$d\epsilon_\gamma$ that for the gamma-ray. The quantity in square brackets is the usual expression for an allowed beta-transition. I_0 is a constant which depends on ϵ_0 and ϵ_1 (through the matrix elements) but not on ϵ_e and ϵ_γ, so that the shapes of the spectra are given exactly by this formula. However, for a forbidden beta-part, the quantity in square brackets must be replaced by the ordinary energy-function appropriate to the degree of forbiddenness.

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The Fission of Thorium with Alpha-Particles

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The fission distribution of fission of thorium with alpha-particles of average energy 37.5 Mev has been measured by the chemical method. The distribution found shows that the characteristic dip in the fission yield mass spectrum has been raised to within a factor of two of the peaks compared to a factor of 600 in slow neutron fission of U^{235}. The raise in the dip has caused a corresponding lowering in fission yield of these elements at the peaks. The cross section for fission of thorium with 37.5-Mev alphas was found to be about 0.6 barn, and the threshold for fission was found to be 23 to 24 Mev.

I. INTRODUCTION

SOON after the discovery of fission, Meitner[1] and Bretscher and Cook[2] found differences in the decay of various chemical fractions separated from uranium irradiated with slow neutrons and thorium irradiated with fast neutrons, respectively, and suggested that a difference existed in the distribution of fission products in the two cases. In 1940, Turner[3] suggested that the distribution in various modes of fission should be investigated. The fact that elements such as tin, cadmium, palladium, and silver[4] were found in fast neutron and deuteron fission of uranium and thorium before they were found in slow neutron fission of uranium suggested that the middle region of the distribution was raised as the energy of the incident particle was increased.

Since the compound nucleus formed in the fission of thorium with alpha-particles is U^{236}, the same compound nucleus formed in the fission of U^{235} with neutrons, it is of interest to study the fission of thorium with alphas and compare the resulting distribution of fission products with that found with uranium with slow[5] and thorium with fast[6] neutrons. Any difference between the various results where the same compound nucleus is formed must be due to differences in energy content and possible differences in distribution of the nucleons in the compound nucleus at the time of fission.

II. EXPERIMENTAL METHODS

Of the various methods available for studying the fission process, the method of chemically isolating the fission products with added carrier and determining the fission yield of each isotope seemed most suitable.[7] The method of studying the energy distribution of fission fragments in an ionization chamber, as used by Jentschke[8] and

[1] L. Meitner, Nature **143**, 637 (1939).
[2] K. Bretscher and L. G. Cook, Nature **143**, 559 (1939).
[3] L. A. Turner, Rev. Mod. Phys. **12**, 9 (1940).
[4] Y. Nishina, K. Kimura, T. Yasaki, and M. Ikawa, Nature **146**, 24 (1940); Phys. Rev. **58**, 660 (1940); Phys. Rev. **59**, 323 (1941); Phys. Rev. **59**, 667 (1941).
[5] "Nucleii formed in fission," Plutonium Project, J. Am. Chem. Soc. **68**, 2411 (1946); Rev. Mod. Phys. **18**, 513 (1946).
[6] A. Turkevich, private communication. Presented at A.A.A.S. Meeting, Chicago, December, 1947.
[7] H. L. Anderson, E. Fermi, and A. V. Grosse, Phys. Rev. **59**, 52 (1941).
[8] W. Jentschke, Zeits. f. Physik **120**, 165 (1943).

Flammersfeld, Jensen and Gentner,[9] is not particularly suited for the present problem because of the difficulties inherent in using an ionization chamber in the presence of the alpha-beam.

The thorium used in the bombardment was special purity thorium metal produced by the group at the Atomic Research Institute at Iowa State College. In a long bombardment, designated as Bombardment *A*, of 3020 μah of 39-Mev helium ions (impinging energy as estimated by Dr. J. G. Hamilton) on the Crocker Laboratory cyclotron, a $3\frac{1}{8}$-inch diameter thorium backing plate $\frac{1}{8}$ inch thick was used. After bombardment a radioautograph of the disk was made to determine the active area. The activity was then removed by milling thin layers from the active area, using a milling set-up in which the millings could be quantitatively collected. Eleven layers, varying from 1 to 3 mils in thickness, were removed from this target so the fission product distribution and the excitation curve for fission could be determined. The milling removed about 95 percent of the total beta- and gamma-activity from the target. In addition to this, three shorter bombardments of 24-hour, 2-hour, and 4-hour duration, designated as bombardments *B*, *C*, and *D*, respectively, were obtained on thorium foils of thickness about 30 mg/cm^2 using the interceptor set-up of the Crocker cyclotron. In these cases it was assumed that all of the 2-cm^2 area of the interceptor received bombardment.

The technique used in determining the fission yields was the standard chemical technique as used on the Plutonium Project. The bombarded sample was dissolved in either hydrochloric or nitric acid, a small amount of ammonium fluosilicate being added as a catalyst. The solution was diluted to a known volume and stored in Lusteroid tubes inside a glass tube to prevent loss of activity by adsorption on glass and evaporation of the solution. Aliquots were taken for determination of each desired element. In each fraction, carrier was added for the element to be separated, and the element chemically separated and decontaminated from other activities. The element was then precipitated in a suitable form, weighed, and the chemical yield determined. The samples were mounted on cardboard, covered with Cellophane, and counted on the second shelf of a standard counter set-up. Corrections were made in the counting rate for coincidences, geometry, and absorption to obtain the disintegration rate of the sample. Decay curves were taken to characterize all activities, and absorption curves were taken where possible. If absorption curves could not be taken, absorber corrections were made on the basis of published absorption curves for that activity which were known to have been taken on similar equipment. In order to obtain the fission yield the activities were all extrapolated back to a given time shortly after the end of the bombardment. Since the cyclotron irradiations were irregular, in order to integrate the bombardment and correct for saturation of shorter-lived materials during the bombardment, the bombardment history was treated in small time intervals. The microampere hours of bombardment in each time interval were then allowed to "decay" to some comparison time with the half-life of the activity being considered. At the comparison time the effective irradiations from each time interval were summed to give a total effective bombardment at that time. If the increments were longer than 5 percent of the half-life of the isotope, corrections were made for saturation during each increment.

Since neither the absolute fission yield of a fission product produced in the fission of thorium with alpha-particles nor the fission cross section are known, the cross section for the formation of each isotope which is the product of the fission yield and cross section for fission has been calculated by the equation:

$$\sigma_{\mathrm{F.P.}} = \sigma_f \times \mathrm{F.Y.} = \frac{(\text{atoms F.P.})(\text{area target})}{(\text{atoms Th})(\mu\text{ah})(1.13\times10^{16})},$$

where the term (μah) refers to the effective μah of bombardment at the time of comparison, and the factor 1.13×10^{16} converts μah to the number of helium ions striking the target.

