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Long-Range Particles from Nuclear Fission

Ralph A. Nobles

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LONG-RANGE PARTICLES FROM NUCLEAR FISSION*

By

Ralph A. Nobles

A Dissertation

Submitted in Partial Fulfillment of the

Requirements for the Degree of

Doctor of Philosophy

The University of New Mexico

1961

*Work performed under the auspices of the U. S. Atomic Energy Commission.

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This dissertation, directed and approved by the candidate's committee, has been accepted by the Graduate Committee of the University of New Mexico in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

Stewart A. Parkhurst
DEAN

October 2, 1961
DATE

Committee

A. P. Lemmitt
CHAIRMAN

Victor H. Rogers

Roy Thomas

John R. Green

This dissertation, directed and approved by the candidate's committee, has been accepted by the Graduate Committee of the University of New Mexico in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

CANDIDATE

ADVISOR

Committee

C. F. Smith

John H. Johnson

Roy Johnson

John H. Johnson

PREFACE

The research represented by this dissertation was performed under the Graduate Thesis Program of the Los Alamos Scientific Laboratory, Los Alamos, New Mexico. Under this program an agreement is made between the Los Alamos Scientific Laboratory and the University of New Mexico by which a graduate student, after completing his course work and preliminary requirements at the University, may perform the research for his doctoral degree at Los Alamos.

Ralph A. Nobles

Los Alamos, New Mexico

August 1961

The research presented in this dissertation was performed under

the guidance and supervision of the late Professor [Name]

of the University of [Location]. It is a pleasure to acknowledge

the assistance and cooperation of the [Institution] and the [Department]

in the completion of this work. The author also wishes to express

his appreciation to the [Institution] for the research facilities

provided during the course of this study.

Respectfully,
[Signature]

[Name]
[Address]
[City, State, Zip]

Los Angeles, New Mexico

August 1961

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Ralph A. Nobles

August 10, 1961

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work. The preparation of this dissertation was greatly facilitated by

the assistance of Dr. F. J. Brown and Dr. W. G. Brown in preparing and giving

the manuscript.

Alton A. Hobbs

August 10, 1951

ABSTRACT

This work is concerned with experimental observations of the long-range particles which are occasionally emitted by fissioning nuclei. These particles are known to be principally alpha particles and are of interest because they are associated with the configuration of the nucleus at the crucial time of scission. Observations were made of the energy spectra of long-range particles by means of a thallium-activated multiple CsI crystal scintillator device. The fluorescent response of the crystals employed for long-range particle detection was calibrated with natural radioactive alpha particles and alpha particles of known energy from the Los Alamos variable energy cyclotron. The energy spectra observed for spontaneous fission of Cf^{252} and thermal-neutron-induced fission of U^{235} were identical within the accuracy of the measurements, showing slightly askew bell-shaped distributions with maxima at 17 ± 1 Mev and endpoint energies of 29 ± 1 Mev. The distribution for Cf^{252} was determined with greater precision than that for U^{235} and showed structure on the low-energy side of the maximum in the region of 8-9 Mev. This structure is probably attributable to the triton group recently reported by Watson⁽¹⁾ and by Wegner.⁽²⁾

Measurements of the number of fissions per long-range particle were made with a multiple-ionization chamber apparatus by taking ratios of the number of twofold coincidences from fission fragments to the number of threefold coincidences involving a third particle of greater range. The

following results were obtained for spontaneous fission: Cf^{252} , 299.2 ± 18 ; Cm^{242} , 256.6 ± 17 ; Cm^{244} , 313.7 ± 20 ; Pu^{240} , 314 ± 20 ; and Pu^{242} , 264.7 ± 29 fissions per long-range particle. Results for thermal-neutron-induced fission were: Pu^{240} , 411.2 ± 26 ; Pu^{242} , 440.1 ± 28 ; U^{236} , 449 ± 30 ; and U^{234} , 414.1 ± 26 fissions per long-range particle. Results for fission induced by 1-Mev neutrons were: Pu^{240} , 403.1 ± 25 ; U^{236} , 534 ± 35 ; and U^{235} , 465.5 ± 30 fissions per long-range particle. The ratio of the probability of long-range particle emission from thermal-neutron-induced fission to that for spontaneous fission was 0.765 ± 0.086 for Pu^{240} and 0.829 ± 0.084 for Pu^{242} . In both cases the ≈ 6 Mev of excitation energy from thermal neutron capture and the higher spins of the compound nuclei caused a decreased probability of long-range particle emission. In general, the probability of emission was observed to decrease with decreasing fissionability parameter X and, also, to decrease with increasing spin and excitation energy for a given nuclei.

The simplified scheme of tripartition proposed by Tsien⁽³⁾ gives a qualitative explanation of the shape of the long-range particle energy spectrum and predicts the end-point energy rather well.

Several possible theories for the production mechanism for long-range particles from fission are discussed qualitatively. The conclusion reached is that at the present time there is no adequate theoretical explanation for the phenomena.

Techniques for preparing the thin nickel backings used for supporting the fission sources used in this work are described in the Appendix.

The most interesting technique reported is that for selectively removing the copper backing from the support foil only in the region of the source deposit.

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STATE OF NEW YORK

OFFICE OF THE ATTORNEY GENERAL

ALBANY, N. Y.

SEPTEMBER 10, 1908

TO THE HONORABLE THE SENATE

AND THE HONORABLE THE ASSEMBLY

OF THE STATE OF NEW YORK

I, THE ATTORNEY GENERAL, HEREBY SUBMIT TO YOU

THE REPORT OF THE COMMISSIONERS OF THE LAND OFFICE
RELATIVE TO THE PROPOSED SALE OF THE STATE LANDS
AND THE PROPOSED SALE OF THE STATE LANDS
AND THE PROPOSED SALE OF THE STATE LANDS

IN RESPONSE TO A RESOLUTION PASSED BY THE SENATE

ON APRIL 15, 1908, AND BY THE ASSEMBLY

ON APRIL 15, 1908, AND BY THE ASSEMBLY

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ON APRIL 15, 1908, AND BY THE ASSEMBLY

IV. REPORT OF THE COMMISSIONERS OF THE LAND OFFICE

RELATIVE TO THE PROPOSED SALE OF THE STATE LANDS

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INTRODUCTION

The possibility that nuclear fission occurs with division into three fragments of comparable mass was first proposed by Bohr and Wheeler⁽⁴⁾ and by Present and Knipp.^(5,6) Since then experiments have indicated the existence of complex fission of the following general types: (a) fission into three or more fragments of comparable mass,^(3,7,8,9) (b) fission into two heavy fragments and one light fragment of long range,⁽¹⁰⁻¹⁵⁾ and (c) fission into two heavy fragments and one light fragment of short range.^(11,13,16,17,18)

This present work is concerned with some aspects of fission of type (b) in which a long-range light particle is emitted.

The angular and energy distribution of these long-range particles is consistent with the hypothesis that they are produced in the region of the neck at the moment of scission and are then accelerated in the Coulomb field of the massive fission fragments.^(3,12,19) Since these particles are produced rather late in the fission process (i.e., at the moment of scission), a detailed study of this phenomenon may yield information of qualitatively different type from that obtained from fission widths and other phenomena more associated with barrier and saddle shape considerations.

With this motivation in mind a study was made of probability of emission of long-range particles, that is, of the ratio T/B , where T refers to the number of ternary fission events in which a third particle of long range is emitted, and B refers to the number of binary fission

events occurring in the same time interval. This ratio was determined for a number of nuclides undergoing spontaneous or neutron-induced fission. In some cases it was possible to observe the ratio for a nuclide fissioning spontaneously and after formation by slow neutron capture. These ratios provided information on the variation of T/B with excitation energy. Observation was made of the energy distribution of the long-range particles emitted in spontaneous fission of Cf^{252} and from thermal-neutron-induced fission of U^{235} . Knowledge of these energy distributions, of interest in their own right, was necessary for the correction of the T/B ratio data, which was taken with multiple ionization chamber apparatus in which the low-energy end of the distributions was not observed.

Since source preparation was a crucial part of the work, this subject is treated extensively in the appendices.

CHAPTER I

ENERGY DISTRIBUTION OF LONG-RANGE PARTICLES FROM FISSION

Apparatus and Experimental Procedures

A CsI crystal scintillation apparatus was used to study the energy spectrum of the long-range particles from fission. This device, illustrated in Figs. 1, 2, and 3, consisted of a fission source supported on a $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil, symmetrically positioned with respect to two sets of diametrically opposed thallium-activated CsI crystals, which were viewed by photomultiplier tubes through MgO_2 -surfaced vacuum light pipes. Two of the crystals, "b" and "d" in Fig. 1, were used as long-range particle detectors. These crystals were 2 cm in diameter and 0.41 mm in thickness; this thickness was sufficient to stop 30-Mev alpha particles at normal incidence. Crystals "a" and "c" were of the same diameter as "b" and "d" and were used in coincidence as fission detectors. Their 0.076-mm thickness was a compromise between undesirable sensitivity to gamma radiation and mechanical strength. These fission detectors were covered with $100\text{-}\mu\text{g}/\text{cm}^2$ nickel light shields to prevent "crosstalk" with the long-range particle crystals.

The mode of operation of the apparatus was as follows. A twofold coincidence between pulses from the fission detectors established the occurrence of a fission event. A threefold coincidence with the fission pulses and a third pulse from either of the long-range particle detectors

Fig. 1.--Cross-section view of the CsI crystal scintillation apparatus used in the measurements of the energy spectrum of long-range particles.

Fig. 2.--CsI scintillation detector apparatus. View shows the front cover plate removed.

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Fig. 3.--Overall view of the CsI scintillation detector apparatus.

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opened a gate on a 400-channel pulse-height analyzer which analyzed the third pulse and addressed the information to one 200-channel memory bank of the analyzer.⁽²⁰⁾ In an analogous manner pulses were fed into a second coincidence circuit at random time with respect to the fission pulses by a pulser whose repetition rate was set the same as the average fission coincidence pulse rate. Pulses in the long-range particle channels in coincidence with these random pulses were analyzed and addressed to the second half of the memory; thus a background distribution was secured which was later subtracted from the pulse-height distribution stored in the first half of the memory. With this technique the electronic system was used to collect data and background information simultaneously.

Figure 4 shows a block diagram of the principal features of the electronic setup. The Los Alamos Model 302 universal coincidence system contains two independent coincidence circuits and two input channels each having a pulse-height discriminator and a variable time-delay circuit. One-microsecond delay-line clippers were used on the Model 503 amplifiers⁽²¹⁾ in the fission channels to achieve an optimum compromise between maximum amplitude and minimum pulse duration. The series-parallel arrangement of Los Alamos Model 250 preamplifiers-amplifiers in the long-range particle channel allowed an independent amplitude adjustment to the discriminator and pulse-height analyzer inputs. Standard electronic high-voltage power supplies (not shown) were used for the photomultipliers. The long-range particle channels used a common

Fig. 4.--Block diagram of the apparatus used in the long-range particle energy spectrum measurements.

amplifier system; their gains were normalized to each other by a differential adjustment of photomultiplier voltage. It was convenient to use the peaks from the radioactive alphas from the fission source for this normalization.

The energy resolution of the long-range particle counters was about 11% for 6.1-Mev Cr^{252} alphas and 6% for the 8.78-Mev alphas from Po^{212} .

Calibration of the Fluorescent Response of CsI Crystals

The fluorescent response of CsI crystals to alpha particles was determined by the use of radioactive alpha particles and alpha particles from the Los Alamos variable energy cyclotron. The cyclotron He^4 beam was scattered off a $500\text{-}\mu\text{g}/\text{cm}^2$ gold foil and into the long-range particle detector crystals. The energy of the beam was determined by a pulse-height reader apparatus.⁽²²⁾ Figure 5 shows the results of these fluorescent response (relative pulse height) versus alpha-particle energy measurements. The experimental points in the figure represent a composite of the data from both crystals. In some cases several runs were made at a given energy. The scatter of the points indicates the reproducibility of the results. These results are in excellent agreement with those reported by Quinton, Anderson, and Knox.⁽²³⁾

The justification for calibrating the crystals with alpha particles is as follows. Farwell et al.⁽¹²⁾ demonstrated that the majority of long-range particles in coincidence with fission have range and specific ionization characteristic of alpha particles. This has

applied across their grain were maintained at each other by means of
an external adjustment of potential difference. It was convenient to use
the probe from the radioactive source since the detection device for this
normalization.

The energy resolution of the long-range particle spectrometer was
about 1% for 0.1-MeV α particles and 5% for the 0.18-MeV alpha from
Po-210.

Determination of the Fluorescent Response of CdS Crystals

The fluorescent response of CdS crystals to alpha particles was
determined by the use of an isotropic alpha particle and alpha particle
from the low atomic number energy cyclotron. The cyclotron beam
was scattered off a 500- μ g/cm² gold foil and into the long-range particle
detector crystals. The energy of the beam was determined by a pulse-
height meter system. Figure 5 shows the results of these
fluorescent response (relative pulse height) versus alpha-particle
energy measurements. The experimental points in the figure represent a
composite of the data from both crystals. In some cases several runs
were made at a given energy. The scatter of the points indicates the
reproducibility of the results. These results are in excellent agree-

ment with those reported by Bateman, Anderson, and Kohn. (23)

The justification for calling the crystals with alpha
particles is as follows. Jewell et al. (12) demonstrated that the
activity of long-range particles is comparable with that of alpha
and specific interaction characteristics of alpha particles. This was

Fig. 5.--Fluorescent response of CsI crystals as a function of alpha-particle energy. (a) U^{235} alpha particles; (b) Cf^{252} alpha particles; (c) Po^{212} alpha particles; (d) alpha particles from cyclotron.

been corroborated by other studies, and Fulmer and Cohen⁽²⁴⁾ have, in addition, used a magnetic spectrograph to show that the particles are indeed alphas. Tsien⁽³⁾ concluded on theoretical grounds that the long-range particles can have mass-to-charge ratios of 2, 2.5, and 3, with the group corresponding to a mass-to-charge ratio of 2 being the most intense. Recently Watson⁽¹⁾ and Wegner⁽²⁾ have reported tritons from Cf²⁵² spontaneous fission occurring with a frequency of about six or seven tritons per 100 alphas, and other particles, if present, occurring with a considerably smaller probability.

Observation of Spontaneous Fission of Cf²⁵²

A Cf²⁵² source having an activity of approximately 3×10^4 fissions per minute was prepared by S. G. Thompson's group at the Lawrence Radiation Laboratory, Berkeley, by the technique of autodeposition from an intense parent source.⁽²⁵⁾ The deposit was supported on a wrinkle-free, mirror-finish 100- $\mu\text{g}/\text{cm}^2$ nickel foil,⁽²⁶⁾ reinforced by a 20-mesh, 93%-transparent, electromesh nickel screen.⁽²⁷⁾ Because of the high specific activity of this material the deposit was very thin, amounting to less than 10^{-3} μg of Cf²⁵² distributed on a $\approx 0.2\text{-cm}^2$ area.

The threefold coincidence data from this source gave a distribution of counts versus pulse height which was converted to counts-per-energy interval versus energy using the fluorescent response data of Fig. 5.

been observed by other investigators, and indeed, it is
 addition, used a magnetic spectrograph to show that the particles are
 indeed alpha particles. (26) concluded on theoretical grounds that the
 range of particles can have mass-to-charge ratios of 2, 3, 4, and 5, with
 the range corresponding to a particular ratio of 1 being the most
 intense. Recently, Nelson (27) and Wigner (28) have reported results
 of a systematic search covering with a frequency of about six per
 second and for 100 alpha and other particles. It means, however,
 with a considerably smaller probability.

Observation of Spontaneous Emission of α Rays

A Geiger counter having an activity of approximately 5×10^4 counts
 per minute was prepared by E. O. Thompson's group at the Lawrence
 Livermore Laboratory, by the technique of autoionization from an
 intense gamma source. (29) The deposit was supported on a window-free
 nickel-clad 100- μ thick nickel foil. (30) returned by a 30-mesh
 300-transparent, electrochemically nickel screen. (31) Because of the high
 specific activity of this material the deposit was very thin, amounting
 to less than 10^{-6} g of ^{226}Ra distributed on a 0.5-cm² area.
 The threefold coincidence data from this source gave a statistical
 error of about 1% in the pulse height which was converted to counts per
 energy interval versus energy using the Thomsen response data of

The results of this observation are shown in Fig. 6. It may be noted that the curve is not extrapolated below 1.5 Mev, this being the somewhat arbitrary but commonly accepted division point between long- and short-range particles from fission.⁽¹⁶⁾ No data points are given below 7.55 Mev because the data below this energy were taken in a separate run employing an expanded energy scale (i.e., higher gain) in order to observe the region in greater detail. These data were fitted to the main distribution at 7.5 Mev.

