for electrons) is destroyed and the possibility of a tensor-tensor interaction alone is open. Furthermore it has been pointed out by Heitler[4] that the existence of discrete states would be very important in the scattering of mesons by nuclei.

In conclusion we may sum up the status of the "exact" solution of the meson-pair theory of nuclear forces as follows: Coulomb interaction and motion of heavy particles are neglected; neutron and proton are assumed to interact with mesons in a particular state $v(p)$ and if the interaction constant η is equal to or greater than

$$\left\{8\pi\mu\int_0^\infty v(p)^2dp\right\}^{-1}$$

discrete states are formed. If η is less than the critical value it is sufficient to consider the effect of the nuclear particle on the continuum of filled meson levels of negative energy.

The author would like to express his thanks to Professor Eugene Paul Wigner for suggesting an investigation of excited states of nuclear particles.

[4] W. Heitler, Nature **145**, 29 (1940).

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Branching Ratios in the Fission of Uranium (235)*

H. L. ANDERSON, E. FERMI AND A. V. GROSSE
Pupin Physics Laboratories, Columbia University, New York, New York

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A survey has been made of the percentages of slow neutron-induced uranium fissions that give rise to the formation of various radioactive series. These percentages were obtained by measuring the number of β-disintegrations of a suitable member of the series after an irradiation under specified geometrical conditions and a quantitative chemical separation. The percentages found for the series thus far investigated vary from about 0.1 percent to 10 percent.

A LARGE number of radioactive series found among the fission products of uranium have been reported. Thus far, however, the work has been confined mostly to the identification and the genetic relationships of the radio-elements which arise from uranium fission. The present work is part of a systematic attempt to determine in a quantitative way the probability that when uranium fission occurs a given radioactive series will appear. We shall call this probability the branching ratio of the radioactive series.

In Table I† are listed those radio-elements which have been found to date with indication, wherever possible, of their genetic relationships and atomic weights. In compiling this table we have relied mainly on the critical survey of Livingood and Seaborg,[1] supplemented by data which have appeared subsequently. In Table I the fission fragments have been arranged in two groups; a light group having atomic weights in the range from 82 to 100 and a heavy group with weights ranging from 127 to 150. To date, 10 radioactive chains have been identified in the light group and 12 in the heavy group. Because of greater analytical ease we have concentrated our effort on the heavy group and have determined the branching ratios of 9 out of 12. The sum of these heavy group fission series amounts to only about 50 percent, (see Table I) indicating that our present knowledge of fission fragments is still incomplete.

The presence of a variety of series may be interpreted by assuming that the original splitting may take place into different fragments. The emission of one or more neutrons further increases the number of possibilities. If no neutrons were emitted the sum of the weights of the two fragments ought to be equal to 236, in the case of fission produced by slow neutrons in U^{235}. If

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† *Note added in proof.*—The 22-hour and the 6.6-hour iodines have now been assigned to atomic weights 133 and 135, respectively, by C. S. Wu, Phys. Rev. **58**, 126 (1940).

[1] J. J. Livingood and G. T. Seaborg, Rev. Mod. Phys. **12**, 30 (1940).

TABLE I. *Radioactive series from uranium fission. S indicates a stable isotope.*

HEAVY GROUP

	127	128	129	130	131	132	133	134	135	136	137	138	139	140	—	—	—	—	—	—	—
Sb 51	80 hr.		4.2 hr.												5 min.	10 min.	10 min.				
Te 52	10 hr.	S	70 min.	S	25 min.										77 hr.	60 min.	43 min.	15 min.			
I 53	S				8 day										24 hr.	22 hr.	54 min.	6.6 hr.			
Xe 54		S	S	S	S	S		S		S			30s	short		5 day		9.4 hr.	15 min.		
Cs 55							S						6 min.	40s?					32 min.		
Ba 56				S		S		S	S	S	S	S	86 min.	300 hr.						14 min.	1 min
La 57													S	40 hr.						2.5 hr.	18 min
%	0.18		0.34		1.6								6.4	8.4	5.2	7.6	12	9(?)			