Variations in the cross section for various bombardments have not been normalized to any given value; since the values of $\sigma_f\times$F.Y. have been calculated on an absolute basis there is no *a priori* reason for normalizing the values to any given value of the barium, the element with which comparisons are usually made. Since there

[9] A. Flammersfeld, P. Jensen, and W. Gentner, Zeits. f. Physik **120**, 450 (1943).

are inaccuracies in estimating the total bombardment and the distribution of the bombardment, variations can be expected when long and short-lived periods are compared in any one bombardment. In general, the results are probably correct to within a factor of two when comparisons are made between different bombardments.

III. ISOTOPES STUDIED

In all bombardments the 300-hr. Ba^{140} was separated in order to offer a rough check on the bombardment. The chemical methods used in the isolation were modifications of those developed for the fission products by the Plutonium Project. These will be described in detail by the individual authors,[10] so only the general methods will be described here.

1. *Zinc* was separated by precipitation as $ZnHg(CNS)_4$, which was dissolved in dilute nitric acid, and mercuric and bismuth sulfide scavenging precipitations were made. Zinc sulfide was precipitated, dissolved in hydrobromic acid, and fumed twice to dryness. From a sodium hydroxide solution, ferric hydroxide and barium carbonate scavenging precipitates were made. Zinc sulfide was precipitated, dissolved, and the zinc finally precipitated as $ZnHg(SCN)_4$. The final yield of carrier added was only 10 percent, and only a small activity was found which could not be identified as the 49-hr. Zn^{72}. An initial observed activity of 250 c/m decayed with a 3-hr. period, followed by a period longer than 5 days, and could conceivably be silver contamination. An upper limit of 50 c/m of the 49-hr. period could have been present and not resolved, setting an upper limit of 8×10^{-29} for the value of $\sigma_f\times$F.Y. for mass 72.

2. *Arsenic* was separated to study the chain $Ge^{77} \xrightarrow{\text{12 hr.}} As^{77} \xrightarrow{\text{40 hr.}} Se^{77}$ (stable). The arsenic was separated about 24 hours after bombardment, so 75 percent germanium had decayed to arsenic. Since in uranium fission the independent yield of members of this chain is nearly all 40-hr. As^{77},[11] it is believed the result found represents >95 percent of the chain. Arsenic and germanium were separated by precipitation as the sulfide from 6*N* HCl. Germanium was distilled from HCl and $KClO_3$. Then arsenic was distilled as arsenic trichloride from concentrated hydrochloric acid containing cuprous chloride with antimony, tellurium, and tin as holdback carriers. Arsenic sulfide was precipitated from the distillate and redistilled. The arsenic was weighed as As_2S_3. The yield of carrier was 40 percent, and an observed activity of 50 c/m decayed to less than 5 c/m with a 40-hr. half-life. It was assumed to be all arsenic. Since no absorption curve could be taken, the activity was extrapolated to zero absorber on the basis of absorption curves of Steinberg and Engelkemeier.[11] The activity found gives a value of $\sigma_f\times$F.Y. of 2.8×10^{-29}.

3. *Bromine* was isolated by extraction with carbon tetrachloride after oxidation with permanganate. The extract was decontaminated from iodine by oxidation with sodium nitrite and extraction of the iodine with carbon tetrachloride. The bromine was then oxidized by permanganate and extracted. Two such decontamination cycles were run, and the bromine was then precipitated as silver bromide.

The 2.4-hr. Br^{83} was found and identified, it being found with a $\sigma_f\times$F.Y. of 5.69×10^{-27} and 4.83×10^{-27} in two determinations. A search for the 35-hr. Br^{82}, a shielded isotope, from a larger sample was not successful, the low activity found being resolvable into 22-hr. and 8-day curves which were probably iodine contamination. A maximum of $\frac{3}{4}$ of the observed 22-hr. period might have been 35-hr. activity and not resolved, placing the maximum value of $\sigma_f\times$F.Y. at 1.85×10^{-29}.

4. *Strontium* was isolated by precipitation of strontium and barium nitrates from fuming nitric acid. After two nitrate precipitations a ferric hydroxide decontamination precipitate was made. Barium was separated by precipitating as barium chromate from an acetate buffered solution. The strontium was precipitated from an ammoniacal solution as $Sr(C_2O_4)\cdot H_2O$ with ammonium oxalate.

The 9-hr. Sr^{91}, 2.7-hr. Sr^{92}, 53-day Sr^{89}, and 25-yr. Sr^{90} periods were studied. The 53-day Sr^{89} was observed in both bombardments *A* and *B*. Two values of $\sigma_f\times$F.Y., 3.3×10^{-26}, and 2.25×10^{-26}, respectively, were found. The 25-yr. Sr^{90} was measured by extracting the 60-hr. Y^{90} daughter

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[10] Plutonium Project Record, Vol. **9B** (to be issued).

[11] E. P. Steinberg and D. W. Engelkemeier, PPR Vol. **9B**, No. 7.2.1 (to be issued).

after allowing a strontium sample to equilibrate with the daughter. This gave a value of $\sigma_f \times$F.Y. of 2.16×10^{-26} for the 25-yr. Sr^{90}. The 9.7-hr. Sr^{91} was observed directly after allowing the 2.7-hr. Sr^{92} and its daughter to decay. The absorption correction used included an estimation of the effect of the 40 percent branching to an excited state of Y^{91} and the 9 percent conversion of the Y^{91} gamma-ray. A value of 2.08×10^{-26} was found for $\sigma_f \times$F.Y. for this isotope. The 2.7-hr. Sr^{92} decaying to a 3.5-hr. Y^{92} was found by subtraction of the 9-hour and 53-day periods. The observed curve was compared to a synthetic growth and decay curve for the two isotopes and found to fit quite well, the assumption being made that the two isotopes were counted with equal efficiency. A value of 2.23×10^{-26} was found for $\sigma_f \times$F.Y. of Sr^{92}.

5. *Zirconium* was separated by precipitation as barium fluozirconate after precipitation of lanthanum fluoride. The barium fluozirconate was dissolved in boric acid, reprecipitated, dissolved, and barium sulfate precipitated. The zirconium was then precipitated with cupferron and ignited to ZrO_2 for weighing and mounting.

The 65-day Zr^{95} was separated several months after bombardment *A*, counted, and an absorption curve taken immediately to eliminate effects caused by the growth of the 35-day columbium daughter. The activity observed was corrected back to the comparison time on the basis of the published half-life of 65 days. Two samples separated from the same bombardment gave values of $\sigma_f \times$F.Y. of 1.94×10^{-26} and 1.79×10^{-26}.

The 17-hr. Zr^{97} was counted after equilibration with the 75-min. daughter Cb^{97}. The observed activity of the parent plus the daughter gave values $\sigma_f \times$F.Y. of 1.98×10^{-26} and 2.09×10^{-26} for the yield of Zr^{97}.

6. *Molybdenum* was separated by extraction with ether after oxidation with bromine. The ether was washed with hydrochloric acid and evaporated. The residue was taken up in nitric acid and oxalic acid to complex the columbium, and α-benzoin oxime to precipitate molybdenum added. The precipitate was fumed with perchloric acid, made basic with ammonia, and two ferric hydroxide scavenging precipitates made. Molybdenum was then again precipitated with α-benzoin oxime after acidification. The α-benzoin oxime precipitate was dissolved in perchloric acid, and silver molybdate precipitated from an acetate buffered solution.