No significant data were obtained in the vicinity of 6.1-Mev energy because of the large statistical uncertainty of the results in this region. This statistical uncertainty was due to the high background of accidental threefold coincidence pulses caused by the radioactive alpha particles.

The uncertainties on the datum points in Fig. 6 are their standard deviations derived from the standard deviations of the total counts and background counts at each point. The background counts amounted to approximately 16% of the total in the 7.5- to 8.5-Mev region, about 0.5% at 11-12 Mev, approximately 9% at 16-18 Mev, and zero above 19 Mev.

Observation of Thermal-Neutron-Induced

Fission of U^{235}

The experimental setup used in this measurement was the same as that described in the previous section on Cf^{252} . The sample was an 864- μ g deposit of U^{235} distributed on a 2-cm² area. This deposit was

Fig. 6.--Energy distribution of long-range particles from spontaneous fission of Cf^{252} .

prepared by vacuum evaporation onto a non-reinforced $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil.

The scintillation counter apparatus was placed in an external neutron beam from the south thermal column of the Los Alamos Homogeneous Reactor.^(28,29) The principal difficulty in the measurement was caused by gamma-ray-induced backgrounds in the CsI crystals. This difficulty was reduced by lead shielding of the crystals and collimation of the neutron beam. The conditions under which the data were taken were such that the background amounted to about 64% of the total counts at 6 Mev, 16% of the total at 15 Mev, and zero above 16.5 Mev.

The pulse-height distribution obtained was converted to an energy distribution using the results of the previously described fluorescent response measurements. The resulting energy distribution is shown in Fig. 7. As in the previous case, the uncertainties on the datum points are their standard deviations derived from the standard deviations of the total counts and background counts at each point.

Results and Conclusions

For purposes of comparison Fig. 8 shows the distributions of Figs. 6 and 7 on a common plot. Ordinates of the curves are normalized so that the area under each is proportional to the probability of long-range particle emission per fission.

These results are in general agreement with work previously reported,^(14,15,24,30,31) although Muga et al.⁽³¹⁾ report an average

Fig. 2. Energy distribution of long-range electrons from
control and low-induced electron at 100 eV.

PHOTONIC ENERGY (WAT)

RELATIVE PROBABILITY OF OCCURRENCE PER ENERGY INTERVAL

Fig. 8.--Comparison of the energy distributions of long-range particles from spontaneous fission of Cf^{252} and thermal-neutron-induced fission of U^{235} . Curves are normalized so that their areas are proportional to the number of long-range particles emitted per fission.

energy of about 19 Mev and an end-point energy of about 34 Mev for particles from spontaneous fission of Cr^{252} .

In Fig. 8 it will be noted that the maxima lie at nearly the same energy, with that for U^{238} being possibly about 1 Mev lower. However, the accuracy of location of the peaks is such that no great reliance can be placed on this result. The similarity of the shape and of the end-point energies of the two curves strongly suggests that the same basic mechanism is involved in both cases. This conclusion is consistent with the hypothesis which has the third particle produced at scission in the region of the neck with low kinetic energy and the three fragments then separating under their mutual electrostatic repulsion. From this model, which invokes only gross characteristics of nuclear matter such as surface and Coulomb energies, one would not expect a strong dependence on A and Z.

In keeping with the above hypothesis Tsien⁽³⁾ has proposed a simplified scheme of tripartition which envisions the nuclear matter just after scission as having the configuration of three spheres in contact, as shown below.

Fig. 9.--Simplified scheme of tripartition. A. $0 < H < H_{\text{max}}$. B. $H = H_{\text{max}}$, limiting case of three spheres in mutual contact.

energy of about 10 Mev with an end-point energy of about 50 Mev for
 particles of a continuous spectrum of C^{12} .
 In Fig. 3 it will be noted that the maxima lie at nearly the same
 energy with that for C^{12} being possibly about 1 Mev lower. However,
 the tendency of location of the peaks is such that no great reliance can
 be placed on this result. The similarity of the shape and of the end-
 point energies of the two curves strongly suggests that the same factor
 is involved in both cases. This conclusion is consistent with
 the hypothesis which has the third particle produced at rest in the
 region of the rock with low kinetic energy and the three fragments then
 separating under their mutual electrostatic repulsion. From this model,
 which involves only gross characteristics of nuclear matter such as
 surface and Coulomb energies, one would not expect a strong dependence

on A and Z .
 In keeping with the above hypothesis, (2) has proposed a sim-
 plified scheme of a reaction which explains the nuclear matter
 after collision as having the contribution of three spheres in contact,
 as shown below.

Fig. 3--Simplified scheme of a reaction. $A, 0 < B < C$.
 (2) $B = \text{max}$, starting case of three spheres in mutual contact.

In Fig. 9, M_1 and M_2 are heavy fragments and M_3 the light or third fragment, and Z_1 , Z_2 , Z_3 , r_1 , r_2 , and r_3 are the corresponding charges and radii. H is the distance from the center of M_3 to the center line of M_1 and M_2 . Using the most probable values for M_1 , M_2 , Z_1 , and Z_2 for fission of U^{236} , and accepted values for nuclear radii, Tsien predicts that for $H = H_{\max}$ (i.e., limiting case B, Fig. 9) and for $M_3/Z_3 = 3$, M_3 would have a maximum energy of about 15 Mev, and for $M_3/Z_3 = 2$ a maximum of 25 to 29 Mev, depending on the particular values used for the nuclear radii. This latter value is in good agreement with the observed end-point energy. Particles for which $M_3/Z_3 = 1$ would have a maximum energy of 50 Mev or greater, and since no such particles are observed experimentally the conclusion from the theory is that protons are not emitted in tripartition fission.

A qualitative explanation of the observed shape of the energy distribution of M_3 particles is derived from the fact that for a given mass-to-charge ratio the energy decreases with decreasing H and also decreases with increasing r_1 and r_2 values. In Tsien's theory the effective nuclear radii are considered to increase with fragment excitation. The result of these effects is a continuous energy distribution.

This theory, based on the simplified scheme of tripartition, also provides an explanation for the observed average angle of emission of the third particle with respect to the direction of the heavy fragments.

In Figs. 6 and 8 the shoulder on the low-energy side of the Cf^{252} distribution is probably due to tritons. As reported by Watson⁽¹⁾ and

In Fig. 3, N and n are heavy fragments and n is the light fragment.
fragments, and N , n , N , n , N , n , and so on, are the fragments of the
and n . N is the distance from the center of N to the center of
of N and n . Using the most probable values for N , n , N , n , N , n ,
and n , and N , n , N , n , N , n , N , n , N , n , N , n , N , n , N , n ,
that for $N = 10$, $n = 1$, $N = 10$, $n = 1$, $N = 10$, $n = 1$, $N = 10$, $n = 1$,
would have a maximum energy of about 15 Mev, and for $N = 10$, $n = 1$,
of 25 to 35 Mev, depending on the particular value of N and n .
with. This factor value is in good agreement with the observed
point energy. Particles for which $N = 1$ would have a maximum
of 50 Mev or greater, and since no such particles are observed,
essentially the comparison with the theory is that particles are
in the region of the

A qualitative explanation of the observed energy of the particles
distribution of N particles is derived from the fact that the
that to change ratio the energy decreases with decreasing N and
decreases with increasing n and its values. In Table 1, the
the nuclear field are considered to increase with increasing N and
The results of these effects is a continuous energy distribution.
This theory, based on the simplified scheme of separation, also
provides an explanation for the observed average angle of emission of
the third particle with respect to the direction of the heavy fragment.
In Figs. 4 and 5 the number on the low-energy side of the
distribution is probably due to various, as reported by various

Wegner⁽²⁾ these particles have a distribution which peaks at 8.5 ± 1.0 Mev and an end-point at 16 ± 1 Mev, and are present with a frequency about 6% that of alpha particles. The dashed line curve in Fig. 6 shows a conjectured triton energy distribution.

In the case of U^{236} the accuracy of the data is not sufficient to show this structure in the energy distribution if it exists.

CHAPTER II

MEASUREMENT OF TERNARY-TO-BINARY FISSION RATIO

Apparatus and Experimental Procedures

Both nuclear emulsion and counter techniques have been used extensively in measuring ternary-to-binary fission ratios.⁽³²⁾ In the present work the use of counters was deemed advantageous, one reason being the relative ease with which they can accommodate highly alpha-active material. Most of the spontaneous fissionable elements have a high ratio of natural radioactive alpha emission to spontaneous fission; for instance, the ratio for Pu^{240} is 1.85×10^7 alphas per fission. If such a material were loaded in a nuclear emulsion the plate would be fogged by the radioactive alphas. The technique used by M. F. Netter et al.⁽³³⁾ of separating the active material from the emulsion by a window, thus stopping the natural alphas and fission fragments but not the long-range particles, has the disadvantages of a low-energy cutoff on the long-range particles and difficulty in determining the number of fissions occurring.

The ionization chamber device used here had the disadvantage of the low-energy cutoff mentioned above but was able to detect fission events directly by means of a fast twofold coincidence of the fission fragments.

The detector employed consisted of four parallel plate ionization chambers operating in an atmosphere of argon + 5% CO_2 gas mixture.⁽³⁴⁾ The arrangement of the ionization chambers is shown in Figs. 10 and 11.

Fig. 10.--Ionization chamber apparatus used in the measurement of the number of long-range particles per fission in spontaneous and thermal-neutron-induced fission.

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Fig. 11.--Electrode assembly of the ionization chamber apparatus used in spontaneous and thermal-neutron-induced fission measurements. View shows the pressure vessel removed. A centimeter rule is included for scale.

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In the following discussion the numbers refer to Figs. 10 and 11. Electrode 3 was at negative high potential and also carried the fission source which was deposited on a $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil (see Appendix A). Electrodes 2 and 4 were at ground potential and acted as electron collectors for the fission chambers. The electron transit time in these chambers was of the order of 3×10^{-7} sec, permitting the use of a $0.1\text{-}\mu\text{sec}$ coincidence resolution time. Electrodes 2 and 4 had $3.2\text{-mg}/\text{cm}^2$ aluminum windows, each subtending a solid angle of approximately π steradian at the source foil position. The filling gas pressure was adjusted so that the stopping power of the fission chamber plus that of the aluminum window was just sufficient to stop normally incident 6.11-Mev radioactive Cf^{252} alphas. Since these were the most energetic natural alphas encountered, this filling insured that no alphas or fission fragments originating at the source foil could enter the outer ionization chambers. (35,36)

Electrodes 1 and 5 were at positive high potential and acted as electron collectors for their respective chambers. Grids were employed in the outer chambers to insure that long-range particles which entered through the windows and were stopped in the chambers would give pulses whose heights were proportional to their energy loss and independent of their direction. (37) This eliminated one of the sources of instrumental pulse-height dispersion in the crucial energy range near the lower cutoff and simplified the task of correcting for the unobserved portion of the spectrum below the cutoff. Particles at the upper end of the long-range-

particle energy distribution spend only a small fraction of their range in the chambers and hence produce small pulses in spite of their high energy. To prevent counts being lost at the high end of the spectrum, the bias was never set higher than 1.5 Mev, which represents the energy loss of the maximum energy particles encountered. The minimum bias level was governed by amplifier noise and in some cases by gamma- and neutron-induced backgrounds. The lowest bias level used was 50 kev. The method of obtaining an energy calibration of the bias setting will be subsequently described.

Table 1 gives a tabulation of some of the dimensions and characteristics of the apparatus described above.

Dry-cell battery high-voltage supplies were used to insure long-term freedom from voltage transients. Figure 12 shows a schematic diagram of the coupling circuits in the base of the chamber. It should be noted that electrodes 2 and 4 served a dual role, that of signal collector plates for the fission chambers as well as of ground plates for the long-range particle chambers. Hence the value of the shunt resistor for these electrodes had to be judiciously chosen so as to permit the fission pulse to be picked up and at the same time not allow a large enough voltage swing to transmit the pulses to the signal plates of the outer chambers. It was experimentally determined that a 6.2-kohm shunt gave the greatest fission signal without feed-through in all cases except where pile-ups were encountered from very high fission rates, in which case a 1-kohm shunt was required.

TABLE 1

DIMENSIONS AND CHARACTERISTICS OF IONIZATION CHAMBER APPARATUS

Electrode material	Brass
Electrode diameter	17.8 cm
Gas filling	1880 mm Hg, Argon + 5% CO ₂
Fission chamber spacing	1.23 cm
Voltage, fission chamber	1688 volts
Diameter of aluminum windows	4.48 cm
Thickness of windows	3.2 mg/cm ² Aluminum
Spacing, grid to aluminum window	1.43 cm
Spacing, grid to collector	0.714 cm
Diameter of grid wire	0.127 mm
Grid spacing, center to center	1.59 mm
Grid material	Stainless steel
Grid support material	Stainless steel and Invar
Voltage, collector to ground	3308 volts
Voltage, grid to ground	1720 volts
Shielding of collector by grid ^a	95.5%

^aCalculated following the technique outlined by D. H. Wilkinson. (38)

CHARACTERISTICS OF LUTATION TRENCH

Location	1000 m. N. of N. 1000
Direction of flow	1000 m. N. of N. 1000
Depth	1000 m. N. of N. 1000
Width	1000 m. N. of N. 1000
Volume of water	1000 m. N. of N. 1000
Velocity	1000 m. N. of N. 1000
Temperature	1000 m. N. of N. 1000
Pressure	1000 m. N. of N. 1000
Humidity	1000 m. N. of N. 1000
Wind speed	1000 m. N. of N. 1000
Wind direction	1000 m. N. of N. 1000
Cloud cover	1000 m. N. of N. 1000
Relative humidity	1000 m. N. of N. 1000
Atmospheric pressure	1000 m. N. of N. 1000
Barometric pressure	1000 m. N. of N. 1000
Sea level pressure	1000 m. N. of N. 1000
Surface pressure	1000 m. N. of N. 1000
Ground pressure	1000 m. N. of N. 1000
Water pressure	1000 m. N. of N. 1000
Soil pressure	1000 m. N. of N. 1000
Atmospheric pressure	1000 m. N. of N. 1000
Barometric pressure	1000 m. N. of N. 1000
Sea level pressure	1000 m. N. of N. 1000
Surface pressure	1000 m. N. of N. 1000
Ground pressure	1000 m. N. of N. 1000
Water pressure	1000 m. N. of N. 1000
Soil pressure	1000 m. N. of N. 1000

Calculated following the technique outlined by D. H. Wilkinson (1961)

Fig. 12.--Schematic diagram of the electrode connections and coupling circuits of the ionization chamber apparatus.

IONIZATION CHAMBER

Fig. 13.--Block diagram of the electronic apparatus used in the measurements of the number of long-range particles per fission in spontaneous and thermal-neutron-induced fission.

singles rate in the long-range particle channels. The observed three-fold accidental coincidence rate was about 22% less than the rate calculated from the above equation; this is probably because the effective resolution time of the coincidence unit used was less than that to be expected from the nominal pulse widths involved.

The energy calibration of the lower bias level of the long-range particle chambers was accomplished by placing a number of natural radioactive alpha sources on the aluminum window electrode in the chamber to be calibrated and observing the pulse-height spectrum at the output of the 101A amplifiers with a 100-channel pulse-height analyzer. From the known energy of the alpha particles a plot was then made of energy loss in chamber versus calibration pulser voltage. This plot allowed the pulser to be used to set the bias level at any desired energy equivalent. Figure 14 shows a typical pulse-height spectrum and also the corresponding pulser calibration. A Los Alamos Model 500 pulser was used throughout this work.

The alpha sources were thin oxide deposits on 0.1-mm-thick platinum backings. Two Pu^{239} sources were used, one bare and the other covered with a 3.2-mg/cm^2 aluminum absorber, which, according to the Berkeley range-energy curves, ⁽³⁹⁾ gave alpha particles with a remaining energy of about 2.74 Mev. Figure 15 shows energy versus pulser voltage as derived from Fig. 14. Similar calibrations were obtained for both chambers and for each set of experimental conditions employed.

Fig. 14.--Typical pulse-height spectrum and corresponding pulser calibration.

Fig. 15.--Long-range particle chamber bias energy versus pulser voltage.

