm of air from a ^{252}Cf fission source contained no atmospheric Kr and Xe attributable to scattering events. Assuming the observed short-range ^4He particles to be due to atmospheric He scattered by fission fragments into the catcher foils, and assuming this same process to be at work in the ^{252}Cf experiment, the abundance of Kr seen in this latter experiment should have been larger by a factor of >80, and would have easily been detected. Based on the above considerations it is considered highly unlikely that a measurable amount of short-range ^4He extracted from the Pb foils adjacent to the ^{235}U source was due to scattered atmospheric He.

4. *Fission*

The present experiments indicate that the observed short-range ^4He particles are produced in the fission act itself (short-range ^3H or ^3He may also be produced, but the evidence is less certain). It has been established that these particles are emitted with an energy $E \leq 7.7$ MeV and with a frequency of one particle in 478 ± 24 fissions. Other experiments (see Introduction) have indicated that short-range particles are produced with a frequency of about one in every hundred fissions of $^{236}U^*$, although no positive identification of the emitted light isotopes was obtained in those studies. The energy of the particles was estimated to be about 1 MeV, and emission was observed to be predominantly at right angles to the direction of the heavy fragments.[3]

It may be assumed now that at least some of the events seen in the early experiments were short-range ^4He particles emitted at the instant of scission. If we accept the results of measurements of energy and angle of emission obtained using nuclear emulsions,[3] it must be concluded that these short-range ^4He particles, just as the long-range ^4He particles, are released in the region between two heavy fragments. In order to escape this region of relatively high Coulomb potential energy with as little as 1 MeV of kinetic energy, the particles must be released only in cases of extreme deformation of the heavy fragments, and at a very late stage of the scission act, such that unusually large distances exist between the centers of the heavy fragments and the α particle. It is hoped that theoretical investigations into the various modes of deformation occurring during fission may lead to an understanding of this phenomenon.

B. Energy Distribution of the Long-Range Particles

Measurements of the energy distributions of ^3H and ^4He produced in fission were originally intended to provide information on the short-range components discussed above. Apart from revealing the existence of these low-energy particles, however, the energy-distribution measurements are the first to be carried out using the mass-spectrometric technique. The results obtained give independent confirmation of earlier, entirely different experiments. For comparison, Table II summarizes the present and other recently published data for ^{3}H and ^{4}He emitted in $^{236}U^*$, ^{252}Cf, $^{234}U^*$, and $^{240}Pu^*$ fission.

The values obtained in this work for most probable energies and widths of distributions are in excellent agreement with the results of other work. All other experiments[9–11, 28, 29, 31, 32] referred to in Table II employed counter telescopes and were subject to a low-energy cutoff. As already indicated, the present technique allows detection of particles with essentially zero energy, although with poor energy resolution. Similar experiments using thinner foils and higher-sensitivity mass-spectrometric measurements should reveal much more detail than the present work.

C. Probability of Emission of Long-Range ^{4}He Particles in Fission of $^{236}U^*$

The absolute frequency of formation of long-range α particles has been measured by a variety of techniques, but results obtained are in generally poor agreement. Table III summarizes some of the published data. It can be seen that the values vary by more than a factor of 2.5.

A possible explanation for the variation of quoted values may be the inclusion of short-range ^{4}He particles in some experiments and not in others. It should be noted that the nuclear-emulsion experiments generally give higher frequencies, whereas solid-state detectors give the lowest observed frequencies. The former experiments can detect particles with very low energies, whereas the latter would exclude all ^{4}He particles below ~6 MeV. In the present work a total (short-range plus long-range) ^{4}He emission frequency of 1 : 234 ± 7 was obtained. All values listed in Table III, except the two obtained using solid-state detectors, lie between the limits 1 : 234 ± 7 and 1 : 459 ± 22, representing, respectively, total ^{4}He and long-range ^{4}He only, measured in this work.

The mass-spectrometric technique used in this study is considered more reliable than others. This method eliminates spurious events by achieving unambiguous particle identification and attains necessary resolution from short-range particles. It is evident that further accurate measurements of emission probabilities for light charged particles in fission will be required before any reliable conclusions can be drawn from the variation of this parameter with different fissioning systems.