The 67-hr. Mo^{99} was observed and decayed to below background with no tailing of the curve. No growth of the 6.6-hr. Tc^{99} daughter was observed through the 7.5 mg of absorber present. From the observed activity a value of $\sigma_f \times$F.Y. of 1.81×10^{-26} was calculated.

7. *Ruthenium* was separated from bombardment *A* some six months after bombardment, at which time only the 1-yr. Ru^{106} was found. The separation was made by distillation of ruthenium tetroxide from perchloric acid and sodium bismuthate. The ruthenium tetroxide was collected in sodium hydroxide, reduced with alcohol, and the oxide separated. This was dissolved in hydrochloric acid and reduced with magnesium to ruthenium metal.

An absorption curve of the ruthenium showed the presence of no radiation other than the 4-Mev beta from the Rh^{106} daughter. The decay was followed over several months, and no activity shorter than 330 days was seen. A value of $\sigma_f \times$F.Y. of 2.27×10^{-26} was calculated for Ru^{106}.

8. *Palladium* was precipitated with dimethylglyoxime from a diluted nitric acid solution of the thorium. The dimethylglyoxime precipitate was dissolved in nitric acid, and made basic with ammonia and silver iodide precipitated from the ammonia solution. The solution was then acidified with hydrochloric acid, and palladium was again precipitated with dimethylglyoxime. In one experiment the palladium was mounted and counted, and, in a second, the palladium dimethylglyoxime precipitate was dissolved in nitric acid and aliquots of this solution periodically milked for silver after equilibration for one day. The palladium solution was stored in Lusteroid to avoid adsorption of silver by glass, which occurs to considerable extent when silver is present only in tracer quantities. The chemical yield of palladium was determined on that portion of the palladium solution not used for milkings.

The gross decay curve of palladium is not too useful, since two isotopes of palladium, Pd^{109} and Pd^{112}, are present with half-lives of 13.4 hr. and 21 hr., respectively. The final portion of the palladium gross decay curve was extrapolated back on a 21-hr. line. Using the absorption data of Seiler[12] for the 3.2-hr. Ag^{112} and 21-hr. Pd^{112}, $\sigma_f \times$F.Y. for the 21-hr. Pd^{112} was calculated to be 1.6×10^{-26} in bombardment *B*. In bombardment *D* the palladium was milked for its 3-hr. silver daughter, and the yield was calculated to be 0.8×10^{-26}.

9. *Silver* was separated by precipitation as silver chloride which was dissolved in ammonia, and two ferric hydroxide scavengings were made. Silver sulfide was then precipitated, dissolved in nitric acid, and the purification repeated. The silver was finally precipitated as silver chloride. The 7.5-day Ag^{111} was the only isotope found after decay of the 3.2-hr. Ag^{112}. A value of 1.32×10^{-26} was found for $\sigma_f \times$F.Y. for Ag^{111}.

10. *Cadmium* was separated as the sulfide, dissolved in acid, made ammoniacal, and hydroxide scavengings made with ferric and lanthanum hydroxides. From acid solution silver chloride was precipitated. From $2M$ HCl solution palladium and antimony sulfides were precipitated. The cadmium was finally precipitated as cadmium ammonium phosphate.

From bombardment *A*, the 43-day Cd^{115} period was separated and found to have a $\sigma_f \times$F.Y. of 1.01×10^{-27}. From a short bombardment both the 2.3-day Cd^{115} and the 43-day Cd^{115} were separated, and the 43-day isomer was found to give a value of 1.28×10^{-27} for $\sigma_f \times$F.Y. The decay of the 2.3-day isomer through the 4.5-hr. In^{115} and the fact that the gamma-ray from the 4.5-hr. In^{115} is 49 percent converted[13] was considered in calculating the fission yield of the 2.33-day cadmium. Two values of $\sigma_f \times$F.Y. obtained were 1.32×10^{-26} and 1.43×10^{-26} for this isomer.

11. *Tin* was separated as stannic sulfide. This was dissolved in concentrated acid, and an antimony sulfide scavenging was made from hot $2N$ HCl. From sodium hydroxide solution, ferric hydroxide, cadmium hydroxide, and columbic oxide were precipitated, followed by a second ferric hydroxide scavenging. In an acid solution the antimony sulfide scavenging was repeated. The tin was precipitated as metastannic acid from boiling nitric acid containing ammonium nitrate to which had been added a few drops of ruthenium carrier. The metastannic acid was ignited to SnO_2 and mounted on thin mica or glass and covered with Cellophane.

The isotopes of tin are in a rather confused state. On the Plutonium Project[5] periods of 62 hours, 10 days, and 130 days were found. Grummit and Wilkinson[14] found a 7-day and a 17-day period in place of the 10-day. Recently the 62 hour has been changed to 26 hour,[15] and is apparently identical with an isotope reported earlier by Livingood and Seaborg.[16] Lindner and Perlman[17] have recently shown this isotope to be Sn^{121} by deuteron bombardment of the separated tin isotope, Sn^{120}, and have further shown that no other period than the 26-hour one is formed by deuteron bombardment of Sn^{120}. Therefore, the 26-hr. period is the only long-lived isotope at mass 121. This leaves the 10 day, or 7 and 17 day, and the 130 day to be placed at Sn^{123} and Sn^{125}. Already at Sn^{125} is a 9-minute period which is apparently the parent of the 2.7-yr. Sb^{125}.[18] Further, Wilkinson[19] found that no antimony activity could be milked from any of the longer-lived tin isotopes they observed in the fission of U^{235} with slow neutrons.

In a long bombardment, tin was separated about six months after bombardment and only the 130-day period observed, decaying with a 130-day period over two half-lives. From a shorter bombardment, a 26-hr. period, a period of about 7.5 days, and two longer periods were found. The longer periods have not yet been resolved, but apparently one is the 130-day period and the other may have a half-life of about 20 days. The aluminum absorption curve of the 7.5 day, plus a small amount of the longer periods, had a Feather range of 1230 mg/cm² (2.5 Mev) which checks the reported energy of 2.6 Mev for the 10-day period[20] and is considerably greater than the 1.8-Mev energy reported by Grummit and Wilkinson for their 7- and 17-day periods.

Values of $\sigma_f \times$F.Y. have been calculated for the 26-hr., 7.5-day, and 130-day periods, the values

[12] J. A. Seiler, PPR Vol. **9B**, No. 7.21 (to be issued).

[13] J. M. Lawson and J. M. Cork, Phys. Rev. **57**, 982 (1940).

[14] W. E. Grummit and G. Wilkinson, Nature **158**, 163 (1946).

[15] E. P. Steinberg, private communication.

[16] J. J. Livingood and G. T. Seaborg, Phys. Rev. **54**, 775 (1938).

[17] M. Lindner and I. Perlman, Phys. Rev. **73**, 1124 (1948).