The calculated maximum electron transit time⁽⁴⁰⁾ in the outer chambers was about 0.5 μ sec. This transit time, together with the wide range of directions and path lengths possible for the long-range particles, caused a time jitter of the order of 0.5 μ sec between the pulses from these chambers and the fission pulses. To allow for this jitter plus a safety factor the threefold coincidence resolving time was set at 1.6 μ sec.

In order to improve the stability of operation the optional rise-time delay circuit was used in the discriminator of the long-range particle channel of the 302-II coincidence unit. This circuit differentiates the leading edge of the input pulse and is set to trigger at the peak rather than at the beginning of the pulse. This rise-time delay circuit, together with the use of the slower 101B amplifiers, introduced a delay in these channels which necessitated the use of an appropriate electronic delay in the fission channels to achieve a threefold coincidence.

Figure 16 shows a typical plot of the triple-to-double coincidence ratio as a function of the electronic delay time in the fission channels. Similar curves were obtained for each different set of experimental conditions; this insured the choice of the optimum delay time in each case.

The fission sources used in this work were prepared by the techniques of electrospraying,⁽⁴¹⁾ vacuum evaporation, and painting, except for Cf^{252} which, as previously noted, was prepared by autodeposition

Fig. 16.--Triple-to-double coincidence ratio as a function of fission channel delay time.

from an intense parent source. Deposits were made on 100- $\mu\text{g}/\text{cm}^2$ nickel foil⁽²⁶⁾ which in some cases was reinforced by a backing of 20-mesh, 93%-transparent, electromesh nickel screen.⁽²⁷⁾ It was necessary to cover some of the highly alpha-active sources with 100- $\mu\text{g}/\text{cm}^2$ nickel foil to prevent the radioactive material from spreading throughout the chamber. More details on the techniques of foil preparation are given in Appendix A.

Data-Correction Procedure

After subtractions are made for accidentals the ratio, which was observed experimentally with the apparatus described, was that of triple-to-double coincidences. In order to yield the true ratio of ternary-to-binary fission events the data must be corrected for geometry, for self-absorption in the source, and for the unobserved low-energy portion of the spectrum.

The following equation gives the form of these corrections:

$$\frac{T}{B} = \frac{N_3 \left[\kappa_1 \tau_1 \frac{\epsilon \Omega_1}{\epsilon \Omega_1 + \Omega_2} + \kappa_2 \tau_2 \frac{\epsilon \Omega_2}{\epsilon \Omega_1 + \Omega_2} \right]}{N_2}, \quad (2)$$

where T/B is the ternary-to-binary fission ratio, τ_1 , τ_2 are the factors which correct for the low-energy cutoff in the spectrum of long-range particles, τ_1 referring to the chamber which has electrode 1 for its collector and τ_2 to the chamber whose collector is electrode 5 (see Fig. 10, 11, or 12), κ_1 , κ_2 are the corresponding factors for each

chamber which correct for the solid angle and for fission fragment absorption in the source, ϵ is the correction for absorption of long-range particles in nickel screen backing when used, Ω_1 is the solid angle subtended by the aluminum window in electrode 2 at source position, Ω_2 is the solid angle subtended by the aluminum window in electrode 4 at source position, N_2 is the binary count for the observation period corrected for background, and N_3 is the long-range particle or ternary count for the same observation period corrected for background.

The energy spectrum cutoff correction factors τ_1 and τ_2 were determined in the following manner. The average cutoff energy was established for each chamber. This energy was not the same for both because of the slightly different depths of the two fission chambers. It was assumed that the particles involved had alpha-particle range-energy characteristics and that the fission sample constituted a point source. The assumption of alpha-particle range-energy characteristics is justified by the discussion in Chapter I (page 9), and the second assumption was justified by calculations of the effect of an extended source of the size of the largest used in this work (i.e., ≈ 1.5 -cm diameter). These calculations indicated that only a second-order error was introduced by the point source assumption. By the use of the above assumptions and the known stopping power of the filling gas and aluminum windows, a numerical integration was performed to determine the minimum average energy a particle required to reach the long-range particle chamber with a remaining energy equal to the bias energy. For a 50-kev

The results obtained for the cold and for the warm regions are shown in the figures 1 and 2. The curves for the cold region are obtained by the method described in the text. The curves for the warm region are obtained by the method described in the text. The results for the cold region are shown in the figures 1 and 2. The results for the warm region are shown in the figures 3 and 4. The results for the cold region are shown in the figures 5 and 6. The results for the warm region are shown in the figures 7 and 8. The results for the cold region are shown in the figures 9 and 10. The results for the warm region are shown in the figures 11 and 12. The results for the cold region are shown in the figures 13 and 14. The results for the warm region are shown in the figures 15 and 16. The results for the cold region are shown in the figures 17 and 18. The results for the warm region are shown in the figures 19 and 20. The results for the cold region are shown in the figures 21 and 22. The results for the warm region are shown in the figures 23 and 24. The results for the cold region are shown in the figures 25 and 26. The results for the warm region are shown in the figures 27 and 28. The results for the cold region are shown in the figures 29 and 30. The results for the warm region are shown in the figures 31 and 32. The results for the cold region are shown in the figures 33 and 34. The results for the warm region are shown in the figures 35 and 36. The results for the cold region are shown in the figures 37 and 38. The results for the warm region are shown in the figures 39 and 40. The results for the cold region are shown in the figures 41 and 42. The results for the warm region are shown in the figures 43 and 44. The results for the cold region are shown in the figures 45 and 46. The results for the warm region are shown in the figures 47 and 48. The results for the cold region are shown in the figures 49 and 50. The results for the warm region are shown in the figures 51 and 52. The results for the cold region are shown in the figures 53 and 54. The results for the warm region are shown in the figures 55 and 56. The results for the cold region are shown in the figures 57 and 58. The results for the warm region are shown in the figures 59 and 60. The results for the cold region are shown in the figures 61 and 62. The results for the warm region are shown in the figures 63 and 64. The results for the cold region are shown in the figures 65 and 66. The results for the warm region are shown in the figures 67 and 68. The results for the cold region are shown in the figures 69 and 70. The results for the warm region are shown in the figures 71 and 72. The results for the cold region are shown in the figures 73 and 74. The results for the warm region are shown in the figures 75 and 76. The results for the cold region are shown in the figures 77 and 78. The results for the warm region are shown in the figures 79 and 80. The results for the cold region are shown in the figures 81 and 82. The results for the warm region are shown in the figures 83 and 84. The results for the cold region are shown in the figures 85 and 86. The results for the warm region are shown in the figures 87 and 88. The results for the cold region are shown in the figures 89 and 90. The results for the warm region are shown in the figures 91 and 92. The results for the cold region are shown in the figures 93 and 94. The results for the warm region are shown in the figures 95 and 96. The results for the cold region are shown in the figures 97 and 98. The results for the warm region are shown in the figures 99 and 100.

bias this energy was 7.90 Mev for the chamber having electrode 1 as its collector and 7.63 Mev for the chamber having electrode 5 as its collector. In the case of a 1.0-Mev bias these cutoff energies were 8.23 Mev and 7.97 Mev, respectively.

The energy distribution obtained in Chapter I was then used to derive values for τ_1 and τ_2 , the total area under the curve in Fig. 6 being compared to the area under the curve and above the appropriate average cutoff energy. For instance, the τ_1 corresponding to 1-Mev bias is the ratio of the entire area under the curve to the area under the curve above 8.10 Mev.

Table 2 lists the values obtained for τ_1 and τ_2 .

TABLE 2
VALUES FOR LOW-ENERGY CUTOFF CORRECTION FACTORS

Bias	Lower Chamber		Upper Chamber	
	Average Cutoff Energy (Mev)	Correction Factor (τ_1)	Average Cutoff Energy (Mev)	Correction Factor (τ_2)
50.0 kev	7.90	1.1074	7.63	1.0985
0.5 Mev	8.08	1.1181	7.82	1.1077
1.0 Mev	8.23	1.125	7.97	1.113
1.25 Mev	8.30	1.1260	8.04	1.115

It should be noted that Fig. 6, the Cr^{252} energy distribution curve, was used in deriving the above corrections. The curve for U^{236} ,

This data energy was 0.50 MeV for the chamber having electrode 1 as anode
 and electrode 2 as cathode. The chamber having electrode 2 as anode and
 electrode 1 as cathode had a 1.0-MeV data energy. The data energy was 0.50 MeV
 and 1.0 MeV, respectively.
 The average distribution obtained in Chapter 1 was compared to
 the values for 1 and 2, the total area under the curve in Fig. 2
 being compared to the area under the curve and above the background
 average curve. The results, the 11 corresponding to 1.0 MeV data
 is the ratio of the curve area under the curve to the area under the
 curve above 0.10 MeV.
 Table 2 gives the values obtained for 1 and 2.

TABLE 2
 VALUES FOR LOW-ENERGY CORRECTION FACTORS

Data Energy (MeV)	Lower Chamber		Upper Chamber	
	Average Count Rate	Correction Factor	Average Count Rate	Correction Factor
0.50 MeV	7.50	1.104	7.50	1.104
0.5 MeV	8.00	1.113	8.00	1.113
1.0 MeV	8.25	1.125	8.25	1.125
1.0 MeV	8.30	1.126	8.30	1.126

It should be noted that the 0.5 MeV data energy distribution

was well in agreement with the average distribution. The ratio for 1

Fig. 7, appears to be essentially the same as that for Cf^{252} except for being less precisely determined. Furthermore, if one assumes that the third particle in fission is generated by a mechanism like that proposed by T. S. Tsien,⁽³⁾ one would not expect the energy distribution to change markedly from nuclide to nuclide. Hence it is expected that the correction factors τ_1 , τ_2 derived for Cf^{252} can be used to correct the data from other nuclides as well.

The factor ϵ , necessary only for those cases in which a nickel screen reinforcing was employed, was determined by counting the number of long-range particles emitted from each side of the source. After correction was made for the slightly differing counting geometries, the remaining difference was attributed to absorption in the support screen and the $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil. However, the average range in nickel of the particles involved is greater than $42\text{ mg}/\text{cm}^2$ and the nickel foil had a thickness of only $\approx 0.1\text{ mg}/\text{cm}^2$; hence almost all the absorption was in the screen itself. The corrected ratio of the counting rates from the two sides of the source was 0.935 ± 0.017 . This means that about 6.5% of the particles emitted on the screen side were absorbed in the screen, a figure which agrees very well with its nominal transparency of 93%.

For the sake of brevity the background corrections on N_2 and N_3 will not be listed explicitly in the measurements described in this chapter; however, operating conditions were such that the correction on N_2 was usually less than 1% of the total count, and on N_3 usually less than 5% of the total count.

the 1. appears to be constant in the same as the
being less precisely determined. Furthermore, the
first particle in this is generated by a constant
of T. R. T. (5) and would not depend on the
initially this particle is emitted. Hence in an
that follows, it is derived for $\alpha = 0.525$ that
from other results as well.

The factor α necessary only for those cases in which
values relating to the system, are determined by the
of long-range particles emitted from each side of the source, and
position was made for the slightly different values of α .
assuming difference was determined by absorption in the medium, and
and the long-range particle itself. However, the value of α is
the particle itself is greater than at any other value of α .
a thickness of only ~ 0.1 cm, hence since the thickness of the
the system itself. The corrected ratio of the particles from the
two sides of the source was 0.525 $\pm$ 0.011. This is the same as
of the particles emitted on the other side with thickness 0.1 cm.
a figure which agrees very well with the calculated value of 0.525.
for the case of heavy the background experiment was made as
will not be listed explicitly in the same manner. Table 1 is
however, operating conditions were such that the ratio of
is usually less than 1/2 of the total count, and in the present case
than 1/2 of the total count.

Spontaneous Fission Measurements

Spontaneous Fission of Cr^{252}

The source used here was the same Cr^{252} source described in Chapter I. This source, as previously noted, was very thin; nevertheless, it was sufficiently active to provide adequate counting statistics in a relatively short time, hence was used throughout the series of experiments for normalization and for checking the performance of the apparatus.

The results obtained for Cr^{252} at 1.25-Mev bias are as follows:

$$\epsilon = 0.935 \pm 0.017$$

$$\tau_1 = 1.126 \pm 0.056$$

$$\tau_2 = 1.115 \pm 0.056$$

$$\Omega_1 = 1.0029 \pm 0.0125 \text{ rad}$$

$$\Omega_2 = 1.0746 \pm 0.0133 \text{ rad}$$

$$\kappa_1 = 2.0738 \pm 0.021 \text{ (see Appendix B)}$$

$$\kappa_2 = 1.8954 \pm 0.019 \text{ (see Appendix B)}$$

$$N_2 = 16,457,804 \text{ fissions}$$

$$N_3 = 24,821 \pm 158 \text{ long-range particles}$$

Substitution in Eq. (2) gives

$$T/B = (3,343 \pm 0.20) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 299.2 \pm 18 \text{ fissions per long-range particle.}$$

The estimated uncertainties in the quantities listed above arise from the following considerations. Those in Ω_1 and Ω_2 are due to the

uncertainties in determining the spacing of the electrodes and the diameters of apertures of the aluminum windows. The error in ϵ involves the statistics of counts taken on each side of the source as well as the errors in Ω_1 and Ω_2 . The uncertainty in τ_1 and τ_2 is more difficult to assess, as an estimate had to be made of the possible error engendered by an uncertainty in the low-energy end of the distribution in Fig. 6 (see Chapter I for a discussion of the energy distribution). A study of this factor, as well as that of the accuracy of the determination of the area under the curve in Fig. 6 and of the precision of the location of the lower energy cutoff points, etc., leads to an overall estimate of the uncertainty in τ_1 and τ_2 of approximately $\pm 5\%$. The geometry and source fission fragment absorption correction factors K_1 and K_2 are believed to be known to better than 1% (see Appendix B).

The error on N_3 is the standard deviation of the number of counts. Equation (2) involves N_2 and N_3 only in the ratio N_3/N_2 and, since $N_2 \gg N_3$, the standard deviation on N_2 is neglected.

The uncertainty in the ratio T/B is its standard deviation derived from the uncertainties listed above.

In the description of the measurements on the remainder of the nuclides only the results of the error analysis will be given.

Spontaneous Fission of Cm^{242}

A $0.06\text{-}\mu\text{g}/\text{cm}^2$ source having an alpha activity of 5×10^8 α/min was electrospayed⁽⁴¹⁾ on $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil which was reinforced by a 93%-transparent electromesh nickel screen. The deposit was covered by

100- $\mu\text{g}/\text{cm}^2$ nickel foil to prevent the material from spreading throughout the chamber. The Cm^{242} sample was prepared by R. W. Hoff and his group at UCLRL, Livermore, California, by irradiating pure Am^{241} in the Materials Testing Reactor for approximately 90 days in a neutron flux of $\approx 2 \times 10^{14}$ n/cm²/sec. The sample was composed mostly of Cm^{242} and Am^{241} in a ratio of about one to two, and had a Cm^{243} - Cm^{244} content well below 10 atom percent, and a trace amount of Pu^{238} . Consideration of the spontaneous fission half-lives^(42,43) involved indicates that more than 95% of the fissions observed were from Cm^{242} ; therefore no correction was made for the isotopic content in the analysis of the data. Cm^{242} has an alpha half-life of 162.5 days and a spontaneous fission half-life of 7.2×10^6 years; thus, there are on the average 1.54×10^7 radioactive alphas per fission. In practice, alpha pile-up difficulties placed an upper limit on the amount of material which could be tolerated in the chamber. The source as used gave an average fission counting rate of approximately 17 fissions per minute over a 31-day period of observation. The results obtained for Cm^{242} at 1.0-Mev bias are as follows:

$$\epsilon = 0.935$$

$$\tau_1 = 1.125$$

$$\tau_2 = 1.113$$

$$\kappa_1 = 2.1013$$

$$\kappa_2 = 1.9585$$

$$N_2 = 709,306 \text{ fissions}$$

$$N_3 = 937.4 \times 1.302 = 1,220 \text{ long-range particles.}$$

Substitution in Eq. (2) gives (the unlisted factors are as in the preceding Cf^{252} case)

$$T/B = (3.897 \pm 0.27) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 256.6 \pm 17 \text{ fissions per long-range particle.}$$

The factor 1.302 which appears in the value for N_3 above is a correction for the fact that these data were taken under conditions such that some of the counts were missed because of a timing error. This correction factor was determined by normalizing Cf^{252} N_3/N_2 ratios taken at the time to ratios taken after the error had been eliminated.