LIGHT GROUP

	82	83	84	85	86	87	88	89	90	—	—	—	—	—	—	—
Br 35		2.3 hr.								40 min.	3.8 hr.					
Kr 36	S	1.9 hr.	S		S		3 hr.	5 min.								
		S														
Rb 37				S		S	18 min.	15 min.								
Sr 38			S		S	S	S	51 day				6 hr.	7 min.			
Y 39								S				3.3 hr.				
Zr 40									S					17 hr.	70 day	
Cb 41														75 min.		
Mo 42																67 hr.
— 43														6.1		6.6 hr.
%																

neutrons are emitted this sum is correspondingly less. Since, presumably, the same number of neutrons is not always emitted, we need not expect that when a given series in the heavy group occurs it will always be accompanied by the same series in the light group. Very little information is available as to the relative probability of the formation of the various fission products. We have, therefore, undertaken a study of these probabilities. Since we shall be unable to continue this research for some months because of the rebuilding of the Columbia cyclotron now in progress, it was thought worth while to report on the results obtained thus far.

EXPERIMENTAL METHOD

In each measurement we irradiated a solution containing 50 grams of uranium element inside a paraffin block placed in a fixed position near the cyclotron. Usually, uranyl nitrate was used, but the sulphate was employed for the antimony determination. Neutrons were produced in the cyclotron by the bombardment of Be with 6-Mev protons. These neutrons have an energy up to about 2 Mev and our geometrical arrangement was such that practically only slow neutrons were effective in producing fission. Our results, therefore, refer to the fission of U^{235}. The intensity of irradiation was monitored by means of a gold foil placed in a fixed position inside the paraffin block. The activity of the gold foil was compared to that of a standard uranium plaque by means of an ionization chamber. In a few cases a Rn+Be source was used for the irradiation instead of the cyclotron.

For the investigation of each element a known amount of the element in a suitable chemical form was added as a carrier to the uranium solution, together with smaller quantities of all other known fission products. The desired element was subsequently purified and separated. A weighed fraction of the added amount was painted on a thin aluminum strip, covered with Scotch cellulose tape and wrapped around a thin-walled silvered-glass counter.[2] The strip was held by means of a suitable aluminum container so as to insure the reproducibility of its disposition relative to the counter. Taking into account the absorption of the β-rays, the geometrical efficiency of the counting arrangement, and the finite time of irradiation, as explained below, the number of β-disintegrations per second at saturation of the element in question was obtained. The branching ratio, i.e., the fraction of fissions giving rise to a particular radioactive series, was calculated from the formula,

$$R = I\eta/gfMFN.$$

[2] Made by Eck and Krebs, New York City.

I is the observed initial activity of the preparation measured in counts per minute; f is the ratio of the weights of carrier element used for the activity measurement to that added to the irradiated solution; M is the intensity of irradiation as measured in arbitrary units with the gold monitor; F is the fraction of saturation of the activity in question accumulated during the time of irradiation; η is the correction factor for the absorption of the β-rays. For its determination an aluminum cylinder of thickness 0.043 g/cm^2, equivalent to the combined thickness of the counter wall and the Scotch tape, was inserted between the sample and the counter. If an exponential law of β-ray absorption is assumed, η is the ratio of the measured activity without the aluminum to that with it. Values of η are given in Table II. The thickness of the counter wall was found by comparing, with a second counter as detector, the absorption for β-rays of radium E of the counter used with an aluminum cylinder of known thickness under the same geometrical conditions. In this way the thickness of the counter wall was found to be equivalent to 0.029 g/cm^2 of aluminum. The Scotch tape had a thickness equivalent to 0.012 g/cm^2. A small increment was added to take into account the absorption in the average sample measured.

The symbol g is the geometrical efficiency of the counting arrangement. It was determined by measuring in our standard way the β-activity of a weighed amount of U_3O_8 and was found to be $\frac{1}{3}$. Differences in this efficiency caused by possible differences in the back-scattering of the various β-rays were neglected. The measurement of g was repeated several times with varying amounts of U_3O_8 and was found to give the same result to within a few percent.

N is the number of fissions per minute taking place in the uranium solution for unit intensity of irradiation. In order to determine N we proceeded in the following way: In place of the uranium solution we used 140 cc of a solution of $MnSO_4$. In order to alter the distribution of neutrons as little as possible the concentration of this solution was adjusted so that the number of neutron captures per second would be roughly the same as in the uranium case. The activity of a known fraction of these manganese atoms was then determined with the counter in our standard way. The number of fissions per uranium atom is equal to the number of disintegrations per manganese atom times the ratio of the fission cross section of uranium to the capture cross section of manganese for thermal neutrons. We found that for unit intensity of irradiation 960,000 fissions took place in our uranium solution per minute.