[18] C. W. Stanley and L. E. Glendenin, PPR Vol. **9B**, No. 7.23 (to be issued).

[19] G. Wilkinson, private communication (April, 1948).

[20] J. A. Seiler, PPR Vol. **9B**, No. 7.26.1 (to be issued).

found for the 26-hr. period being 1.09×10^{-26} and 0.98×10^{-26} in one bombardment and 0.62×10^{-26} in another. For the 7.5-day period values of 0.64×10^{-26} and 0.59×10^{-26} were found in one bombardment, and 0.78×10^{-26} and 0.91×10^{-26} in another bombardment. The 130-day period gave a value of $\sigma_f\times$F.Y. of 0.76×10^{-26}, an average of five values for the same solution. For the reasons given in the above discussion both the 7.5-day and 130-day periods have been tentatively placed at mass number 123.

12. *Antimony* was separated by precipitation as the sulfide, after oxidation and reduction of the carrier to ensure exchange. The sulfide was dissolved in nitric acid and fumed with sulfuric acid. The antimony was electrolyzed in 4–6 N H_2SO_4 from a lead cathode to give stibene, which was collected in silver nitrate solution as silver antimonide. This was treated with hydrochloric acid, and antimony was precipitated as the sulfide from the supernatant. The yields in this separation were poor, but according to Wilkinson[19] the antimony is very pure. The sulfide is not a desirable method of weighting antimony, and probably contains considerable impurities, making the results low.

Some 12 months after bombardment A, antimony was separated to obtain the 2.7-yr. Sb^{125}. The activity found has decayed a few percent over a period of four months. The aluminum absorption curve contained two beta-components, the more energetic of which gave a Feather range of 300 mg/cm^2 (0.75 Mev). The activity had a gamma-ray, of energy about 0.5 Mev as estimated from a lead absorption curve on 80 gamma c/m. This is in agreement with the published data for this isotope.[5] A calculation of $\sigma_f\times$F.Y. for this isotope gave values of 0.69×10^{-26} and 0.73×10^{-26}.

13. *Tellurium* was separated by evaporating the sample plus carrier with HBr several times, taking up in HCl and reducing the tellurium with sulfur dioxide. The tellurium was then dissolved in nitric acid, and ferric hydroxide scavengings were made from ammonia solution, after which the sample was precipitated twice more as tellurium metal with sulfur dioxide from hydrochloric acid solution.

The gross tellurium decay curve is quite complex and could not be resolved. However, from the growth of 2.4-hr. iodine a value of $\sigma_f\times$F.Y. for the 77-hr. Te^{132} of 1.44×10^{-26} was calculated. Milking a purified tellurium sample for iodine was not successful, since the iodine radio-chemical exchange was not quantitative from the tellurium solution because of complexing of iodine and tellurium. Following the activity of 2.4-hr. iodine separated periodically from aliquots of the original sample also gave poor results because of poor exchange of the iodine carrier. However, from the highest points on the curve obtained by periodic separations of the 2.4-hr. iodine, a value of 0.9×10^{-26} for $\sigma_f\times$F.Y. of the 77-hr. tellurium was obtained.

14. *Iodine* was separated by oxidizing the sample plus carrier with sodium hypochlorite in sodium carbonate solution to periodate, acidifying this, and reducing the iodine with hydroxylamine to free iodine which was extracted with carbon tetrachloride. The iodine was re-extracted into dilute sulfurous acid, re-oxidized with sodium nitrite and nitric acid, and re-extracted into carbon tetrachloride. Three such extraction cycles were run, and the iodine was then precipitated as silver iodide from nitric acid solution. The activity of the 8-day iodine was observed in samples separated after all the 22-hr. iodine had decayed, so no interference from the 5.3-day xenon daughter of the 22-hr. iodine occurred. The values of $\sigma_f\times$F.Y. were fairly consistent at 0.9×10^{-26}, 0.7×10^{-26}, and 0.8×10^{-26}. However, considering the scatter of points in the activity of the 2.4-hr. iodine observed in these same samples, it is probable that complete exchange with the carrier was not achieved.

15. *Cesium* was separated by precipitation of cesium perchlorate on dilution with alcohol after fuming with perchloric acid. The cesium perchlorate was dissolved, made basic with ammonia, and two ferric hydroxide scavenging precipitates made. The solution was evaporated, the beaker flamed to eliminate ammonium ion, and cesium again precipitated as the perchlorate which was mounted.

About 12 months after bombardment A the 33-yr. Cs^{137} was separated. The aluminum absorption curve showed the presence of two beta-components, the visual range and the Feather range of the more energetic being about 350 mg/cm^2 (0.9 Mev). A gamma-ray is present, to the extent of about 0.65 percent, which has an energy of about 0.9 Mev. This compares well with the published data, and a comparison of the observed aluminum absorption curve with that of Glendenin and Metcalf[21] showed them to be parallel. A calculation of $\sigma_f \times$F.Y. for the 33-yr. Cs^{137} gives a value of 5.1×10^{-26} in each of two samples. Since this value is higher than that of any other mass number, the cesium in one sample was redissolved and run through another series of purification steps, including hydroxide scavenging and sulfide scavenging. No change occurred in the specific activity of the sample, indicating no significant amount of impurities were present.

In bombardment *D*, cesium was separated to study the 13-day Cs^{136}, a shielded isotope. Two samples gave values of $\sigma_f \times$F.Y. of 9.0×10^{-28} and 9.3×10^{-28}.

16. *Barium* was separated by precipitation of the chloride from a concentrated hydrochloric acid and ether solution. After three such precipitations, a ferric hydroxide scavenging precipitate was made, and the barium reprecipitated as the chloride. The 300-hr. Ba^{140} was observed in all bombardments. The equilibrium mixture of 300-hr. barium and its 40-hr. lanthanum daughter was counted, and from this the activity of the barium was calculated. The values of $\sigma_f \times$F.Y. found were 2.44×10^{-26}, 1.96×10^{-26}, 1.73×10^{-26}, and 2.15×10^{-26} in bombardments *A*, *B*, *C*, and *D*, respectively.

17. *Cerium* was separated by first separating the thorium as the iodate from 5*N* nitric acid. Then a cerium hydroxide followed by a cerium fluoride precipitation was made. The fluoride was dissolved in boric acid and ceric iodate precipitated in the presence of lanthanum hold-back carrier. The precipitation of ceric iodate was repeated, and then a zirconium iodate scavenging precipitation was made. The cerium was precipitated as the hydroxide and finally precipitated as cerous oxalate.