Spontaneous Fission of Cm^{244}

A $2\text{-}\mu\text{g}/\text{cm}^2$ source having an alpha activity of $5.4 \times 10^8 \alpha/\text{min}$ was electrosprayed on $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil which was reinforced by a 93%-transparent electromesh nickel screen. To prevent the source from spreading, it was covered with a $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil. Virtually all of the spontaneous fission events from the sample were attributable to Cm^{244} since the impurities consisted of only a trace of Cm^{242} and a few atom percent of Am^{243} and Am^{241} which, being odd-A nuclides, have exceedingly long spontaneous fission half-lives. (42,43)

Data were taken at 50-kev and 1.25-Mev biases. The results are given in Table 3.

Deposition in 20. (2) since the relative amount of the two

isotopes is the same.

It is a 0.27 x 10⁻¹⁰ mole fraction per fraction, or

0.27 x 10⁻¹⁰ mole fraction per fraction.

The factor 1.00 which appears in the value for 0.27 is a

factor for the fact that these data were taken under conditions

that some of the samples were placed under a higher error. This

error is shown by the value of 0.27 x 10⁻¹⁰ mole fraction

at the time of fractionation when the error had been eliminated.

Spontaneous fractionation of C¹³

A 2-gram sample having an initial activity of 0.4 x 10⁶ dpm was

electrolyzed on 100-gram nickel foil which was refined by a 30-

percent electrolytic nickel anode. To prevent the source from

overheating it was covered with a 100-gram nickel foil. After 1.4

of the spontaneous fractionation the sample was removed for analysis.

Since the fractionation consisted of only a trace of C¹³ and a few

atoms percent of C¹² and C¹⁴ which, being non-radioactive, have no effect

(48.43)

data were taken at 50-gram and 100-gram masses. The results are

given in Table I.

TABLE 3

CORRECTION FACTORS AND DATA FOR SPONTANEOUS FISSION OF Cm^{244}

	50-kev Bias	1.25-Mev Bias
ϵ	0.935	0.935
τ_1	1.1074	1.1260
τ_2	1.0985	1.1150
K_1	2.1863	2.1863
K_2	2.0329	2.0329
N_2	6,956,049	5,654,892
N_3	9,695	7,507
T/B^a	$(3.235 \pm 0.20) \times 10^{-3}$	$(3.1309 \pm 0.20) \times 10^{-3}$

^aT/B values are obtained by substitution in Eq. (2).

The weighted average of these results yields

$T/B = (3.188 \pm 0.20) \times 10^{-3}$ long-range particles per fission, or

$B/T = 313.7 \pm 20$ fissions per long-range particle.

Spontaneous Fission of Pu^{240}

The source was electrosprayed on 100- $\mu\text{g}/\text{cm}^2$ nickel foil which was reinforced by a 93%-transparent electromesh nickel screen. To prevent spreading, the source was covered with 100- $\mu\text{g}/\text{cm}^2$ nickel foil. The deposit had an area of 1.25 cm^2 and contained $\approx 180 \mu\text{g}$ of metal as determined by spontaneous fission counting. The material was sprayed

TABLE 2

RELATIVE YIELDS AND DATA FOR SEVERAL FISHES OF THE

Species	Yield (%)	Yield (%)
1. Yellow Perch	0.03	0.03
2. Rock Bass	1.00	1.00
3. White Crayfish	1.00	1.00
4. Bluegill	2.00	2.00
5. Pumpkinseed	1.00	1.00
6. Spottail	2.00	2.00
7. Rock Bass	2.00	2.00
8. Rock Bass	2.00	2.00
9. Rock Bass	2.00	2.00
10. Rock Bass	2.00	2.00
11. Rock Bass	2.00	2.00
12. Rock Bass	2.00	2.00
13. Rock Bass	2.00	2.00
14. Rock Bass	2.00	2.00
15. Rock Bass	2.00	2.00
16. Rock Bass	2.00	2.00
17. Rock Bass	2.00	2.00
18. Rock Bass	2.00	2.00
19. Rock Bass	2.00	2.00
20. Rock Bass	2.00	2.00

The values are obtained by substituting in Eq. (2).

The weighted average of these results yields

$$Y = (0.10 \times 0.03) + (0.90 \times 2.00) = 1.80 \text{ g/g}$$

Y = 0.10 g/g ± 0.03 g/g (10% error)

Experimental Results of Y

The source was electroplated on 100-μg/cm² nickel foil which was reinforced by a 535-translucent electroplated nickel screen. To prevent swelling, the source was covered with 100-μg/cm² nickel foil. The deposit had an area of 1.25 cm² and contained ~150 μg of metal as determined by monochromatized neutron activation. The material was covered

as the nitrate and then converted to the oxide by heating. This process is capable of completely converting the nitrate to the oxide; however, because of the fragile nature of the nickel foil it was necessary to carry out the conversion at a reduced temperature. This introduced a degree of uncertainty as to the extent of the conversion and hence of the stopping power of the deposit. Nevertheless, an assumption of complete conversion is given credence by the following evidence. The results of calculations described in Appendix B indicate that if the source were a uniform deposit of PuO_2 , 2.2% of the fission events would not be detected because of source absorption, and that if it were $\text{Pu}(\text{NO}_3)_4 \cdot 4(\text{H}_2\text{O})$, 5% would not be detected. An extrapolation to zero bias of the single fragment pulse-height distribution of the pulses from this source in a gridded ionization chamber revealed that approximately 2.5% of the pulses lie below the fission counter threshold. This is consistent with the pure oxide assumption, and also serves to set an upper bound on the error which could be introduced in the ratio T/B by preferential absorption of those fission events which emit long-range particles directed toward the outer chamber aluminum windows. This upper limit amounts to twice the percentage of counts which lie below the threshold or about 5%. This is because the long-range-particle counting geometry is approximately 50% and is such that the particles detected are those associated with fragments which lie closest to the source plane, and hence are most likely to be absorbed. The K_1 and K_2 factors from the calculations described in Appendix B correct for this preferential absorption.

as the system and then continued to the center of heating. This process
is capable of completely converting the system to the solid phase
because of the large amount of the initial fall in the temperature
only but the conversion of a reduced temperature. This indicates
change of temperature as the result of the solidification process
the typical lower of the system. Nevertheless, an assumption of the
phase conversion is given previously by the following evidence. The
weight of crystalline substance is assumed to be constant from the
start with a uniform deposit of 100, 2.3% of the fraction events which
not be detected because of minor absorption, and that it is
100% at 100% of the weight was detected. An extrapolation to zero
value of the change in weight yields a linear distribution of the phase
this source is a further localized chamber revealed that approximately
2.3% of the weight is below the fraction constant threshold. This is
consistent with the phase change assumption, and also serves to set an
upper bound on the error which could be introduced in the extrapolation
extrapolated assumption of those fraction events which are long-range
pertinent directed toward the outer chamber aluminum window. This upper
limit remains to show the percentage of source which lies below the
threshold at about 5%. This is because the long-range events counting
accuracy is approximately 5% and is such that the particles detected are
those associated with fragments which lie closest to the source plane,
and hence are most likely to be absorbed. The K_{α} and K_{β} factors that
the calculations described in Figure 2 correct for this absorption
absorption.

The plutonium used in this measurement is described in the March 23, 1959, Research Pool Inventory, Batch #13A-D2S, as 99.79% pure Pu^{240} . The source gave a counting rate of 5.13 fissions per minute and observations were made over a period of one month, during which time 253 long-range particles were recorded. Background runs totaling approximately 11.5 days were also made. In these background runs the aluminum windows were blanked so that no true triple coincidences could occur. Five accidental triple coincidence counts were recorded during the period, which implies an accidental rate of approximately 5.4% of the true rate. This was the highest percentage accidental rate encountered in the spontaneous fission measurements. All the other sources gave higher long-range particle counting rates and lower percentage backgrounds.

The results obtained for Pu^{240} at 50-kev bias are as follows:

$$\epsilon = 0.935$$

$$\tau_1 = 1.1074$$

$$\tau_2 = 1.0985$$

$$K_1 = 2.2232$$

$$K_2 = 2.0665$$

$$N_2 = 231,291 \text{ fissions}$$

$$N_3 = 239.6 \times 1.302^* = 312 \text{ long-range particles.}$$

*Timing correction described in the Cm^{242} case above.

Substitution in Eq. (2) gives

$$T/B = (3.18 \pm 0.30) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 314 \pm 30 \text{ fissions per long-range particle.}$$

Spontaneous Fission of Pu²⁴²

The source was electrosprayed on 100- $\mu\text{g}/\text{cm}^2$ nickel foil which was reinforced by a 93%-transparent electromesh nickel screen. To prevent spreading the source was covered with a 100- $\mu\text{g}/\text{cm}^2$ nickel foil. The deposit had an area of 1.25 cm^2 and contained $\approx 290 \mu\text{g}$ of Pu²⁴² as determined by spontaneous fission counting. The source gave a rate of about 12.4 fissions per minute and was physically similar to the Pu²⁴⁰ source described in the previous section, the same uncertainty existing with respect to its final chemical form. A study of its pulse-height distribution indicated that no more than approximately 2% of the fission fragments were absorbed in the source and backing material, a fact which was again consistent with the assumption that the chemical form of the deposit was mostly PuO₂. The Pu²⁴² sample, which was supplied by Paul Fields of the Chemistry Division of the Argonne National Laboratory, had the following composition (in atom percent):

$$\text{Pu}^{239} - 0.391 \pm 0.007$$

$$\text{Pu}^{240} - 0.226 \pm 0.005$$

$$\text{Pu}^{241} - 7.51 \pm 0.07$$

$$\text{Pu}^{242} - 88.51 \pm 0.08$$

$$\text{Pu}^{244} - 0.033 \pm 0.002$$

Identification in Sp. (2) gives

$P_{1.25} = 0.19 \pm 0.03$ for range particles per liter, or

$P_{1.25} = 0.19 \pm 0.03$ for range particles

Statistical Analysis of Data

The source was electromagnetically on 100-1000 m² nickel foil which was

exposed by a 35% transparent electrochromic nickel screen. No previous

exposure the source was covered with a 100-1000 m² nickel foil. The

exposure had an area of 1.25 cm² and contained a 200 mg of $P_{1.25}$ as

detected by spontaneous fission counting. The source gave a rate of

about 12.4 fissions per minute and was physically similar to the $P_{1.25}$

source described in the previous section, the same underlying existing

with respect to the final chemical form. A study of the nuclear decay

characteristics indicated that no more than approximately 25% of the fission

fragments were absorbed in the source and taking account of that which

was again consistent with the assumption that the chemical form of the

source was mostly $P_{1.25}$. The $P_{1.25}$ sample, which was supplied by Paul

Smith of the Chemistry Division of the Argonne National Laboratory, had

the following composition (in atom percent):

$$P_{1.25} = 0.291 \pm 0.007$$

$$P_{1.25} = 0.226 \pm 0.005$$

$$P_{1.25} = 1.81 \pm 0.05$$

$$P_{1.25} = 22.51 \pm 0.03$$

$$P_{1.25} = 0.025 \pm 0.002$$

It may be noted that none of the constituents other than Pu^{242} was present in sufficient quantity to add significantly to the spontaneous fission rate. However, the presence of Pu^{241} in the amount of 7.51% allowed this same source to be used for an observation of thermal-neutron-induced fission. The results obtained for Pu^{242} at 50-kev bias are as follows:

$$\epsilon = 0.935$$

$$\tau_1 = 1.1074$$

$$\tau_2 = 1.0985$$

$$\kappa_1 = 2.2401$$

$$\kappa_2 = 2.0820$$

$$N_2 = 518,662 \text{ fissions}$$

$$N_3 = 459.5 \times 1.302^* \approx 598.3 \text{ long-range particles.}$$

Substitution in Eq. (2) gives

$$T/B = (2.742 \pm 0.21) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 364.7 \pm 29 \text{ fissions per long-range particle.}$$

Thermal-Neutron-Induced Fission Measurements

In the following measurements the ionization chamber assembly was placed in an external beam from the south thermal column of the Los Alamos Homogeneous Reactor, which was operated at 10- to 25-KW power.

*Timing correction described in Cm^{242} case above.

It may be noted that none of the constituents other than Pu^{239} was present in sufficient quantity to add significantly to the spontaneous fission rate. However, the presence of Pu^{239} in the amount of 1.51g allowed this source to be used for an observation of thermal-neutron induced fission. The λ value obtained for Pu^{239} at 50-kev was as follows:

$$\lambda_1 = 0.982$$

$$\lambda_2 = 1.104$$

$$\lambda_3 = 1.098$$

$$\lambda_4 = 2.261$$

$$\lambda_5 = 2.0820$$

$$M = 518.686 \text{ fissions}$$

$$M_1 = 459.3 \times 1.302 = 598.3 \text{ long-range particles}$$

Substitution in Eq. (2) gives

$$T/D = (2.74 \pm 0.2) \times 10^{-7} \text{ long-range particles per fission, or}$$

$$S/T = 364.7 \pm 22 \text{ fissions per long-range particle}$$

Thermal-Neutron-Induced Fission Measurements

In the following measurements the ionization chamber assembly was placed in an external beam from the south thermal column of the Los Alamos Neutron Source, which was operated at 1- to 25-MeV power.

*Timing corrected described in Eq. (2) also above.

In all other respects the experimental conditions were identical to those used in the spontaneous fission measurements.

Thermal-Neutron-Induced Fission of Pu²⁴¹

The source used here was the same as that used in the preceding measurement on spontaneous Pu²⁴² fission. A consideration of the isotopic composition of the source material given in the preceding section, together with the fact that the thermal neutron fission cross-section ratio of Pu²⁴¹ to that of Pu²⁴² is greater than 5000 to 1,⁽⁴⁴⁾ indicates that more than 99% of the fissions observed were from thermal-neutron-induced fission of Pu²⁴¹.

The results obtained at 1.25-Mev bias are as follows:

$$\epsilon = 0.935$$

$$\tau_1 = 1.1260$$

$$\tau_2 = 1.115$$

$$K_1 = 2.2401$$

$$K_2 = 2.0820$$

$$N_2 = 3,356,431 \text{ fissions}$$

$$N_3 = 3,158 \text{ long-range particles.}$$

Substitution in Eq. (2) gives

$$T/B = (2.272 \pm 0.15) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 440.1 \pm 28 \text{ fissions per long-range particle.}$$

It is found that the experimental conditions are identical to those

described in the preceding section.

Experimental Results

The first result is that the rate of reaction is independent of the

concentration of the reactants. A plot of $\log k$ versus $\log [\text{A}]$ gives a

straight line with a slope of 1.0, indicating a first-order reaction.

From the slope of this line, the rate constant k was calculated to be

$k = 0.015 \text{ s}^{-1}$ at 25°C . The activation energy E_a was determined to be

$E_a = 50.0 \text{ kJ mol}^{-1}$ from a plot of $\log k$ versus $1/T$.

The experimental results are summarized in Table I.

The rate constant k at 25°C is compared with values reported in the

literature in Table II.

The rate constant k at 25°C is 0.015 s^{-1} .

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The rate constant k at 25°C is 0.015 s^{-1} .

The rate constant k at 25°C is 0.015 s^{-1} .

It is of interest to compare this result with that obtained for spontaneous fission of Pu^{242} . The same nuclide is involved in both cases, the principal differences being in the spins and in the 6.4 Mev of binding energy available from the neutron capture in the latter case. The intrinsic ground-state spin of a Pu^{242} nucleus is $I = 0^+$ and its spin after formation by thermal-neutron capture is a combination of the Pu^{241} ground-state spin⁽⁴⁵⁾ of $5/2^+$ and the spin of the neutron (i.e., $5/2 \pm 1/2$). Thus the spin difference between the two cases is either 2 or 3, depending upon the neutron orientation.

Thermal-Neutron-Induced Fission of Pu^{239}

The source was a vacuum-evaporated 123- μg deposit of 2- cm^2 area on 100- $\mu\text{g}/\text{cm}^2$ nickel foil and had no reinforcing screen or cover foil. The isotopic composition of the sample was Pu^{239} - 94.0%, Pu^{240} - 5.33%, and Pu^{241} - 0.33 to 0.5%. The results obtained at 1.25-Mev bias are as follows:

$$\epsilon = 1.0$$

$$\tau_1 = 1.126$$

$$\tau_2 = 1.1150$$

$$K_1 = 2.2053$$

$$K_2 = 2.0501$$

$$N_2 = 12,106,356 \text{ fissions}$$

$$N_3 = 12,363 \text{ long-range particles.}$$

Substitution in Eq. (2) gives

$$T/B = (2.432 \pm 0.15) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 411.2 \pm 26 \text{ fissions per long-range particle.}$$

It may be noted that here again the same nuclide is undergoing fission as in the previous case of spontaneous fission of Pu^{240} , the differences being in the spins and in the excitation due to the binding energy of the neutron captured in the latter case. The intrinsic ground-state spin of Pu^{240} is $I = 0^+$ and the Pu^{239} ground-state spin⁽⁴⁵⁾ is $1/2^+$, hence $I = 1/2 \pm 1/2$ and the spin difference is either 0 or 1 depending upon the neutron orientation.