TABLE III. Probability of emission of long-range ^{4}He particles in thermal-neutron fission of ^{235}U.

Reference	Experimental technique	No. atoms ^{4}He to No. fissions
This work	Mass spectrometry	1 : 459 ± 22
12	Nuclear emulsions	1 : 230 ± 26
3	Nuclear emulsions	1 : 340 ± 40
5 [a]	Nuclear emulsions	1 : 401 ± 50
13	Nuclear emulsions	1 : 333 ± 111
14	Ionization chambers in coincidence	1 : 505 ± 50
15	Ionization chamber	1 : 220 ± 33
16	Ionization chambers in coincidence	1 : 250
17	CsI scintillator	1 : 449 ± 30
18	CsI scintillator with magnetic spectrograph	1 : 310
19 [b]	Solid-state detectors	1 : 507 ± 10
20 [a]	Solid-state detectors	1 : 594 ± 65

[a] Values shown are corrected values given in Table V of T. D. Thomas and S. L. Whetstone, Phys. Rev. 144, 1060 (1966).

[b] The value shown is an average of three values given in Ref. 19.

D. Relative Yields of Long-Range ^{3}H, ^{3}He, and ^{4}He in Fission of $^{236}U^*$

Relative yields of the various light particles emitted in ternary fission may ultimately provide a sensitive test for any theory attempting to explain this process. According to Halpern,[8] it does not seem possible to explain the relative yields on the basis of conventional evaporation theory. Such an approach could not account for the observed angular distribution (one would expect isotropic emission), and would predict unreasonably high neutron/(light charged-particle) ratios. Generally, one expects an inverse correlation between particle yield and required release energy, but lack of sufficient and accurate experimental data has not yet permitted determination of the exact functional dependence.

TABLE IV. Relative yields for long-range ^{3}H, ^{3}He, and ^{4}He particles.

Fissioning nuclide	$^{3}H/^{4}He$	$^{3}He/^{4}He$
$^{236}U^*$ [a]	$(3.1 \pm 0.8) \times 10^{-2}$	$(2^{+6}_{-2}) \times 10^{-6}$
$^{236}U^*$ [b]	$(6.2 \pm 0.5) \times 10^{-2}$	...
$^{234}U^*$ [c]	$(2.8) \times 10^{-2}$	$\simeq 1.8 \times 10^{-2}$
$^{240}Pu^*$ [d]	$(6.8 \pm 0.3) \times 10^{-2}$	...
^{252}Cf [e]	$(5.9 \pm 0.2) \times 10^{-2}$	$\lesssim 9 \times 10^{-3}$
^{252}Cf [f]	$(8.46 \pm 0.28) \times 10^{-2}$	$\leq 7.5 \times 10^{-4}$

[a] This work.
[b] Reference 29.
[c] Reference 11.
[d] Reference 32.
[e] Reference 9.
[f] Reference 10.

In Table IV some recent published relative yields for ^3H, ^3He, and ^4He are listed for several different fissioning species. A discrepancy exists between the value for $^3H/^4He$ found in this work and that of Dakowski *et al.*[29] If the results of the present work are accepted, however, we conclude that the $^3H/^4He$ ratios for $^{236}U^*$ and $^{234}U^*$ are roughly equal but differ significantly from those found for $^{240}Pu^*$ and ^{252}Cf fission.

Cambiaghi, Fossati, and Pinelli[11] claim positive observation of ^3He emitted in fission of $^{234}U^*$. Their measured yield is in sharp contrast to the value obtained in this work for $^{236}U^*$, as well as to the upper limits reported for ^{252}Cf. Cambiaghi, Fossati, and Pinelli consider the differences between their values and those for ^{252}Cf significant.

The extremely low (possibly zero) yield for ^3He obtained in this work for $^{236}U^*$ should provide data against which experiments using counter telescopes for particle identification may test particle resolution. If the large differences for $^3He/^4He$ ratios in various fission systems are verified in future experiments, they may provide a very sensitive test for any model of the ternary-fission process.

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