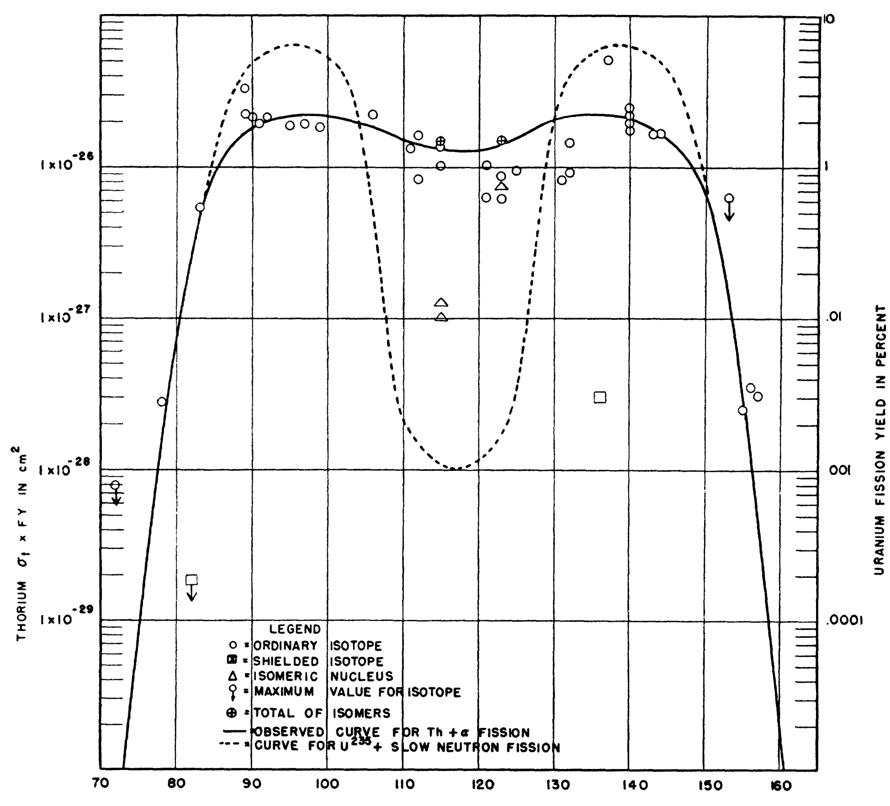

FIG. 1. Fission yield spectrum of thorium for fission with 38-Mev helium ions compared to that of U^{235} for fission with slow neutrons.

[21] L. E. Glendenin and R. P. Metcalf, PPR Vol. 9B, No. 7.39.1 (to be issued).

TABLE I. Values of $\sigma_f \times$F.Y. for various isotopes found in the bombardment of thorium with 37.5-Mev helium ions.

Isotope	Bombardment	$\sigma_f \times$F.Y. (cm^2)	Isotope	Bombardment	$\sigma_f \times$F.Y. (cm^2)
Zn^{72}	*B*	$<8 \times 10^{-29}$	Sn^{123}?(130d)	*A*	0.71×10^{-26}
As^{77}	*D*	$\sim 2.8 \times 10^{-29}$	Sn^{123}?(130d)	*A*	0.65×10^{-26}
Br^{82}	*C*	$<1.85 \times 10^{-29}$	Sn^{123}?(130d)	*A*	0.85×10^{-26}
Br^{83}	*C*	5.69×10^{-27}	Sn^{123}?(130d)	*A*	0.77×10^{-26}
Br^{83}	*C*	4.83×10^{-27}	Sn^{123}?(130d)	*A*	0.79×10^{-26}
Sr^{89}	*A*	3.34×10^{-26}	Sb^{125}	*A*	0.89×10^{-26}
Sr^{89}	*B*	2.25×10^{-26}	Sb^{125}	*A*	0.93×10^{-26}
Sr^{90}	*A*	2.16×10^{-26}	I^{131}	*D*	0.98×10^{-26}
Sr^{91}	*B*	2.08×10^{-26}	I^{131}	*D*	0.68×10^{-26}
Sr^{92}	*B*	2.23×10^{-26}	I^{131}	*D*	0.82×10^{-26}
Zr^{95}	*A*	1.94×10^{-26}	Te^{132}	*B*	1.44×10^{-26}
Zr^{95}	*A*	1.79×10^{-26}	Te^{132}	*D*	0.91×10^{-26}
Zr^{97}	*D*	2.09×10^{-26}	Cs^{136}	*D*	3.0×10^{-28}
Zr^{97}	*D*	1.98×10^{-26}	Cs^{136}	*D*	3.1×10^{-28}
Mo^{99}	*B*	1.81×10^{-26}	Cs^{137}	*A*	5.15×10^{-26}
Ru^{106}	*A*	2.27×10^{-26}	Cs^{137}	*A*	5.15×10^{-26}
Ag^{111}	*B*	1.32×10^{-26}	Ba^{140}	*A*	2.44×10^{-26}
Pd^{112}	*B*	$\sim 1.65 \times 10^{-26}$	Ba^{140}	*A*	2.47×10^{-26}
Pd^{112}	*D*	0.83×10^{-26}	Ba^{140}	*B*	1.96×10^{-26}
Cd^{115}(2.33d)	*B*	1.43×10^{-26}	Ba^{140}	*C*	1.74×10^{-26}
Cd^{115}(2.33d)	*B*	1.32×10^{-26}	Ba^{140}	*C*	1.72×10^{-26}
Cd^{115}(43d)	*A*	1.01×10^{-27}	Ba^{140}	*D*	2.23×10^{-26}
Cd^{115}(43d)	*B*	1.28×10^{-27}	Ce^{143}	*B*	1.66×10^{-26}
Sn^{121}	*B*	0.62×10^{-26}	Ce^{144}	*A*	1.67×10^{-26}
Sn^{121}	*C*	1.09×10^{-26}	Ce^{144}	*A*	1.73×10^{-26}
Sn^{121}	*C*	0.98×10^{-26}	Sm^{153}	*B*	6.3×10^{-27}
Sn^{123}?(7.5d)	*C*	0.98×10^{-26}	Eu^{155}	*A*	2.5×10^{-28}
Sn^{123}?(7.5d)	*C*	0.78×10^{-26}	Eu^{156}	*B*	3.5×10^{-28}
Sn^{123}?(7.5d)	*B*	0.64×10^{-26}	Eu^{157}	*B*	3.1×10^{-28}
Sn^{123}?(7.5d)	*B*	0.59×10^{-26}			

The 270-day Ce^{144} was separated six months after bombardment *A*. The activity decayed with a 270-day half-life with no shorter periods observed. The absorption curve showed the presence of no activity other than the 270-day cerium and its 17-min. praseodymium daughter, the activity at zero absorber determined by extrapolating the beta-absorption curve for the 17-min. praseodymium back to zero absorber being about equal to half the total activity extrapolated to zero absorber. The value of $\sigma_f \times$F.Y. for Ce^{144} was found to be 1.67×10^{-26} and 1.73×10^{-26}. The 33-hr. Ce^{143} was determined on a short bombardment and $\sigma_f \times$F.Y. for this isotope was 1.66×10^{-26}.

18. *Samarium* was separated along with the rest of the rare earths by fluoride precipitation after separation of the thorium and zirconium by iodate precipitation. The cerium was then separated by iodate precipitation. Samarium and europium were then separated from the rest of the rare earths by extraction with sodium amalgam from an acetate buffered solution. The amalgam was washed with water, and the samarium and europium were then extracted back into dilute hydrochloric acid. Three such sodium amalgam extraction cycles were run, lanthanum hold-back carrier being added in each case. The samarium and europium were then separated by reducing the europium with amalgamated zinc and precipitating samarium hydroxide with ammonia while the europium was in the reduced state. The samarium was further purified by precipitation from added inactive europium in a second cycle. The samarium was finally precipitated as the oxalate.