Thermal-Neutron-Induced Fission of U^{235}

In this measurement four sources of differing thickness were used to check experimentally the source self-absorption calculations described in Appendix B. These sources had the following amounts of 99.84%-pure U^{235} : 53 μg , 307 μg , 330 μg , and 860 μg . The sources were 2 cm^2 in area and were vacuum-evaporated on 100- $\mu\text{g}/\text{cm}^2$ nickel foil and had no reinforcing screens or cover foils. The results obtained at 1.25-Mev bias are given in Table 4.

The weighted average of the results from the four samples gives

$$T/B = (2.004 \pm 0.12) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 499 \pm 30 \text{ fissions per long-range particle.}$$

distillation in 24 hr (2) after

$$W_2 = (2.42 \pm 0.12) \times 10^{-5} \text{ long-range particles per flashon, or}$$

$$W_2 = 4.9 \pm 2.0 \text{ flashons per long-range particle}$$

It may be noted that here again the same method is used.

Flashon 22 is the previous case of spontaneous flashon of W_2 at

distillation before in the flash and in the calculation due to the flash

energy of the reaction captured in the flash. The intrinsic flash

event again at W_2 is $W_2 = 0$ and the W_2 flashon flash

flash, hence $W_2 = 1/2$ and the same difference is either 0 or 1

depending upon the reaction orientation.

Flashon-Flashon-Induced Flashon of W_2

In this experiment four sources of different thickness were used

to check experimentally the source self-absorption relationship. Flashon 22

is reported. These sources had the following amounts of W_2 flashon:

Flashon 22, 30, 40, 50, and 60 mg. The sources were 2 mm and 4 mm

thick and were vacuum-evaporated on 10- μ metal foil and had a

reflecting surface of cover foil. The results obtained at 1.50 MeV

are given in Table 4.

The weighted average of the results from the four samples give

$$W_2 = (2.00 \pm 0.12) \times 10^{-5} \text{ long-range particles per flashon, or}$$

$$W_2 = 4.9 \pm 2.0 \text{ flashons per long-range particle}$$

TABLE 4
CORRECTION FACTORS AND DATA FOR
THERMAL-NEUTRON-INDUCED FISSION OF U^{235}

	53 μg	307 μg	330 μg	864 μg
ϵ	1	1	1	1
τ_1	1.126	1.126	1.126	1.126
τ_2	1.115	1.115	1.115	1.115
κ_1	2.2193	2.2560	2.2660	2.3550
κ_2	2.0636	2.1000	2.1060	2.1870
N_2	7,268,229	13,077,126	12,387,099	22,778,952
N_3	6,128	10,713	10,089	18,012
T/B^a	$(2.020 \pm 0.125) \times 10^{-3}$	$(1.996 \pm 0.123) \times 10^{-3}$	$(1.994 \pm 0.120) \times 10^{-3}$	$(2.009 \pm 0.123) \times 10^{-3}$

^aT/B values are obtained by substitution in Eq. (2).

The errors on the ratios T/B listed in Table 4 are the estimated absolute errors previously discussed in the Cr^{252} measurement. However, the deviation from the mean of each of the T/B ratios falls within its relative error which is based on the standard deviation on N_3 only. This indicates that the source self-absorption calculation of Appendix B gives consistent results when used on deposits of differing thicknesses.

Thermal-Neutron-Induced Fission of U^{235}

A 2-cm² area source was vacuum-evaporated on 100- $\mu\text{g}/\text{cm}^2$ nickel foil and had no reinforcing screen or cover foil. The weight of the

deposit was ≈ 1.0 mg as determined by its fission counting rate in the known flux from the reactor. Its isotopic composition was as follows (in atom percent):

$$U^{233} - 98.02 \text{ to } 98.19$$

$$U^{234} - 1.33$$

$$U^{238} - 0.5 \text{ to } 0.6$$

Most of the remainder was U^{235} with a trace of U^{232} . It was necessary to separate the sample chemically shortly before use in order to break radioactive decay chains which produce high-energy alphas. These chains start with U^{232} and U^{233} and pass through Po^{212} and Po^{214} which emit alpha particles of 8.78 and 8.34 Mev, respectively. These high-energy alphas were objectionable because their ranges permitted them to enter the long-range particle chambers and increase the accidental triple coincidence rate to an objectionable level.

The results obtained at 1.25-Mev bias are as follows:

$$\epsilon = 1.0$$

$$\tau_1 = 1.126$$

$$\tau_2 = 1.115$$

$$K_1 = 2.380$$

$$K_2 = 2.208$$

$$N_2 = 22,946,539 \text{ fissions}$$

$$N_3 = 21,587 \text{ long-range particles.}$$

deposit was ~ 1.0 mg as determined by the titration method.

known from the reaction. The isotopic composition was as follows:

(in atom percent):

$U^{235} = 98.02$ to 98.19

$U^{238} = 1.81$

$U^{234} = 0.17$ to 0.2

Most of the remaining was U^{235} with a trace of U^{238} .

To separate the sample chemically, firstly before use in order to avoid

radioactive decay chains which produce high-energy alpha.

start with U^{235} and U^{238} and pass through Th^{231} and Pa^{231} which emit

alpha particles of 5.76 and 5.54 MeV, respectively. These high-energy

alpha were objectionable because their ranges exceeded those of alpha

the long-range particle chambers and increase the accidental counts

consequently rate to an objectionable level.

The results obtained at 1.55-MeV are as follows:

$\epsilon = 1.0$

$r_1 = 1.125$

$r_2 = 1.115$

$k_1 = 2.330$

$k_2 = 2.308$

$N_0 = 22,940,000$ fission

$N_1 = 21,887$ long-range particles

Substitution in Eq. (2) gives

$$T/B = (2.415 \pm 0.15) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 414.1 \pm 26 \text{ fissions per long-range particle.}$$

CHAPTER III

MEASUREMENT OF THE TERNARY-TO-BINARY FISSION RATIO FOR FISSION INDUCED BY 1-MEV NEUTRONS

For the measurement of the ternary-to-binary fission ratio for fission induced by 1-Mev neutrons, the ionization chambers were arranged in the following manner. The long-range particle chamber having electrode 5 as its collector (see Fig. 10, 11, or 12) was removed and the remaining electrode assembly moved closer to the end wall of the chamber. A neutron target 3 cm in length and filled with tritium gas was placed coaxially with the chamber and as near the end wall as possible. The elimination of the one long-range particle chamber and moving of the electrodes decreased the detection efficiency for long-range particles by a factor of one-half and also decreased the average distance to the neutron source by approximately the same factor. Because of the $\approx 1/r^2$ neutron flux variation from the target there was a net gain of a factor of almost two in counting rate from this modification.

Measurements taken with the Cf^{252} spontaneous fission source normalized the long-range particle counting efficiency to that of the previous double chamber setup.

One of the difficulties encountered in this phase of the work was a neutron-induced background which was produced mostly by proton recoils and n-p reactions in the chamber electrodes, walls, and filling gas. The difficulty was most acute in the long-range particle channel where

MEASUREMENT OF THE EFFECT OF TEMPERATURE ON THE

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the relatively low bias setting of 1 Mev permitted a large number of pulses to be formed, and the slow coincidence time (0.8 μ sec) increased the probability of their making an accidental threefold coincidence. (See Chapter II for a discussion of the bias settings and coincidence resolution time of this channel.) A large accidental background in the threefold coincidence channel is especially deleterious because of the low counting rate of this channel.

To minimize background difficulties, a system of guard electrodes was installed (Fig. 17). These electrodes restricted the sensitive volume of the chambers to that which was useful for detecting particles from fission. The electrodes and walls of the chamber were covered with gold wherever practical and the filling gas was changed from an argon-CO₂ mixture to a krypton-CO₂ mixture, these both being well known techniques^(46,47) for decreasing the background in ionization chambers used in intense high-energy neutron fluxes. With the new gas filling it was necessary to change the gas pressure from 1880 mm Hg to 1240 mm Hg in order to maintain the same stopping power. Energy calibration and timing measurements similar to those described in Chapter II were made to establish the proper operating conditions.

All surfaces inside the chamber were kept as clean as possible and then given a final wash with electronics grade trichloroethylene just before assembly. The krypton and CO₂ were spectroscopically and chemically pure grades respectively and were dried with anhydrous P₂O₅ to remove residual moisture. In use, the gas mixture was circulated through

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Fig. 17.--Electrode assembly of the ionization chamber apparatus used in the 1-Mev neutron-induced fission measurements. View shows the pressure vessel removed. A centimeter rule is included for scale.

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a convective 350°C calcium purifier and then passed over activated uranium.

In spite of these precautions a pernicious energy-dependent neutron-induced background was encountered which in practice placed an upper limit of about 1 Mev on the usable neutron energy. The amplitude of these background pulses varied in approximately direct proportion to the neutron energy. Therefore the pulses were probably due to proton recoils produced in residual hydrogenous vapors in the filling gas and in hydrogenous compounds and occluded hydrogen on the electrode surfaces.

Neutrons were produced by the $T(p,n)He^3$ reaction at the Los Alamos large Van de Graaff accelerator from a tritium gas target 3 cm in length. The target had a 9.9-mg/cm² molybdenum window and was operated at a pressure of ≈ 62 lbs/in² absolute. The target design was similar to that described in reference (48) except that it had only a single entrance window and no gas cooling. Beam currents were in the 4- to 7- μ amp range and produced neutron fluxes up to about 10^8 n/cm²/sec at the fission sample.

The correction equation which pertains to the single long-range particle chamber setup as it was used in this part of the experimental work is

$$T/B = \frac{N_3 K_1 \tau_1}{N_2}$$

where T/B is the ternary-to-binary fission ratio, τ_1 is the correction factor for the low-energy cutoff of the long-range particle spectrum,

K_1 is the factor which corrects for geometry and source fragment absorption (see Appendix B), N_2 is the binary count for the observation period corrected for background, and N_3 is the long-range particle or ternary count for the same observation period corrected for background.

Pu^{239} Fission Induced by 1-Mev Neutrons

The source was a vacuum-evaporated 1.1-mg deposit of 2-cm² area on 100- $\mu\text{g}/\text{cm}^2$ nickel foil and had no reinforcing screen or cover foil. The isotopic composition of the sample was the same as that described in the section on thermal-neutron-induced fission of Pu^{239} . The target provided an average neutron energy of 1.0 Mev with a thickness of approximately 500 kev. The results obtained at 1.0-Mev bias are as follows:

$$\tau_1 = 1.125 \pm 0.056$$

$$K_1 = 4.3460 \pm 0.043$$

$$N_2 = 1,700,851 \text{ fissions}$$

$$N_3 = 1035 - 172 \text{ BG} = 863 \pm 34.6 \text{ long-range particles.}$$

Substitution in Eq. (3) gives

$$T/B = (2.481 \pm 0.16) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 403.1 \pm 25 \text{ fissions per long-range particle.}$$

The errors on τ_1 and K_1 are discussed in the section on spontaneous fission of Cf^{252} . The background or accidental count correction on N_2

At the lowest values of α the observed spectrum is
 almost identical with the theoretical spectrum for the
 case of a uniform distribution of particles of energy
 E_0 (see Appendix A). As α increases the spectrum
 becomes more complicated and the observed spectrum
 deviates from the theoretical spectrum for a uniform
 distribution of particles.

2. Theoretical spectrum for a uniform distribution of particles

The source was a vacuum-evaporated ^{238}Pu deposit of 5-mg weight
 on a 1-cm 2 area of a 1-cm-thick aluminum foil. The
 thickness of the foil was 0.01 cm. The energy of the
 alpha particles was 5.5 MeV. The theoretical spectrum
 for a uniform distribution of particles is shown in
 Figure 1. The spectrum is a function of the energy
 of the alpha particles and the thickness of the foil.

$$I = I_0 \exp(-\alpha E)$$

$$I = I_0 \exp(-\alpha E)$$

$$I = I_0 \exp(-\alpha E)$$

$$I = I_0 \exp(-\alpha E)$$

where

$$I = \text{intensity of the spectrum}$$

$$I_0 = \text{intensity of the spectrum for a uniform distribution of particles}$$

$$\alpha = \text{absorption coefficient of the foil}$$

The energy of the alpha particles is 5.5 MeV.

The thickness of the foil is 0.01 cm.

was of the order of 0.02%, hence insignificant. Here, as well as in the sections which follow, the listed uncertainty in the ratio T/B is its standard deviation derived from uncertainties on the factors involved.

U²³⁵ Fission Induced by 1-Mev Neutrons

The fission source used here was the same 864- μ g sample described in the section on thermal-neutron-induced fission of U²³⁵. The average neutron energy was 1 Mev with a target thickness of approximately 500 kev. The results obtained at 1.0-Mev bias are as follows:

$$\tau_1 = 1.125 \pm 0.056$$

$$K_1 = 4.3125 \pm 0.043$$

$$N_2 = 2,519,205 \text{ fissions}$$

$$N_3 = 1269 - 297 \text{ BG} = 972 \pm 40 \text{ long-range particles.}$$

Substitution in Eq. (3) gives

$$T/B = (1.872 \pm 0.12) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 534.2 \pm 35 \text{ fissions per long-range particle.}$$

U²³⁴ Fission Induced by 1-Mev Neutrons

The source had a U²³⁴ weight of $\approx 890 \mu\text{g}$ and an area of 2 cm^2 and was prepared by the painting technique on $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil. No reinforcing screen or cover foil was used. The U²³⁴ sample had an admixture of about 2% U²³⁵; this allowed a thermal neutron measurement

was of the order of 0.01, being insignificant. There is also a 1.5

section which follows the linear uncertainty in the ratio of the

average deviation between the two measurements on the same samples.

2.1. Results obtained by 1.5 MeV neutrons

The neutron source used here was the same 200- μ sample described

in the section on thermal neutron-induced reaction of ^{235}U . The average

neutron energy was 1.5 MeV when a narrow spectrum of approximately 10% was

the results obtained at 1.5 MeV were as follows:

$$\sigma = 1.125 \pm 0.008$$

$$\sigma = 1.125 \pm 0.008$$

$$\sigma = 1.125 \pm 0.008$$

$$\sigma = 1.125 \pm 0.008$$

$$\sigma = 1.125 \pm 0.008$$

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$$\sigma = 1.125 \pm 0.008$$

$$\sigma = 1.125 \pm 0.008$$

2.2. Results obtained by 1.5 MeV neutrons

The source had a V_{eff} value of ≈ 200 μ and an area of 2 cm² and

was prepared by the following technique on 100- μ sample, typical of

radioactive source of lower foil was used. The V_{eff} sample had an

thickness of about 25 μ , this allowed a thermal neutron measurement

to be made and hence a normalization to previous U^{235} thermal neutron data. The use of this normalization permitted an accurate correction for source thickness effects with respect to previous U^{235} data. One-Mev neutrons were obtained from the target arrangement described previously. The results obtained at 1.0-Mev bias are as follows:

$$\tau_1 = 1.125 \pm 0.056$$

$$K_1 = 4.400 \pm 0.088$$

$$N_2 = 2,733,341 \text{ fissions}$$

$$N_3 = 1534 - 348 \text{ BG} = 1187 \pm 43 \text{ long-range particles.}$$

Substitution in Eq. (3) gives

$$T/B = (2.148 \pm 0.14) \times 10^{-3} \text{ long-range particles per fission, or}$$

$$B/T = 465.5 \pm 30 \text{ fissions per long-range particle.}$$

to 100% and hence a concentration of particles in the central region.

data. The use of this transformation permitted an accurate correction

for source thickness effects with respect to previous ^{137}Cs data. The data

obtained were corrected from the largest arrangement described previously.

The results obtained at 1.0-MeV, 1.5-MeV and 2.0-MeV are:

$$N_1 = 1.125 \pm 0.025$$

$$N_2 = 1.125 \pm 0.025$$

$$N_3 = 1.125 \pm 0.025$$

$$N_4 = 1.125 \pm 0.025$$

particles.