On the assumption that a series of radio-elements has no branchings we chose for each radioactive series one convenient radio-element for determining the percentage of fissions of the series. The chemical elements investigated were I, Sb, Ba and Zr. The series investigated are those of Table I for which the percentages are given.

Iodine

Iodine was separated by the following method: A standard solution of 5 KI+1 KIO_3 was added to the uranium solution and acidified with dilute H_2SO_4 to free all of the iodine. The free iodine was then steam distilled into a flask containing water and converted into iodide ion by titrating with $NaHSO_3$; the iodine was then precipitated as either PdI_2 or AgI.

Originally, our practice was to paint PdI_2 precipitated in the presence of Br ion on the aluminum strip. Since on several occasions the PdI_2 reacted with the aluminum with some loss of iodine, these measurements could be relied on for giving only relative branching ratios of the various iodine isotopes. For absolute determinations we used a precipitate of AgI. Bromine is not separated in this case, but since the bromine products are short-lived the determinations could be made on the longer-lived iodines. For these last measurements we used a Rn+Be source.

Separation of iodine has permitted us to

TABLE II. *Absorption factors η for 0.043 g/cm^2 of aluminum.*

Element	Half-Life	η	Element	Half-Life	η
$_{51}Sb^{129}$	4.2 hr.	1.67	$_{53}I$	22 hr.	1.85
$_{52}Te^{129}$	70 min.	1.81	$_{53}I^{131}$	8 days	4.9
$_{51}Sb^{127}$	80 hr.	1.61	$_{56}Ba^{139}$	86 min.	1.26
$_{52}Te^{127}$	10 hr.	1.66	$_{56}Ba^{140}$	300 hr.	1.52
$_{53}I$	54 min.	1.60	$_{57}La^{140}$	36 hr.	
$_{53}I$	2.4 hr.	1.60	$_{40}Zr$	17.2 hr.	1.39
$_{53}I$	6.6 hr.	2.0	$_{41}Cb$	75 min.	

investigate the following series,[3]

(1) Te^{131} (30 min.) or (30 hr.)→I^{131} (8 day),

(2) Sb (10 min.)→Te (60 min.)
→I (22 hr.)→Xe (5 day),

(3) Te (~15 min.)→I (6.6 hr.)→Xe (9.4 hr.),

(4) Sb (5 min.)→Te (77 hr.)→I (2.4 hr.),

(5) Sb (<10 min.)→Te (43 min.)→I (54 min.).

In one experiment iodine was separated 24 hours after the end of a 7-hour irradiation. The decay curve was readily analyzed into an 8-day and a 22-hour activity; no evidence for the presence of the 5-day Xe could be found and it was assumed that this gas escaped from the sample during measurement.

We were able to determine that the 8-day iodine arises principally from the 30-minute rather than the 30-hour isomer of tellurium. This was done by irradiating for 2 hours and separating the iodine after 22 hours and again after 89 hours. If the 8-day iodine arose from the 30-hour tellurium the ratio of its activity from the second separation to that from the first would be 1.1. If instead, it arose from the 30-minute tellurium this ratio would be zero. A ratio of 0.1 was found which indicates that the 8-day iodine arises principally from the 30-minute tellurium.

The branching ratio of the 2.4-hour activity was obtained as follows. The parent substance of this radio-element has a much longer life (77 hours) than the parent elements of all other radio-iodines produced in the fission. It follows that the 2.4-hour radio-iodine is the only radio-iodine which can be regenerated in an irradiated uranium solution from which iodine has been removed once one or two days after irradiation. We therefore irradiated a solution and after about one day performed a first iodine separation. The collected iodine has a decay curve in which all the following periods were represented: 8 days, 2.4 hours, 66 hours and 22 hours. The solution was then allowed to stand for about one more day; the 77-hour tellurium, which was still present in the solution, reproduced during this time the 2.4-hour iodine; a second iodine separation collected, therefore, the 2.4-hour iodine almost pure.

The activity of the 54-minute iodine could easily be distinguished from all other activities because of the large difference in period from the other activities and the fact that a separation of iodine soon after a short irradiation yields this activity with a much greater intensity than any other.

We were, however, not able to get a very convincing decay curve of the 6.6-hour iodine, perhaps because the daughter substance (9.4-hour Xe) was occluded only in part in our sample. The corresponding branching ratio given in the table is somewhat doubtful. The branching ratios in the case of the 22-hour and the 2.4-hour iodines were measured using AgI precipitation. The branching ratios of the other isotopes were obtained by analyzing decay curves of PdI_2 precipitations and comparing initial activities of the 22-hour and 2.4-hour activities with the others. Irradiations of various lengths of time gave reasonably consistent results.