In bombardment *B* the 47-hr. Sm^{153} was studied. The activity found contained a 4.5-hr., a 45-hr., and 13-day periods, so the sample was undoubtedly contaminated with praseodymium, neodymium, and probably lanthanum. The 45-hr. component observed was probably composed of a mixture of 40-hr. lanthanum and 47-hr. samarium, so the value of $\sigma_f \times$F.Y. for this period, 0.6×10^{-26}, represents only a maximum value, and is probably considerably lower since the 13-day activity was quite pronounced indicating considerable impurities.

19. *Europium* was separated by isolating a rare earth fraction and separating europium from the rare earths by reduction with amalgamated zinc. In this case, after the first separation more lanthanum carrier was added, and two more decontamination cycles from other rare earths run. The europium was finally precipitated as the oxalate.

About eight months after bombardment *A*, europium was separated and a low activity found which was not removed by further decontamination cycles and was assumed to be the 2-yr. Eu^{155}. No absorption curve was possible, and the activity was extrapolated to zero absorber on the basis of Winsberg's data.[22] The value of $\sigma_f \times$F.Y. found was 2.5×10^{-28}.

From bombardment *B* the europium separated from the samarium fraction previously described gave activities of 15.4 hours and 15 days. Winsberg's data[22] was used to extrapolate the 15.4-hr. period to zero absorber. The value of $\sigma_f \times$F.Y. found for the 15.4-hr. Eu^{157} was 3.12 $\times 10^{-28}$. From the 15-day portion of the curve, and assuming the activity to be the daughter of a 10-hr. Sm^{156} parent as stated by Winsberg,[22] and further assuming that no significant separation of samarium and europium occurred in the sodium amalgam extraction, a process requiring several

[22] L. E. Winsberg, PPR Vol. 9B, No. 7.56.3 (to be issued).

TABLE II. Excitation data on fission products of thorium with helium ions.

Layer	Mg/cm² Th in layer	$\sigma_f \times$F.Y. of fission products in layer (10^{-26} cm²) Ce^{144}	Ba^{140}	Sn$^{123?}$(130d)	Cd^{115}(43d)	Ru^{106}	Sr^{89}	Specific gross β^- activity in layer c/m/mg Th
1	46.2	1.67 1.73	2.44 2.45	0.75 (av. of 5 values)	0.101	2.27	3.34	8.63×10^5
2	47.4	1.20	1.9	0.356 0.361	0.046	1.38	2.17	6.02×10^5
		1.23	1.53	0.364 0.377				
3	38.6	0.852 0.835	1.31 1.38	0.22	0.032	0.54	1.8	3.66×10^5
4	74.4	0.223 0.225	0.27 0.32	0.036	0.0049	0.16	0.37	9.57×10^4
5	71.3	0.00995	0.0115	0.0017	0.0018	0.0057	0.016	2.74×10^3
6	29.1	0.0041	0.0086	0.0012	0.00017	0.0021	0.0054	1.22×10^3
7	4.78	0.0034	0.0052	0.00082		0.0018	0.0040	1.09×10^3
8	46.1	0.0028	0.005	0.00094		0.0015	0.0039	9.4×10^2
9	29.3	0.0021	0.0039	0.00075		0.00095	0.0033	8.0×10^2
10	126.3	0.0011	0.0030	0.00016 0.00025		0.00061	0.0015	4.8×10^2
11	18.3	0.0024	0.0024	0.00012		0.00039		3.3×10^2

hours, a value of $\sigma_f\times$F.Y. for the chain of mass 156 was calculated to be 3.5×10^{-28}.

IV. THE FISSION YIELD SPECTRUM AND THE CROSS SECTION FOR FISSION

The values of the yields of the various fission products investigated at an average alpha-energy of 37.5 Mev have been tabulated in Table I. These are plotted in Fig. 1, in which the distribution of yields of isotopes formed in the bombardment of thorium with 37.5-Mev helium ions has been compared to that obtained from the slow neutron fission of U^{235}. Despite the spread of the points, it is at once apparent that the spectrum is quite different in the middle region of the distribution. Instead of a factor of 600 in yield between the peaks and the minimum as in slow neutron fission of U^{235}, there is only a factor of about two in the depth of the dip in the fission of thorium with 37.5-Mev helium ions.

It is also seen that the shielded isotopes Br^{82} and Cs^{136} were both formed in very low yield compared to the unshielded isotopes. The value of unshielded Cs^{137} is far off the drawn curve, and there is no obvious explanation. This isotope was taken from only one long bombardment, but other long-lived isotopes, e.g., Sr^{90}, give more reasonable values. It may be that another isotope is present, e.g., 2.3-yr. Cs^{134}, in about 10 percent abundance relative to Cs^{137}, though from the results with slow neutron fission where the 2.3-yr. Cs^{134} has not yet been detected this appears to be improbable. A second explanation is that the half-life of the Cs^{137} is about 15 to 20 years rather than 33 years. Since the Cs^{137} absorption curve contains two components, it would be difficult to find 10 percent of a shorter-lived isotope by absorption measurements, and the half-life of any second isotope must be long enough to make its detection by decay over a few months impossible with ordinary counting methods.

In Fig. 1 the sum of isomeric nuclei have been plotted. The isomers of Cd^{115} and the assumed isomers of Sn^{123} have been summed to give the total yield of those masses. No account was taken in the curve for the possible existence of any other tin isotope.

A summation of the values of $\sigma_f\times$F.Y. over the entire curve, and assuming the total fission yield to add up to 200 percent, a value for the cross section of thorium for fission with 37.5-Mev helium ions of 0.58±0.1 barn is found. The probable error of 0.1 barn is estimated from the distribution of points and the types of weighted curves which could be drawn through those points other than the one drawn. This gives a value of about 2.8 percent for the fission yield of Ba^{140}.

V. THE FISSION EXCITATION CURVE OF THORIUM WITH ALPHA-PARTICLES

The layers separated in the milling of the thorium disk used in bombardment *A* were analyzed separately for Ba, Sr, Sn, Ce, Ru, and Cd; hence it is possible to obtain a rough excitation curve for the fission of thorium by helium ions.

In Table II the measured yields are expressed as $\sigma_f \times$F.Y. for the isotopes studied in each of the various layers of thorium together with the specific beta-activity of that layer. This data is plotted on a logarithmic scale in Fig. 2. It is seen that fission continues to occur well beyond the range of the impinging 39-Mev alphas, which in thorium is about 340 mg/cm², and this is probably due to a slight deuteron contamination of the alpha-beam, to fast neutron fission, or to a combination of the two effects.