Concentration in g/cm^3 gives

$$N_1 = (1.125 \pm 0.025) \times 10^{10} \text{ long-range particles per flask, or}$$

$$N_1 = 1.125 \pm 0.025 \text{ long-range particles per flask, or}$$

CHAPTER IV

RESULTS AND CONCLUSIONS OF THE MEASUREMENT OF THE TERNARY-TO-BINARY FISSION RATIO

The results of the ternary-to-binary fission ratio, T/B , measurements are given in Table 5. For convenience the inverse ratios, B/T , are also tabulated since this is the value usually quoted in the literature. Also listed are the Z^2/A and X values for each nuclide. Bohr and Wheeler⁽⁴⁾ first introduced the so-called fissionability parameter, X , which can be written

$$X = \frac{Z^2/A}{(Z^2/A)_0}$$

where $(Z^2/A)_0$ is the limiting value for a spherical nucleus which is just unstable with respect to small distortions. In a recent review article on nuclear fission Hyde⁽⁴⁹⁾ presents a detailed discussion of the fissionability parameter X and gives 50.13 as the best value for $(Z^2/A)_0$.

Figure 18 shows a plot of the number of long-range particles per fission (i.e., ternary-to-binary fission ratio) as a function of X and also as a function of Z^2/A . It may be noted that the number increases for increasing X values and that for a fixed X the ratio decreases with increasing spin and excitation energy.

Table 6 gives the observed variation of the relative probability of long-range particle emission as a function of excitation energy.

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$$\frac{E_1}{T_1} = \frac{E_2}{T_2}$$

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of long-range particle emission as a function of ...

Fig. 18.--The number of long-range particles per fission as a function of Z^2/A and X .

X

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TABLE 5
RESULTS OF MEASUREMENTS OF THE NUMBER OF LONG-RANGE PARTICLES PER FISSION

Nuclide	Type Fission	Z^2/A	X^a	Long-Range Particles per Fission (T/B)	Fissions per Long-Range Particle (B/T)
Cf ²⁵²	Spontaneous	38.11	0.7602	$(3.343 \pm 0.20) \times 10^{-3}$	299.2 ± 18
Cm ²⁴²	Spontaneous	38.08	0.7596	$(3.897 \pm 0.27) \times 10^{-3}$	256.6 ± 17
Cm ²⁴⁴	Spontaneous	37.77	0.7534	$(3.188 \pm 0.20) \times 10^{-3}$	313.7 ± 20
Pu ²⁴⁰	Spontaneous	36.82	0.7345	$(3.180 \pm 0.30) \times 10^{-3}$	314 ± 20
Pu ²⁴⁰	Pu ²³⁹ + n _{Th}	36.82	0.7345	$(2.432 \pm 0.15) \times 10^{-3}$	411.2 ± 26
Pu ²⁴⁰	Pu ²³⁹ + n ₁ Mev	36.82	0.7345	$(2.481 \pm 0.16) \times 10^{-3}$	403.1 ± 25
Pu ²⁴²	Spontaneous	36.51	0.7283	$(2.742 \pm 0.21) \times 10^{-3}$	364.7 ± 29
Pu ²⁴²	Pu ²⁴¹ + n _{Th}	36.51	0.7283	$(2.272 \pm 0.15) \times 10^{-3}$	440.1 ± 28
U ²³⁶	U ²³⁵ + n _{Th}	35.86	0.7153	$(2.004 \pm 0.12) \times 10^{-3}$	499 ± 30
U ²³⁶	U ²³⁵ + n ₁ Mev	35.86	0.7153	$(1.872 \pm 0.12) \times 10^{-3}$	534.2 ± 35
U ²³⁵	U ²³⁴ + n ₁ Mev	36.02	0.7185	$(2.148 \pm 0.14) \times 10^{-3}$	465.5 ± 30
U ²³⁴	U ²³³ + n _{Th}	36.17	0.7215	$(2.415 \pm 0.15) \times 10^{-3}$	414.4 ± 26

^aWhere $X = \frac{Z^2/A}{50.13}$

TABLE 1. Results of measurements of the amount of water vapor in the air.

Point	Time of day	Temp. of air, °C	Humidity, g/m ³	Relative humidity, %
1	8:00	18.5	12.5	62
2	9:00	19.0	13.0	63
3	10:00	19.5	13.5	64
4	11:00	20.0	14.0	65
5	12:00	20.5	14.5	66
6	13:00	21.0	15.0	67
7	14:00	21.5	15.5	68
8	15:00	22.0	16.0	69
9	16:00	22.5	16.5	70
10	17:00	23.0	17.0	71
11	18:00	23.5	17.5	72
12	19:00	24.0	18.0	73
13	20:00	24.5	18.5	74
14	21:00	25.0	19.0	75
15	22:00	25.5	19.5	76
16	23:00	26.0	20.0	77
17	24:00	26.5	20.5	78
18	1:00	27.0	21.0	79
19	2:00	27.5	21.5	80
20	3:00	28.0	22.0	81
21	4:00	28.5	22.5	82
22	5:00	29.0	23.0	83
23	6:00	29.5	23.5	84
24	7:00	30.0	24.0	85

Table 1. Results of measurements of the amount of water vapor in the air.

TABLE 6
VARIATION OF RELATIVE PROBABILITY OF LONG-RANGE
PARTICLE EMISSION WITH EXCITATION ENERGY

Nuclide and Spin	Type Fission	Neutron Energy (Mev)	Excitation (Mev)	Relative Probability of Long-Range Particle Emission		
				Present Data	Netter et al. ^a	Mostovoi ^b
Pu ²⁴⁰ 0 ⁺	Spontaneous	. .	0	1	. .	1
Pu ²⁴⁰ 0 ⁺ , 1 ⁺	Pu ²³⁹ + n	Thermal	6.4	0.765 ± 0.086	1	0.625 ± 0.059
Pu ²⁴⁰	Pu ²³⁹ + n	0.330	6.74	. .	1.05 ± 0.10	. .
Pu ²⁴⁰	Pu ²³⁹ + n	0.690	7.09	. .	0.90 ± 0.10	. .
Pu ²⁴⁰	Pu ²³⁹ + n	1.0	7.4	0.780 ± 0.090	. .	. .
Pu ²⁴⁰	Pu ²³⁹ + n	1.99	8.39	. .	1.075 ± 0.27	. .
Pu ²⁴² 0 ⁺	Spontaneous	. .	0	1	. .	. .
Pu ²⁴² 2 ⁺ , 3 ⁺	Pu ²⁴¹ + n	Thermal	6.3	0.829 ± 0.084	. .	. .
U ²³⁶ 3 ⁻ , 4 ⁻	U ²³⁵ + n	Thermal	6.5	1	1	. .
U ²³⁶	U ²³⁵ + n	0.330	6.83	. .	1.0 ± 0.13	. .
U ²³⁶	U ²³⁵ + n	1.0	7.5	0.934 ± 0.082	. .	. .
U ²³⁶	U ²³⁵ + n	1.165	7.66	. .	0.82 ± 0.09	. .
U ²³⁶	U ²³⁵ + n	2.50	9.0	. .	1.29 ± 0.18	. .
U ²³⁴ 2 ⁺ , 3 ⁺	U ²³³ + n	Thermal	6.8	. .	1	. .
U ²³⁴	U ²³³ + n	0.330	7.13	. .	1.23 ± 0.12	. .
U ²³⁴	U ²³³ + n	0.690	7.49	. .	0.93 ± 0.10	. .
U ²³⁴	U ²³³ + n	1.165	7.96	. .	0.89 ± 0.10	. .
U ²³⁴	U ²³³ + n	1.990	8.79	. .	1.05 ± 0.15	. .

^aReference (33).

^bReference (50).

TABLE 1
RELATIVE HUMIDITY OF AIR IN
VARIOUS PARTS OF THE BUILDING

Room	Time	Relative Humidity of Air (%)	Relative Humidity of Surface (%)	Relative Humidity of Air (%)	Relative Humidity of Surface (%)
Room 1	10:00	75	85	75	85
Room 1	11:00	75	85	75	85
Room 1	12:00	75	85	75	85
Room 1	13:00	75	85	75	85
Room 1	14:00	75	85	75	85
Room 1	15:00	75	85	75	85
Room 1	16:00	75	85	75	85
Room 1	17:00	75	85	75	85
Room 1	18:00	75	85	75	85
Room 1	19:00	75	85	75	85
Room 1	20:00	75	85	75	85
Room 1	21:00	75	85	75	85
Room 1	22:00	75	85	75	85
Room 1	23:00	75	85	75	85
Room 1	24:00	75	85	75	85
Room 2	10:00	75	85	75	85
Room 2	11:00	75	85	75	85
Room 2	12:00	75	85	75	85
Room 2	13:00	75	85	75	85
Room 2	14:00	75	85	75	85
Room 2	15:00	75	85	75	85
Room 2	16:00	75	85	75	85
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Room 2	18:00	75	85	75	85
Room 2	19:00	75	85	75	85
Room 2	20:00	75	85	75	85
Room 2	21:00	75	85	75	85
Room 2	22:00	75	85	75	85
Room 2	23:00	75	85	75	85
Room 2	24:00	75	85	75	85
Room 3	10:00	75	85	75	85
Room 3	11:00	75	85	75	85
Room 3	12:00	75	85	75	85
Room 3	13:00	75	85	75	85
Room 3	14:00	75	85	75	85
Room 3	15:00	75	85	75	85
Room 3	16:00	75	85	75	85
Room 3	17:00	75	85	75	85
Room 3	18:00	75	85	75	85
Room 3	19:00	75	85	75	85
Room 3	20:00	75	85	75	85
Room 3	21:00	75	85	75	85
Room 3	22:00	75	85	75	85
Room 3	23:00	75	85	75	85
Room 3	24:00	75	85	75	85

Between 10:00 and 11:00

Between 11:00 and 12:00

Results are given for the present data and for data reported by Netter et al.⁽³³⁾ and Mostovoi.⁽⁵⁰⁾ Excitation energy is taken to be zero in the case of spontaneous fission, and for neutron-induced fission the excitation energy is the binding energy of the neutron plus its kinetic energy. For each set of data in Table 6 the value of the relative probability is taken as unity for the lowest excitation energy observed. In the present data and in those of Mostovoi there is a trend toward decreasing probability of emission with increasing excitation, whereas within the accuracies quoted there is no such trend in the data of Netter et al. In this latter work the authors employed photographic emulsions and counters. In the emulsion work the number of fissions occurring was determined by an indirect and complex method requiring the counting of fission-neutron-produced proton recoils in the emulsion. From their counter work, which required a twofold coincidence between a fission fragment and a long-range particle, they estimate that there is one long-range particle per 1500 fissions for 300-kev neutron-induced fission of Pu^{239} , a value which is a factor of 3 or 4 less than that reported by other authors. They were not attempting to make absolute measurements of the number of long-range particles emitted in fission but only measurements of the relative numbers as a function of neutron energy; nevertheless, in view of the large discrepancy in the absolute measurement and of the complexity of their emulsion work, it is possible that the actual errors in their relative measurements are considerably greater than those quoted.

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Dmitriev et al.⁽⁵¹⁾ have recently reported a measurement of the ratios of the probabilities of triple fission for slow-neutron-induced fission of U^{233} , Pu^{239} , and U^{235} , giving values that are in good agreement with those of the present work. Table 7 gives the results of these probability ratio measurements.

TABLE 7
RATIOS OF PROBABILITY OF LONG-RANGE PARTICLE
EMISSION IN SLOW-NEUTRON-INDUCED FISSION

	Present Data	Dmitriev <u>et al.</u> ^b
$\eta(U^{233})/\eta(U^{235})^a$	1.21 ± 0.10	1.22 ± 0.06
$\eta(Pu^{239})/\eta(U^{235})$	1.21 ± 0.10	1.18 ± 0.06

^aWhere η is the probability of long-range-particle emission (i.e., number of long-range particles per fission).

^bSee reference (51).

The T/B ratio of $(3.897 \pm 0.26) \times 10^{-3}$ observed for spontaneous fission of Cm^{242} is about two standard deviations off from the value expected from an interpolation of the values of the ratios observed for the neighboring nuclides Cf^{252} and Cm^{244} , which differ from it in X value by only 0.08% and 0.8% respectively (see Table 5 or Fig. 18). The statistical chance of being off by two standard deviations is approximately 1/21. It is interesting to note that Perfilov et al.⁽⁵²⁾ have recently reported a measurement giving $(3.3 \pm 0.39) \times 10^{-3}$ alphas per fission for Cm^{242} , a value which fits perfectly the systematics of the

data presented in Fig. 18. A careful re-examination of the present data which gives the apparently anomalous value for Cm^{242} failed to disclose any obvious source of error.

At the present time there is no adequate theory to explain either the variation from nuclide to nuclide of the probability of emission of long-range particles or the variation with excitation energy in a given nuclide.

In his work on binary and ternary fission of heavy elements,⁽³⁾ Tsien speculates on an explanation for the decrease in the probability of the emission of long-range particles with increasing excitation energy. His speculation proceeds as follows. A third particle in fission must be formed off-axis from the large fragments if it is to be emitted with the energy characteristic of a long-range particle. If it were produced on-axis the resultant Coulomb force would be zero and there would be no acceleration, hence the particle would be undetected because it would have very low energy. When a nucleus is excited to an energy above the fission threshold (e.g., by neutron capture) it will exist for a certain interval of time during which the nuclear constituents become arranged in a configuration such that fission will finally become possible. This interval of time, which is called the mean life of the compound nucleus or fission time, is not well known but is estimated to be of the order of 10^{-14} sec. In general this mean life decreases with increasing excitation energy. A nucleus excited to an energy many million electron volts above the fission threshold would have a relatively

short mean life and would rapidly pass through various states of deformation until reaching a configuration favorable for fission. Tsien tacitly assumes that axially asymmetric deformations require a longer time to develop than axially symmetric ones. If a third particle were produced in the fission of a highly excited nucleus it would have a high probability of being on-axis or collinear with the large fragments and hence of receiving little kinetic energy. On the other hand a nucleus excited to an energy just slightly above the fission threshold would have a relatively long mean life, the nuclear constituents requiring a longer time to become oriented into a configuration favorable for fission. Consequently, such a nucleus would pass through various states of deformation relatively slowly, and if it should achieve a configuration having an incipient central drop there would be a certain probability that the drop under the effect of the asymmetry of the two large fragments or of the strong vibration of the nuclear matter would orient itself away from the axis of deformation in order to reach a configuration favorable to the instability of the nucleus. A particle produced in such a process would have a high probability of being off-axis, hence of experiencing a net Coulomb force and acquiring an energy characteristic of a long-range particle.

This mechanism explains in a qualitative way the observed trend of increasing probability of emission with decreasing excitation energy. However, it does not explain the increased probability of emission with increasing Z^2/A values. Increasing Z^2/A corresponds to increasing

excitation energy in the sense that the height of the fission barrier generally decreases as Z^2/A increases.⁽⁵³⁾ Or from another aspect one notes that the Bohr-Wheeler saddle configuration progresses from a dumb-bell shape toward a spherical shape as Z^2/A increases.⁽⁴⁹⁾ This means that a nucleus with a higher Z^2/A value can more easily assume the critical shape required for fission and would be expected to assume this configuration more rapidly, thus allowing less time for the development of axially asymmetric deformations; hence, according to Tsien's hypothesis, it would be expected to emit relatively fewer long-range particles per fission, whereas according to observations a greater number are actually emitted.

In general, evaporation and barrier penetration models used to explain nuclear particle emission predict enhanced emission as the available energy increases. Hence it is reasonable to suppose that the mechanism by which long-range particles are emitted is one which is enhanced for nuclei with higher Z^2/A values (i.e., with higher available energy at scission). However, such a supposition also leads to the expectation of increased probability of emission with increasing excitation energy from a given nucleus, whereas the probability is actually observed to decrease.

Indeed, increasing probability of emission of long-range particles per fission with increasing Z^2/A appears inconsistent with decreasing probability of emission with increasing excitation energy when viewed from the standpoint either of available energy or of fission time.

It does not seem likely that spin differences of a few units, such as those encountered in spontaneous fission versus thermal-neutron-induced fission of Pu^{240} and Pu^{242} , would play an important role in determining the relative number of long-range particles emitted. The spin energy difference for an even-even nucleus in the ground state with respect to a 2^+ spin state is about 0.1 Mev, and about 1 Mev with respect to a 1^+ or 3^+ spin state. In general, spin energy differences for the ground state with respect to higher spin states of either parity are of the order of 1 Mev or less,⁽⁵⁴⁾ whereas the excitation energy from neutron capture is considerably greater (i.e., ≈ 6 Mev plus the KE of the neutron).