Antimony

Antimony and some tellurium were added to the uranium sulphate solution in the form of a solution of $SbCl_3$ in 50-percent H_2SO_4 and reduced with granulated zinc and sulphuric acid in a hydrogen generator to SbH_3, and absorbed in a $AgNO_3$ solution. The silver antimony precipitate was filtered, decomposed with concentrated HCl and antimony precipitated with H_2S from the solution in the usual way and weighted as Sb_2S_3 after drying in a stream of CO_2.

The activity of the Sb samples was analyzed in order to determine the branching ratios of the following series:[3]

(1) Sb^{127} (80 hr.)→Te^{127} (10 hr.)→I^{127},

(2) Sb^{129} (4.2 hr.)→Te^{129} (70 min.)→I^{129}.

The decay curves show evidence of the growth of the two radio-telluriums (10 hours and 70 minutes) out of the chemically separated samples of antimony. The branching ratios of these two series are considerably smaller than any other found so far. This fact may perhaps be related to the circumstance that these series are at the low atomic weight end of the heavy group.

[3] P. H. Abelson, Phys. Rev. **56**, 1 (1939).

Barium

This element was separated from the irradiated solution by first eliminating most of the $UO_2(NO_3)_2$ by ether extraction and precipitating Ba and Sr as sulphates in the aqueous extract. These sulphates were melted with $KNaCO_3$ in the presence of the oxides of Te, Mo and Cb, the washed carbonates dissolved in dilute HCl and precipitated, in the presence of Th (to eliminate UX), for a second time as sulphates. After converting these to carbonates Ba was separated from Sr as $BaCrO_4$ in acetic acid solution and reconverted into $BaSO_4$.

From the activity of the barium samples we determined the branching ratios of the two series:[4]

(1) Xe^{139} (<30 s)→Cs^{139} (6 min.)
→Ba^{139} (86 min.)→La^{139},

(2) Xe (short)→Cs (40 s?)
→Ba (300 hr.)→La (40 hr.).

A first barium sample was separated from a uranium solution irradiated for many hours. Special care was taken to purify the sample as thoroughly as possible from UX contamination. Since this purification process took several days only the 300-hour activity was found in the decay curve. Actually, the lifetime appeared from our measurements to be somewhat longer. The first part of the decay curve shows evidence of the growth of the 40-hour lanthanum. This growth was, however, not sufficiently intensive for a reliable separate determination of the absorption factors of Ba and La. In determining the branching ratio we used the average absorption factor of the complex radiation of Ba and La. This procedure may of course introduce some additional error into this determination.

The activity of the 86-minute barium was measured on a $BaCrO_4$ sample separated from a uranium solution irradiated about one hour. Because of the short time available for the purification an appreciable UX background was observed in this sample. The 86-minute activity was, however, much more intensive and could be analyzed without difficulty.

[4] O. Hahn and F. Strassmann, Naturwiss. **27**, 529 (1939); G. N. Glasoe and J. Steigman, Phys. Rev. **58**, 1 (1940).

Zirconium

For the chemical separation of zirconium the U solution was acidified with concentrated HCl to about 25 percent, the Zr precipitated as a phosphate and the ZrP_2O_7 purified as previously described[5] and finally converted into ZrO_2.

The chain investigated was:

Zr (17.2 hr.)→Cb (75 min.).

A zirconium oxide sample separated from a solution irradiated about two hours was measured. The decay curve showed an initial rise caused by the growth of the 75-minute columbium and then a decay with the 17.2-hour period of zirconium. A rather small background, presumably caused by a long-living zirconium was also observed. We were unable to separate the two absorption factors of 17.2-hour zirconium and 75-minute columbium from these curves and we used, therefore, an average absorption coefficient. Two irradiations, one of 2 hours and another of 10 hours, gave, in good agreement, 5.9 and 6.3 for the branching percentages.

Assuming that one radioactive fragment in the light group and one radioactive fragment in the heavy group are produced in each fission, we would expect the sum of the branching percentages to be 100 for each of the two groups. The percentages for the nine series analyzed in the heavy group add to about 50. Apart from possible errors, especially in the determination of the number of fissions, this low result is partly caused by incomplete analysis (9 out of 12) of the known series and probably also by series not yet discovered.

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[5] A. V. Grosse and E. T. Booth, Phys. Rev. **57**, 664 (1940).