In the first 200 mg/cm² of the thorium the distribution does not appear to shift a great deal since the isotopes studied maintain their relative positions and remain in approximately the same proportions to each other and to the gross specific activity. The proportion of the 43-day cadmium

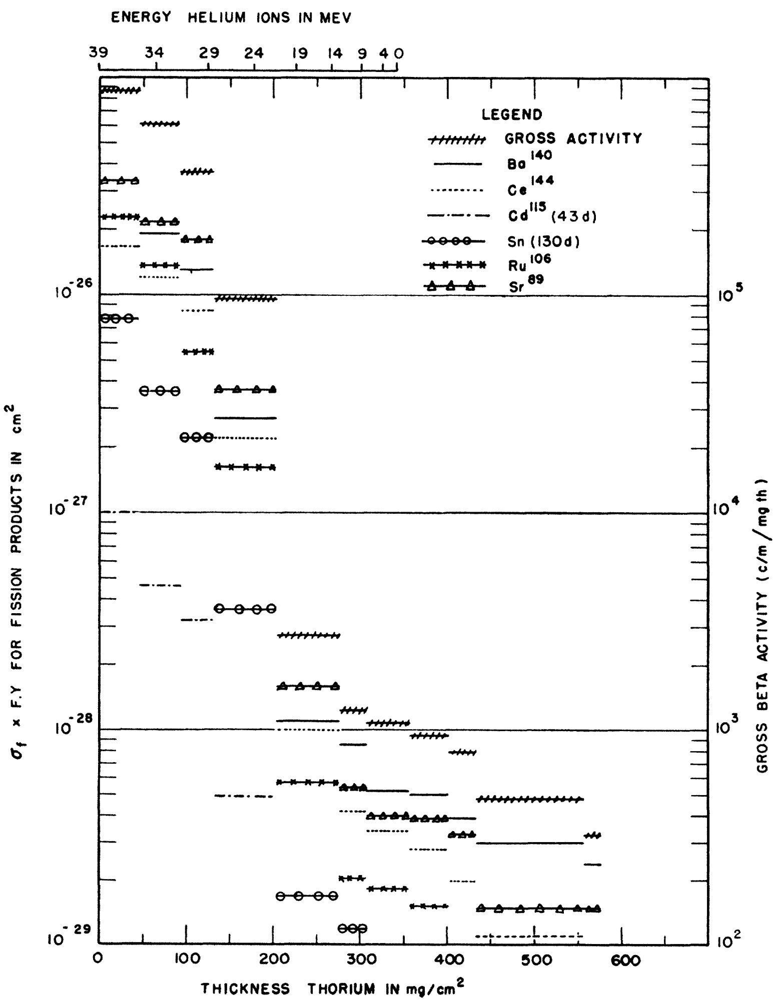

FIG. 2. Yield of some fission products from thorium+helium ions.

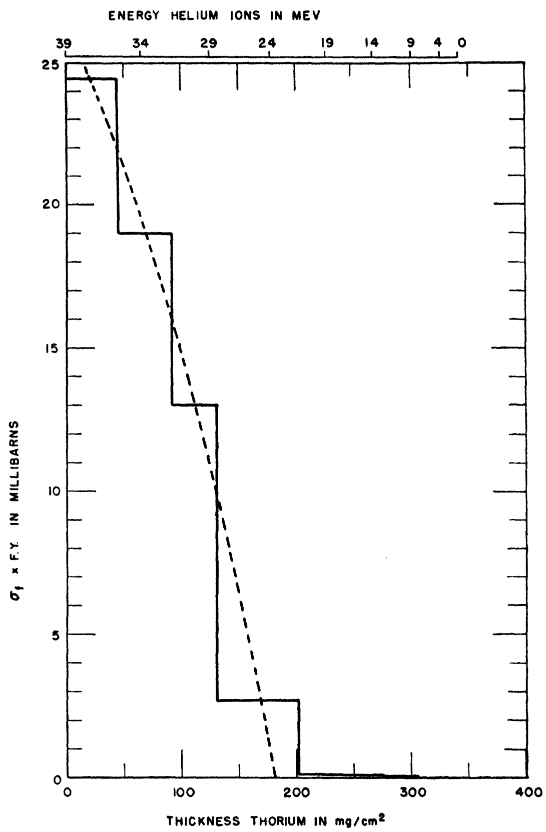

FIG. 3. Excitation curve of gross beta-activity in thorium target.

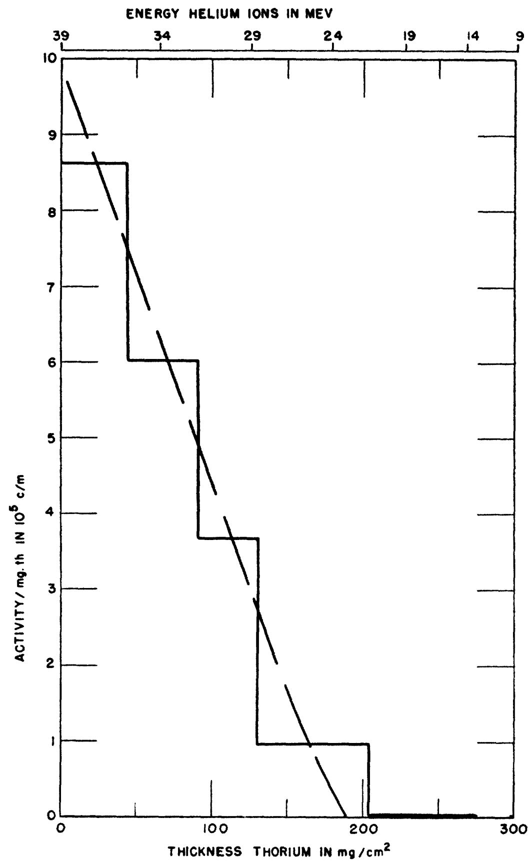

FIG. 4. Excitation curve of barium activity in thorium target.

does decrease somewhat, but whether this is significant or not is doubtful since the yield becomes so low. Below 200 mg/cm² the relative positions are maintained, except in the case of strontium which shifts from a position above barium to one below. In the first four layers Sr^{89} has an abnormally high yield compared to the other isotopes, since it is not expected to be above barium, if the two halves of the curve are symmetrical. The presence of krypton or rubidium impurities in the thorium target to produce Sr^{89} by particle reactions is not likely, and impurities of strontium or yttrium, while more likely as impurities, would require a neutron or deuteron flux of fairly high intensity to raise the yields of Sr^{89} to the values found.

In Figs. 3 and 4 the specific gross beta-activity and the $\sigma_f \times$F.Y. found for barium have been plotted against the thickness of thorium and an excitation curve drawn. The threshold for the alpha-reaction is found to be 23 to 24 Mev, and the cross section for fission is 0.6 barn at an average energy of 37.5 Mev, as found in the preceding section, the gross beta-curve indicating the higher value. This difference might be due to the presence of some long-lived beta-emitters formed at the higher alpha-energies by a process other than fission. These values differ somewhat from those reported by Jungerman and Wright,[23] who found a threshold of about 21 Mev and a cross section at 38 Mev of about 1.5 barn. From the data in this paper, the difference in threshold is probably not significant since too few points were taken to adequately define the curve near the threshold. The difference in cross section indicates that the beam measurements in this experiment may be in error or that the effect of different products of uranium and thorium in the

[23] J. Jungerman and S. C. Wright, Phys. Rev. **74**, 150 (1948).