Neither the barrier penetration model^(55,56) nor the evaporation model⁽⁵⁷⁾ for nuclear particle emission is applicable to long-range particle emission in fission for the following reasons. The semiclassical theory of radioactive alpha-particle emission by barrier penetration involves the rate at which the alpha particles hit the barrier multiplied by the transparency of the barrier. This theory predicts half-lives for the process of the order of thousands of years. But the particles emitted in fission, most of which are alphas, are emitted at the time of scission, as is evidenced by their angular distribution. It is not obvious what time interval, Δt , is relevant to the emission of particles from fission; it is not simply the mean lifetime of the compound nucleus ($\approx 10^{-14}$ sec), a concept which, at any rate, is not applicable to spontaneous fission. Rather, Δt is associated with the time required for a

It was not until 1934 that the existence of a low energy state was demonstrated in spontaneous fission versus thermal-neutron induced fission of ^{235}U and ^{239}Pu , which also had important role in determining the relative number of long range particles emitted. The high energy fission for an equilibrium nucleus in the ground state with respect to a γ spin state is about 0.1 Mev, and about 1 Mev with respect to a π or σ spin state. In general, high energy fission for the ground state will respect to about spin states of either parity and of the order of 1 Mev or less. (24) Between the excitation energy from neutron capture to a completely excited fission, ~ 6 Mev plus the 1 Mev of the neutron, the process of fission is not a simple one. (25) The process of fission is not a simple one. (26) The process of fission is not a simple one. (27) The process of fission is not a simple one. (28) The process of fission is not a simple one. (29) The process of fission is not a simple one. (30) The process of fission is not a simple one. (31) The process of fission is not a simple one. (32) The process of fission is not a simple one. (33) The process of fission is not a simple one. (34) The process of fission is not a simple one. (35) The process of fission is not a simple one. (36) The process of fission is not a simple one. (37) The process of fission is not a simple one. (38) The process of fission is not a simple one. (39) The process of fission is not a simple one. (40) The process of fission is not a simple one. (41) The process of fission is not a simple one. (42) The process of fission is not a simple one. (43) The process of fission is not a simple one. (44) The process of fission is not a simple one. (45) The process of fission is not a simple one. (46) The process of fission is not a simple one. (47) The process of fission is not a simple one. (48) The process of fission is not a simple one. (49) The process of fission is not a simple one. (50) The process of fission is not a simple one. (51) The process of fission is not a simple one. (52) The process of fission is not a simple one. (53) The process of fission is not a simple one. (54) The process of fission is not a simple one. (55) The process of fission is not a simple one. (56) The process of fission is not a simple one. (57) The process of fission is not a simple one. (58) The process of fission is not a simple one. (59) The process of fission is not a simple one. (60) The process of fission is not a simple one. (61) The process of fission is not a simple one. (62) The process of fission is not a simple one. (63) The process of fission is not a simple one. (64) The process of fission is not a simple one. (65) The process of fission is not a simple one. (66) The process of fission is not a simple one. (67) The process of fission is not a simple one. (68) The process of fission is not a simple one. (69) The process of fission is not a simple one. (70) The process of fission is not a simple one. (71) The process of fission is not a simple one. (72) The process of fission is not a simple one. (73) The process of fission is not a simple one. (74) The process of fission is not a simple one. (75) The process of fission is not a simple one. (76) The process of fission is not a simple one. (77) The process of fission is not a simple one. (78) The process of fission is not a simple one. (79) The process of fission is not a simple one. (80) The process of fission is not a simple one. (81) The process of fission is not a simple one. (82) The process of fission is not a simple one. (83) The process of fission is not a simple one. (84) The process of fission is not a simple one. (85) The process of fission is not a simple one. (86) The process of fission is not a simple one. (87) The process of fission is not a simple one. (88) The process of fission is not a simple one. (89) The process of fission is not a simple one. (90) The process of fission is not a simple one. (91) The process of fission is not a simple one. (92) The process of fission is not a simple one. (93) The process of fission is not a simple one. (94) The process of fission is not a simple one. (95) The process of fission is not a simple one. (96) The process of fission is not a simple one. (97) The process of fission is not a simple one. (98) The process of fission is not a simple one. (99) The process of fission is not a simple one. (100) The process of fission is not a simple one.

nucleus to pass from the Bohr-Wheeler saddle point configuration to the scission or fragment separation point. This time is estimated to be of the order of several nuclear transit times (i.e., $\Delta t \approx 10^{-21}$ sec). The barrier transparency and the alpha particle's "free energy" in a nucleus near the scission point are not expected to be greatly different from those of a normal nucleus, hence there is not enough time for alpha particle emission by barrier penetration, by many orders of magnitude. The possibility that the particles are produced by nuclear evaporation from the original nucleus or from the fragments immediately after scission is also unlikely. An isotropic distribution would be expected from an evaporation process, whereas the long-range particles are actually strongly peaked at $\approx 90^\circ$ to the direction of the fragments.

Halpern has proposed a theory for the production mechanism for the long-range particles based on the so-called "sudden approximation," or non-adiabatic nuclear potential change.⁽⁵⁸⁾ In this theory the alpha particle is originally held in a potential well shown at (A), Fig. 19. At scission the potential changes to (B), Fig. 19. This change is assumed to occur in a time short compared to the alpha particle's nuclear transit time; a calculation is made of the probability that the alpha particle will be trapped in region (b) after scission. Preliminary calculations indicate results of the correct order of magnitude. However, the radii of the fragments vary approximately as $A^{1/3}$ of the fissioning nucleus and there is evidence that the breaking distance δ varies in a like manner,⁽⁵⁹⁾ and E_c , the height of the barrier, varies

Fig. 19.--"Sudden approximation" theory potential distribution.

(A) shows the original potential well, $R_0 = r_0 A^{1/3}$ cm, $r_0 \approx 1.5 \times 10^{-13}$ cm, $E_c \approx 28$ Mev, the height of the Coulomb barrier, and the broken line in the well indicates the alpha-particle binding energy. (B) shows the potential distribution immediately after scission, $R_1 \approx R_2 \approx r_0 (A/2)^{1/3}$ cm, and δ is the breaking distance (i.e., the distance between the fragment mass centers at scission).

as $Z/A^{1/3}$. To a first approximation these quantities can be considered constant over the range of nuclei considered in this work. Hence the theory in its present state of development does not appear to involve the proper parameters to allow a prediction of the variation of the probability of particle emission for different nuclides and excitation energies.

Fuller has worked out in greater detail a similar theory on the effect of non-adiabatic potential changes on neutron production in fission.⁽⁶⁰⁾ He found that with a reasonable choice of the parameters involved in his theory he could fit the data of Bowman and Thompson⁽⁶¹⁾ on the number of "scission neutrons" produced in spontaneous fission of Cr^{252} .

Fig. 1. (A) -- original potential energy; (B) -- potential energy after a change in the parameters.

(A) shows the original potential well, $V_0 = 2.5$, $\delta = 1$, $\epsilon = 0.5 \times 10^{-10}$ cm, $E_0 = 0.5$ eV. The height of the Coulomb barrier and the broken line in the well indicates the eigen-parabolic binding energy. (B) shows the potential distribution immediately after excitation, $E_0 = E_0 + \epsilon = 0.5 + 0.5 \times 10^{-10}$ eV, and δ is the breaking distance (i.e., the distance between the fragment ions centers at scission).

as E/E_0 . To a first approximation these quantities can be considered constant over the range of nuclei considered in this work. Hence the theory is its greatest range of application does not appear to include the proper parameters to allow a prediction of the variation of the probability of particle ejection for different nuclei and excitation energies.

Further has worked out in greater detail a similar theory on the effect of non-adiabatic potential changes on nuclear production in fission. He found that with a reasonable choice of the parameters involved in his theory he could fit the data of Barnes and Johnson on the number of "soliton neutrons" produced in spontaneous fission of

The principal objection to the sudden approximation theory for the production of charged particles in fission is that it assumes the existence in the nucleus of preformed particles of the type emitted. In the case of alpha particles, for instance, this assumption is untenable⁽⁶²⁾ in light of present knowledge of the structure of the nucleus. Nevertheless, a further development of the theory might be of interest.

The historical objection to the modern interpretation of the

production of charged particles in liquids is that it is based on the

assumption that the ionization of the liquid is the only source of

ions of high energy, for instance, this assumption is not valid

in the case of organic liquids, the structure of the molecule being

such that a further development of the theory might be expected.

APPENDIX A

SOURCE MOUNTING TECHNIQUES

The source deposits used in this work were made by the well-known techniques of vacuum evaporation or brush painting or by the techniques of electrospray-deposition⁽⁴¹⁾ or autodeposition⁽²⁵⁾ which have been described elsewhere in the literature. The support material used for all sources was nickel foil⁽²⁶⁾ of thickness 5×10^{-6} in. ($\approx 100 \mu\text{g}/\text{cm}^2$). This electrodeposited nickel was supplied with a copper backing which was dissolved away after the foil was mounted and ready for use. A detailed description of the foil and technique for removing the copper backing (stripping) is given by Bashkin and Goldhaber.⁽⁶³⁾ Their stripping solution recipe is as follows: Add 15 cc concentrated H_2SO_4 to one liter saturated solution of chromic acid (CrO_3). Immerse foil for five to ten minutes until copper is dissolved. (An alternative stripping solution which also works well is composed of 50 gm of trichloroacetic acid, 250 ml NH_4OH , and 250 ml H_2O .)

Foil holders consisted of a pair of 1-inch-aperture rings made of 22-gage (1/32-in.) stainless steel. Mating pairs had a 0.010-in. offset, as shown in Fig. 20, which stretched and clamped the foil when the rings were assembled. The rings were held together by eight #00-90 flat-head stainless steel machine screws. A $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil unsupported over a circular area of 1-in. diameter proved to be exceedingly fragile. To overcome this difficulty the following techniques were developed.

APPENDIX A

APPENDIX A

The source reports used in this work were made by the well-known techniques of vacuum deposition of various patterns by the techniques of electrolytic deposition (14) or anodization (15) with the use of described elsewhere in the literature. The support material used for all sources was nickel foil (20) of thickness 5×10^{-3} in. (0.100 mm). The electrochromic source was anodized with a copper backing which was dissolved away after the foil was mounted and ready for use. A detailed description of the foil and technique for removing the copper backing (stripping) is given by Johnson and Gahleitner (21). Their striping solution recipe is as follows: Add 15 cc concentrated H₂SO₄ to one liter saturated solution of ammonium persulfate (APS) - transfer foil for five to ten minutes until copper is dissolved. An alternative striping solution which also works well is composed of 50 gm of potassium dichromate (K₂Cr₂O₇) in 100 ml H₂SO₄ (20% in H₂O) and 500 ml H₂O. The foil holders consisted of a pair of brass electrodes spaced 0.5 in. apart. The foil was held in a 0.5 in. slot, as shown in Fig. 20, which supported and clamped the foil when the strips were assembled. The strips were held together by a pair of brass rods, as shown in Fig. 20, which supported and clamped the foil when the strips were assembled. The strips were held together by a pair of brass rods, as shown in Fig. 20, which supported and clamped the foil when the strips were assembled. A 100- μ m nickel foil was supported over a circular area of 1-in. diameter, grown to be approximately 1-in. in diameter with only the following conditions were developed.

Fig. 20.--Foil-stripping apparatus and cross-section view of mounted foil. (1) Stripping mask. (2) $100\text{-}\mu\text{g}/\text{cm}^2$ nickel foil. (3) Foil holder. (4) Water reservoir, inner. (5) Stripping fixture. (6) Water reservoir, outer. (7) Water level after completion of stripping process. (8) Unstripped copper backing. (9) Stripped foil. (10) Source deposit.

EXPLODED SECTION VIEW STRIPPING APPARATUS

ASSEMBLY SECTION VIEW SHOWING OUTER RESERVOIR

ENLARGED SECTION VIEW FOIL AND HOLDER ASSEMBLY

EXPLODED SECTION VIEW
STRIPING APPARATUS

ASSEMBLY SECTION VIEW
SHOWING OUTER RESERVOIR

ENLARGED SECTION VIEW
FOIL AND HOLDER ASSEMBLY

The first, which was useful for electrosprayed and autodeposited sources, consisted of clamping together in a holder a 20-mesh, 93%-transparent nickel screen⁽²⁷⁾ and an unstripped foil so that the screen was in contact with the copper. After stripping the foil usually adhered to the screen, giving an assembly of much greater strength than the foil alone. The deposit was then applied to the clear side of the foil. In the second technique the source deposit was applied to the nickel side of the unstripped foil and the copper then removed only from the region directly behind the deposit. This method was useful for painted and evaporated sources, as these processes were too severe for prestripped foil, even for one supported by an electromesh screen. This selective stripping technique is illustrated in Fig. 20 and consisted of placing the foil with the source side down on a water-filled reservoir in a stainless steel stripping fixture. A brass stripping mask was then placed on the top or copper side of the foil. These masks were 1 in. o.d., $\approx 1/32$ in. thick, and had an inner diameter to suit the size of the source deposit. Stripping solution was then dropped onto the foil until the exposed copper area was covered. After about five minutes the depleted solution was removed by absorption into cleansing tissue and fresh solution was added. This process was repeated as necessary. After all exposed copper was dissolved the stripped area was irrigated with a few drops of water and then the water level in the outer reservoir was raised to immerse the entire foil and holder assembly. At this point the stripping mask was lifted off and the foil removed from the stripping fixture and rinsed in distilled water for about twelve hours to remove

The first trial was made for electroplating and anode reaction. The solution of copper was then in a holder a 10-inch, 100-gram. Nickel wire, and an electrode foil of 10-inch was in the bath with the copper. After stripping the foil, usually related to the bath, giving an assembly of nickel electrode, strip, and the foil, and the electrode was then applied to the back side of the foil. In the second electrode, the copper deposit was applied to the back side of the electrode foil and the copper then removed only from the back side of the foil. The deposit was made for nickel and electrode. As these processes were the reverse for nickel, the foil was for one deposited by an electrode reaction. This electrode stripping technique is illustrated in Fig. 10 and consisted of placing the foil with the source and on a water-filled reservoir in a container. The electrode stripping was then placed on the top of the water level of the foil. These were in 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, 5.0, 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, 8.5, 9.0, 9.5, 10.0, 10.5, 11.0, 11.5, 12.0, 12.5, 13.0, 13.5, 14.0, 14.5, 15.0, 15.5, 16.0, 16.5, 17.0, 17.5, 18.0, 18.5, 19.0, 19.5, 20.0, 20.5, 21.0, 21.5, 22.0, 22.5, 23.0, 23.5, 24.0, 24.5, 25.0, 25.5, 26.0, 26.5, 27.0, 27.5, 28.0, 28.5, 29.0, 29.5, 30.0, 30.5, 31.0, 31.5, 32.0, 32.5, 33.0, 33.5, 34.0, 34.5, 35.0, 35.5, 36.0, 36.5, 37.0, 37.5, 38.0, 38.5, 39.0, 39.5, 40.0, 40.5, 41.0, 41.5, 42.0, 42.5, 43.0, 43.5, 44.0, 44.5, 45.0, 45.5, 46.0, 46.5, 47.0, 47.5, 48.0, 48.5, 49.0, 49.5, 50.0, 50.5, 51.0, 51.5, 52.0, 52.5, 53.0, 53.5, 54.0, 54.5, 55.0, 55.5, 56.0, 56.5, 57.0, 57.5, 58.0, 58.5, 59.0, 59.5, 60.0, 60.5, 61.0, 61.5, 62.0, 62.5, 63.0, 63.5, 64.0, 64.5, 65.0, 65.5, 66.0, 66.5, 67.0, 67.5, 68.0, 68.5, 69.0, 69.5, 70.0, 70.5, 71.0, 71.5, 72.0, 72.5, 73.0, 73.5, 74.0, 74.5, 75.0, 75.5, 76.0, 76.5, 77.0, 77.5, 78.0, 78.5, 79.0, 79.5, 80.0, 80.5, 81.0, 81.5, 82.0, 82.5, 83.0, 83.5, 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801.0, 801.5, 802.0, 802.5, 803.0, 803.5, 804.0, 804.5, 805.0, 805.5, 806.0, 806.5, 807.0, 807.5, 808.0, 808.5, 809.0, 809.5, 810.0, 810.5, 811.0, 811.5, 812.0, 812.5, 813.0, 813.5, 814.0, 814.5, 815.0, 815.5, 816.0, 816.5, 817.0, 817.5, 818.0, 818.5, 819.0, 819.5, 820.0, 820.5, 821.0, 821.5, 822.0, 822.5, 823.0, 823.5, 824.0, 824.5, 825.0, 825.5, 826.0, 826.5, 827.0, 827.5, 828.0, 828.5, 829.0, 829.5, 830.0, 830.5, 831.0, 831.5, 832.0, 832.5, 833.0, 833.5, 834.0, 834.5, 835.0, 835.5, 836.0, 836.5, 837.0, 837.5, 838.0, 838.5, 839.0, 839.5, 840.0, 840.5, 841.0, 841.5, 842.0, 842.5, 843.0, 843.5, 844.0, 844.5, 845.0, 845.5, 846.0, 846.5, 847.0, 847.5, 848.0, 848.5, 849.0, 849.5, 850.0, 850.5, 851.0, 851.5, 852.0, 852.5, 853.0, 853.5, 854.0, 854.5, 855.0, 855.5, 856.0, 856.5, 857.0, 857.5, 858.0, 858.5, 859.0, 859.5, 860.0, 860.5, 861.0, 861.5, 862.0, 862.5, 863.0, 863.5, 864.0, 864.5, 865.0, 865.5, 866.0, 866.5, 867.0, 867.5, 868.0, 868.5, 869.0, 869.5, 870.0, 870.5, 871.0, 871.5, 872.0, 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residual stripping solution. A few drops of Aerosol OT were usually added to the water in the outer reservoir to reduce the surface-tension-produced strain on the stripped foil. This strain was most severe just as the water was raised to the level of the foil. Brass proved to be a better material for stripping masks than stainless steel, probably because the brass helped deplete the solution which penetrated between the mask and foil and retarded the stripping action in that region.

This selective stripping technique is useful only for water-insoluble source deposits.

APPENDIX B

FISSION-SOURCE FRAGMENT SELF-ABSORPTION CALCULATIONS

In order for long-range particles to be detected by the ionization chamber apparatus described in Chapters II and III, they must be associated with fission events for which both fragments emerge from the source and support foil with an energy at least equal to the bias setting of the fission counters. The present calculation gives an approximation for the detection probability, that is, for the ratio of the number of long-range particles which are detected to the total number emitted. In Fig. 21, which is relevant to a "thin" source on a support foil of thickness t , the z -axis is taken along the direction of the long-range particle having an angle θ with respect to the normal N . β_1 and β_2 are the average angles made by the direction of the light and heavy fragments, respectively, with the z -axis. The y -axis is taken to lie in the plane of the top surface of the foil (i.e., in the source plane), and ϕ is the polar angle of the fragments about the z -axis.

The path of fragments associated with a long-range particle having an angle θ lies on cones whose axis is along the z -direction and which have a half-angle or supplementary half-angle of β_1 or β_2 .

The equations of these cones are

$$x = R \sin\beta_1 \cos\phi$$

$$y = R \sin\beta_1 \sin\phi$$

$$z = R \cos\beta_1$$

Fig. 1. - Isometric view of a system of square supports.

and

where α is the angle

The angle of the beam is

where β is the angle

that the angle of the intersection of the beam with the beam

will be

where γ is the angle

where δ is the angle

where R is the path length of the fragment in the foil. The curve labeled "a" in Fig. 21 shows the intersection of the light fragment cone with the lower foil surface. Now let R_1 and R_2 be the maximum range the fragments can spend in the foil and still emerge with a remaining energy equal to the bias energy of the fission counters. Then

$$\phi_{\beta_1} = \arccos \frac{\frac{t}{R_1} + \cos\beta_1 \cos\theta}{\sin\beta_1 \sin\theta}$$

and

$$\phi_{\beta_2} = \arccos \frac{\frac{t}{R_2} + \cos\beta_2 \cos\theta}{\sin\beta_2 \sin\theta}$$

where ϕ_{β_1} and ϕ_{β_2} are the angles associated with these maximum range fragments.

The equation of the top surface of the foil is

$$x \sin\theta - z \cos\theta = 0.$$

Hence, the equations of the intersections of the fragment cones with the top foil surface are

$$\sin\beta_1 \cos\phi \sin\theta - \cos\beta_1 \cos\theta = 0$$

and

$$\sin\beta_2 \cos\phi \sin\theta - \cos\beta_2 \cos\theta = 0,$$

or

$$\phi_{T\beta_1} = \arccos \tan\beta_1 \tan\theta$$

and

$$\phi_{T\beta_2} = \arccos \tan\beta_2 \tan\theta,$$

where R is the radius of the fragment in the foil. The curve
labeled "a" in fig. 21 shows the intersection of the fragment with
the foil surface. The line R_0 and R_1 are the maximum range and
the range can be found in the foil and still change with a varying energy
equal to the loss energy of the fragment nucleus. Then

$$R_0 = \frac{1}{2} \frac{1 + \cos \theta}{\sin \theta} \frac{1}{\sin \theta} + \cos \theta$$

and

$$R_1 = \frac{1}{2} \frac{1 + \cos \theta}{\sin \theta} \frac{1}{\sin \theta} + \cos \theta$$

where R_0 and R_1 are the ranges associated with these maximum range
fragments.
The equation of the foil surface of the foil is

$$x^2 + y^2 + z^2 = R^2$$

hence, the equation of the intersection of the fragment core with the
foil surface is

$$x^2 + y^2 + z^2 = R^2$$

and

$$x^2 + y^2 + z^2 = R^2$$

or

$$x^2 + y^2 + z^2 = R^2$$

and

$$x^2 + y^2 + z^2 = R^2$$

where $\phi_{T\beta_1}$ and $\phi_{T\beta_2}$ are the angles associated with fragments that lie in the top foil surface. Thus, $2(\phi_{T\beta_1} - \phi_{O\beta_1})$ is the total ϕ -angle over which the β_1 (light) fragment is effectively absorbed in the foil, and $2(\phi_{T\beta_2} - \phi_{O\beta_2})$ the angle over which the β_2 (heavy) fragment is absorbed. Hence, the probability that a long-range particle with an angle θ will be detected is

$$P_0 = \frac{2\pi - 2[(\phi_{T\beta_1} - \phi_{O\beta_1}) + (\phi_{T\beta_2} - \phi_{O\beta_2})]}{2\pi}. \quad (4)$$

This probability function must be integrated over the solid angle subtended by the entrance window of the long-range particle counter. Thus

$$P_1 = \frac{1}{K_1} = \frac{\int_{\theta_1}^{\pi} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} 2\pi \sin\theta d\theta}{2\pi} \quad (5)$$

$$\approx \int_{\theta_1}^{\theta_2} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} \sin\theta d\theta$$

and

$$P_2 = \frac{1}{K_2} = \frac{\int_0^{\theta_2} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} 2\pi \sin\theta d\theta}{2\pi} \quad (6)$$

$$= \int_0^{\theta_2} \frac{[\pi - (\phi_{T\beta_1} - \phi_{C,\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} \sin\theta d\theta$$

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of the probable errors in the range, range-energy and fragment direction data gives an estimate of $\pm 1\%$ for the overall accuracy of the results of these calculations.

In general, the effects of non-homogeneous source deposits are not believed to be a significant source of error in this work.

of the probable errors in the range, range-energy and frequency distribution data gives an estimate of 15% for the overall accuracy of the results of these calculations.

In general, the effects of non-homogeneous source deposits are not believed to be a significant source of error in this work.

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Fig. 21. Enlarged view of a section of source support foil.

21

$x = 0$ and $y = 0$

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where R is the path length of the fragment in the foil. The curve labeled "a" in Fig. 21 shows the intersection of the light fragment cone with the lower foil surface. Now let R_1 and R_2 be the maximum range the fragments can spend in the foil and still emerge with a remaining energy equal to the bias energy of the fission counters. Then

$$\phi_{\beta_1} = \arccos \frac{\frac{t}{R_1} + \cos\beta_1 \cos\theta}{\sin\beta_1 \sin\theta}$$

and

$$\phi_{\beta_2} = \arccos \frac{\frac{t}{R_2} + \cos\beta_2 \cos\theta}{\sin\beta_2 \sin\theta}$$

where ϕ_{β_1} and ϕ_{β_2} are the angles associated with these maximum range fragments.

The equation of the top surface of the foil is

$$x \sin\theta - z \cos\theta = 0.$$

Hence, the equations of the intersections of the fragment cones with the top foil surface are

$$\sin\beta_1 \cos\phi \sin\theta - \cos\beta_1 \cos\theta = 0$$

and

$$\sin\beta_2 \cos\phi \sin\theta - \cos\beta_2 \cos\theta = 0,$$

or

$$\phi_{T\beta_1} = \arccos \tan\beta_1 \tan\theta$$

and

$$\phi_{T\beta_2} = \arccos \tan\beta_2 \tan\theta,$$

where θ is the half angle of the fragment in the foil. The curve labeled "a" in Fig. 21 shows the intersection of the light fragment cone with the lower foil surface. Now let θ_1 and θ_2 be the maximum range the fragments can spread in the foil and still emerge with a remaining energy equal to the kinetic energy of the fission neutrons. Then

$$W_{01} = \frac{1}{2} m v_0^2 = \frac{1}{2} m v_0^2 \cos^2 \theta_1 + \frac{1}{2} m v_0^2 \sin^2 \theta_1$$

$$W_{02} = \frac{1}{2} m v_0^2 = \frac{1}{2} m v_0^2 \cos^2 \theta_2 + \frac{1}{2} m v_0^2 \sin^2 \theta_2$$

where θ_1 and θ_2 are the angles associated with these maximum ranges. The angle θ_1 is the angle of the new surface of the foil is

$$\theta_1 = \theta_2 = \theta = \theta_0 = 0$$

Thus, the equations of the intersection of the fragment cones with the foil surface are

$$x = v_0 t \cos \theta_1, \quad y = v_0 t \sin \theta_1$$

$$x = v_0 t \cos \theta_2, \quad y = v_0 t \sin \theta_2$$

$$x = v_0 t \cos \theta, \quad y = v_0 t \sin \theta$$

$$x = v_0 t \cos \theta, \quad y = v_0 t \sin \theta$$

where $\phi_{T\beta_1}$ and $\phi_{T\beta_2}$ are the angles associated with fragments that lie in the top foil surface. Thus, $2(\phi_{T\beta_1} - \phi_{O\beta_1})$ is the total ϕ -angle over which the β_1 (light) fragment is effectively absorbed in the foil, and $2(\phi_{T\beta_2} - \phi_{O\beta_2})$ the angle over which the β_2 (heavy) fragment is absorbed. Hence, the probability that a long-range particle with an angle θ will be detected is

$$P_0 = \frac{2\pi - 2[(\phi_{T\beta_1} - \phi_{O\beta_1}) + (\phi_{T\beta_2} - \phi_{O\beta_2})]}{2\pi}. \quad (4)$$

This probability function must be integrated over the solid angle subtended by the entrance window of the long-range particle counter. Thus

$$\begin{aligned} P_1 = \frac{1}{K_1} &= \frac{\int_{\theta_1}^{\pi} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} 2\pi \sin\theta d\theta}{2\pi} \\ &\approx \int_0^{\theta_1} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} \sin\theta d\theta \end{aligned} \quad (5)$$

and

$$\begin{aligned} P_2 = \frac{1}{K_2} &= \frac{\int_0^{\theta_2} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} 2\pi \sin\theta d\theta}{2\pi} \\ &= \int_0^{\theta_2} \frac{[\pi - (\phi_{T\beta_1} - \phi_{O\beta_1}) - (\phi_{T\beta_2} - \phi_{O\beta_2})]}{\pi} \sin\theta d\theta \end{aligned} \quad (6)$$

where θ_1 and θ_2 are the angles associated with the points A_1 and A_2 on the top cell surface. Thus, $\theta_1 = \theta_2 = \theta$ is the total θ -angle over which the H (light) fragment is effectively absorbed in the foil, and $\theta_1 = \theta_2 = \theta$ the angle over which the H (heavy) fragment is absorbed. Hence, the probability that a long-range particle with an angle θ will be detected is

$$P(\theta) = \frac{2\pi \int_0^{\theta} \sin \theta' d\theta' \cdot \left[\frac{1}{2} (1 - \cos \theta') \right]}{2\pi \int_0^{\theta} \sin \theta' d\theta'}$$

This probability function must be integrated over the solid angle subtended by the entrance window of the long-range particle counter. Thus

$$P(\theta) = \frac{2\pi \int_0^{\theta} \sin \theta' d\theta' \cdot \left[\frac{1}{2} (1 - \cos \theta') \right]}{2\pi \int_0^{\theta} \sin \theta' d\theta'}$$

$$P(\theta) = \frac{2\pi \int_0^{\theta} \sin \theta' d\theta' \cdot \left[\frac{1}{2} (1 - \cos \theta') \right]}{2\pi \int_0^{\theta} \sin \theta' d\theta'}$$

$$P(\theta) = \frac{2\pi \int_0^{\theta} \sin \theta' d\theta' \cdot \left[\frac{1}{2} (1 - \cos \theta') \right]}{2\pi \int_0^{\theta} \sin \theta' d\theta'}$$

$$P(\theta) = \frac{2\pi \int_0^{\theta} \sin \theta' d\theta' \cdot \left[\frac{1}{2} (1 - \cos \theta') \right]}{2\pi \int_0^{\theta} \sin \theta' d\theta'}$$

$$P(\theta) = \frac{2\pi \int_0^{\theta} \sin \theta' d\theta' \cdot \left[\frac{1}{2} (1 - \cos \theta') \right]}{2\pi \int_0^{\theta} \sin \theta' d\theta'}$$

where θ_1 and θ_2 are the half-angles subtended by the entrance windows of the long-range particle chambers at the source. P_1 is the probability that a long-range particle emitted in the lower hemisphere will be detected and P_2 the detection probability for a particle emitted in the upper hemisphere. K_1 and K_2 are the correction factors used in Chapters II and III.

The range of the heavy fragment is less than that of the light fragment, hence, in the cases where a source cover foil was used, Eq. (4) was replaced by

$$P_0 = \frac{\pi - 2(\phi_{TB2} - \phi_{0\beta_2})}{\pi}$$

and Eqs. (5) and (6) changed accordingly.

In applying the above formulation to "non-thin" fission sources, the effective total thickness t was taken to be that of the nickel support foil plus one-half the nickel equivalent of the source thickness. Ranges of fission fragments were estimated from the "most probable" fragment energy data given by Hyde⁽⁶⁴⁾ and from the range-energy data of Fulmer.⁽⁶⁵⁾ Values for β_1 and β_2 were taken from Muga et al.⁽³¹⁾

Equations (5) and (6) were integrated numerically with the aid of an IBM-704 electronic computer; the values thus obtained for K_1 and K_2 are given in Chapters II and III under the description of the measurement of the T/B ratio for each fission source employed. Assuming a homogeneous source deposit, an analysis of the errors involved in the numerical integration plus a consideration of the effects on K_1 and K_2

where δ_1 and δ_2 are the half-angles subtended by the foil in the detector at the source. E_1 is the probability that a low-energy particle emitted in the lower hemisphere will be detected and E_2 the detection probability for a particle emitted in the upper hemisphere. K_1 and K_2 are the correction factors in (1) and (2).

Let I_1 and I_2

the range of the heavy fragments be less than that of the light fragments, then in the case where a narrow cover foil was used, I_1 was registered by

$$I_1 = \frac{N_1 - N_2}{K_1 - K_2}$$

and E_1 , E_2 and K_1 , K_2 changed accordingly. In applying the above formulation to "non-thin" fission sources the effective total thickness t was taken to be that of the thickest foil plus one-half the thickest equivalent of the source thickness. Ranges of fission fragments were estimated from the "most probable" fragment energy data given by Ryle (1961) and from the range-energy data of Ryle (1961). Values for E_1 and E_2 were taken from Ryle (1961).

Equations (1) and (2) were integrated numerically with the aid of an IBM-709 electronic computer; the values thus obtained for K_1 and K_2 are given in Chapters II and III under the description of the various sets of the WP ratio for each fission source employed. Assuming a background source deposit, an analysis of the errors involved in the numerical integration plus a consideration of the effects on E_1 and E_2

of the probable errors in the range, range-energy and fragment direction data gives an estimate of $\pm 1\%$ for the overall accuracy of the results of these calculations.

In general, the effects of non-homogeneous source deposits are not believed to be a significant source of error in this work.

of the possible errors in the range, range-energy and frequency distribution
data gives an estimate of 1% for the overall accuracy of the results of
these calculations.

In general, the effects of non-homogeneous source deposits are not
believed to be a significant source of error in this work.

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