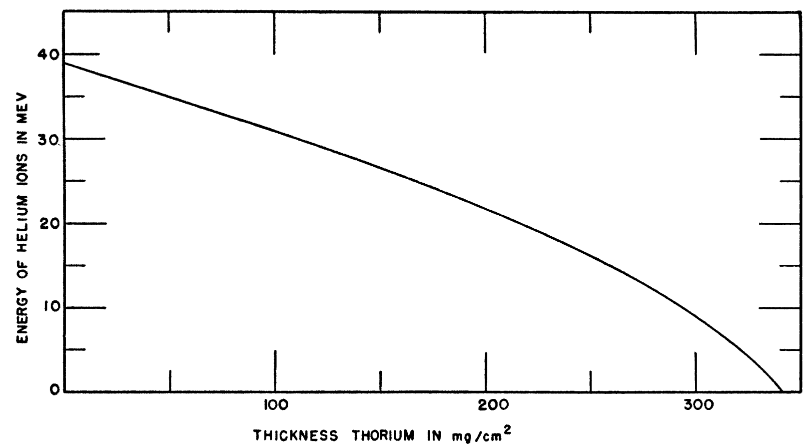

FIG. 5. Energy loss of 39-Mev helium ions in thorium.

experiments of Jungerman and Wright may be more serious than they expected. James[24] has made an extensive study of particle reactions in this region and believes that the number of atoms of thorium undergoing fission at this energy is about equal to those undergoing particle reactions which would be in line with the cross section reported here.

The conversion of thorium thickness to energy has been made on the basis of the curve shown in Fig. 5, which was obtained by integrating a rate of energy loss *vs.* energy curve for thorium. This latter curve was obtained by extrapolating data calculated by the theoretical group at the University of California Radiation Laboratory for the rate of energy loss of alpha-particles in lead, silver, copper, and aluminum to thorium, the extrapolation being made graphically at several energies by plotting $-dE/dx$ against $1/Z$ and extrapolating to thorium along a smooth curve.

VI. DISCUSSION

The distribution curve found for the fission of thorium with energetic alphas confirms a long standing suspicion that the isotopes in the center of the distribution are produced more abundantly in fission by highly energetic particles than is the case in slow neutron fission of U^{235}. In the case of fission of bismuth with 400-Mev alphas, the fission is apparently entirely symmetrical,[25, 26] and the primary fission products formed have an n/p ratio which is constant and equal to that of the fissioning nucleus. Goeckermann and Perlman[26] ascribe the supra threshold at which the fission occurs with decreasing the time between formation by boiling off neutrons of the highly excited nucleus which fissions and the fissioning of that nucleus into two fragments, to a point where rearrangement of the nucleons in the nucleus to give the most energetically favorable distribution of nucleons does not occur.

This same mechanism might be occurring in the fission of thorium with alpha-particles. In this case the compound nucleus U^{236} would be formed in a highly excited state and boil off some neutrons. However, this intermediate nucleus would still be in an energy state far more excited than that of the corresponding nucleus in fission of U^{235} by slow neutrons, and might therefore split much faster and before rearrangement to the most energetically favorable distribution of nucleons could occur. Therefore, one might expect a constant n/p ratio to be found for the primary fission products in this case, as was found in bismuth fission. However, with thorium this n/p ratio is probably that of U^{234} or U^{233}, i.e., ~ 1.53, since only a few neutrons need be boiled off before a nucleus is reached where fission occurs. Hence even if the products have a constant n/p ratio of 1.53, this will not be observed in the distribution curve since nuclei in the fission product region with this n/p ratio still are well above the line of stability and are all beta-emitters. Since the isotopes used in determining the distribution curve are all near the stability line, the primary products will still not be seen, and the only ob-

[24] R. A. James, private communication, August, 1948.
[25] I. Perlman, R. H. Goeckermann, D. H. Templeton, and J. J. Howland, Phys. Rev. **72**, 352 (1947).
[26] R. H. Goeckermann and I. Perlman, Phys. Rev. **73**, 1127 (1948).

servable difference in this respect between fission of thorium with helium ions and U^{235} with slow neutrons would occur in the length of the chains between the primary products and the stable nuclei, those chains on the heavy side of the distribution curve being shortened. This has not been investigated.

The depth of the dip in the mass yield curve must be related to the excess excitation in the fissioning nucleus. The results with slow neutron fission of U^{235}, where the factor in yield between dip and peaks of the distribution curve is about 600, fast neutron fission of thorium where the factor [a]6 is about 10, and fission of thorium with 37.5-Mev helium ions where the factor was found to be only about 2, indicate that there is a definite relation between the excitation of the nucleus and the occurrence of symmetrical fission. While the results at alpha-energies lower than 37.5 Mev do not give a complete picture of the distribution at these energies, enough is given to indicate that the dip is always much shallower than that found in slow neutron fission of U^{235}.

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A Note on the One-Electron States of Diatomic Molecules

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The explicit formulation of an integral of the motion is given for a single electron moving in the field of two fixed nuclei, for both the classical and the quantum mechanical cases. The corresponding quantal operator together with the Schrödinger Hamiltonian and the operator for the component of the orbital angular momentum of the electron about the internuclear axis form a complete set of commuting observables of the problem. This supplies the dynamical interpretation of the separation parameter of the energy equation.

I. INTRODUCTION

IT is well known that the equations of motion of the electron moving in the field of two fixed nuclei are separable in elliptic coordinates in both classical and quantum mechanics. In the latter case the separability of the Schrödinger equation falls under Case VII of Eisenhart's classification.[1] The separated differential equations have been studied in detail for the special case of the hydrogen molecular ion (H_2^+) by Burrau,[2] Wilson,[3] Teller,[4] Hylleraas,[5] and others.

The purpose of the present note is to establish the form of a general integral for the problem. While the use of this integral is implicit in all of the work which has been done on the two-center problem, back to that of Euler,[6] its explicit formulation and dynamical significance have not been given previously, to our knowledge. The possession of the integral does not give one any information which cannot be derived from the separation of the variables in the Schrödinger equation, but it is of theoretical interest as an important example of the use of first integrals in quantum mechanics. Actual examples of this

[1] L. P. Eisenhart, Phys. Rev. **74**, 87 (1948).
[2] Ø. Burrau, K. Danske Viden. Selskab **7**, Nr. 14 (1927).
[3] A. H. Wilson, Proc. Roy. Soc. London **A118**, 617 (1928); **A118**, 635 (1928).
[4] E. Teller, Zeits. f. Physik **61**, 458 (1930).
[5] E. Hylleraas, Zeits. f. Physik **71**, 739 (1931).
[6] E. T. Whittaker, *Analytical Dynamics* (Cambridge University Press, Cambridge, 1927), third edition, p. 97.