Genetic Relationships and Fission Yields of Members of the Mass-115 Decay Chain*

ARTHUR C. WAHL[†] AND NORMAN A. BONNER[‡] Los Alamos Scientific Laboratory, Los Alamos, New Mexico (Received October 29, 1951)

The 21-minute Ag¹¹⁵ has been found to decay to both Cd¹¹⁵ isomers, 9 percent going to the 43-day state and 91 percent to the 53-hour state. No evidence was found for the decay of either cadmium isomer to the other. The 53-hour cadmium isomer decays to the 4.5-hour In^{115m}, but < 0.02 percent of the 43-day cadmium decays to In^{115m}. The U²³⁵ thermal neutron fission yields of the 21-minute silver, 53-hour cadmium, and 43-day cadmium were determined to be 0.0078 percent, 0.0098 percent, and 0.00071 percent, respectively. All of the 43-day cadmium isomer formed in fission grows from the 21-minute silver, but 28 percent of the 53-hour isomer is formed by some other path. The fission yield of each of the cadmium isomers increases one hundred-fold when the energy of the neutrons is increased to 14 Mev. Evidence is presented for the formation of 3.5-year Cd^{113m} in approximately 5×10^{-5} percent yield from the thermal-neutron fission of U²³⁵.

I. INTRODUCTION

THE existence of the Ag¹¹⁵ (21-minute)—Cd¹¹⁵ (53-hour and 43-day)—In^{115m} (4.5-hour) decay chain has been recognized for some time. Each member of the chain is found among the fission products and is produced by at least one nuclear reaction which establishes the mass number. However, except for the observed growth of the 4.5-hour indium from the 53-hour cadmium, the genetic relationships of the cadmium isomers with the other members of the chain were unknown. Therefore, we undertook to establish these relationships. We also measured the fission yields of several chain members for both thermal-neutron-induced and 14-Mev-neutron-induced U²³⁵ fission.

The principal published facts concerning this chain at the time of writing this paper are the following:

21-minute Ag¹¹⁵

A 22-minute silver was identified among the fission products of U²³⁵ by Turkevich,¹ but the mass number and fission yield were not determined. Duffield and Knight² produced a 20-minute silver, presumably the same isotope observed by Turkevich, by the γ ,*p* reaction on cadmium. They assigned this silver activity to mass number 115 by the use of separated cadmium isotopes. However, they were unable to observe the growth of either of the Cd¹¹⁵ isomers because of the small amount of 20-minute silver produced.

53-hour Cd115

The 53-hour cadmium has been assigned to mass number 115 on the basis of a variety of nuclear reac-

tions,³ including the n,γ reaction on an enriched Cd¹¹⁴ sample.⁴ It has been shown to be the parent of the 4.5-hour In^{115m, 5,6}

The half-life has been variously reported to have values in the range 54 to 58 hours.^{3,4}

The decay-scheme³ involves two beta-particles, one of energy 1.10 Mev unaccompanied by gamma-rays, and one of energy 0.56 Mev in coincidence with a 0.54-Mev gamma-ray. The data of Mandeville, Scherb, and Keighton⁷ indicate that about 12 percent (or slightly more) of the cadmium decays by way of the beta-gamma path, the remainder going by way of the 1.10-Mev beta-ray path.

The fission yield (thermal neutrons on U^{235}) has been reported to be 0.011 percent.⁸

43-dav Cd115

The 43-day cadmium has been assigned to mass number 115 principally by means of the n,γ reaction on an enriched Cd¹¹⁴ sample⁴ and the n,p reaction on indium.⁹ It decays almost entirely by emission of a single beta-ray, the energy of the transition being somewhere between 1.4 and 1.8 Mev.^{3,4,10,11} A small fraction (~1 percent or less) decays by way of a soft beta-ray in coincidence with several gamma-rays.^{10,12}

It has been reported¹² that approximately 0.007

⁵Goldhaber, Hill, and Szilard, Phys. Rev. 55, 47 (1939); Nature 142, 521 (1938).

⁶ Nishina, Yasaki, Ezoe, Kimura, and Ikawa, Nature 146, 24 (1940).

¹⁰ Gill, Mandeville, and Shapiro, Phys. Rev. **80**, 284 (1950). ¹¹ G. T. Seaborg and I. Perlman, Revs. Modern Phys. **20**, 585 (1948).

¹² Árgonne National Laboratory Report 4525 (1950).

^{*} This Document is based on work performed at the Los Alamos Scientific Laboratory of the University of California under the auspices of the AEC during the summer of 1950, while the authors were serving as consultants to the laboratory. † Present address: Department of Chemistry, Washington

[†] Present address: Department of Chemistry, Washington University, St. Louis, Missouri. ‡ Present address: Department of Chemistry, Cornell Uni-

[‡] Present address: Department of Chemistry, Cornell University, Ithaca, New York. ¹ A. Turkevich, Argonne National Laboratory Report 4010

¹A. Turkevich, Argonne National Laboratory Report 4010 (1947).

² R. B. Duffield and J. D. Knight, Phys. Rev. 75, 1613 (1949).

⁸ Way, Fano, Scott, and Thew, *Nuclear Data*, Nat. Bur. Standards Circ. 499 (1950).

⁴ Cork, Rutledge, Stoddard, Branyan, and Le Blanc, Phys. Rev. 79, 938 (1950).

⁷ Mandeville, Scherb, and Keighton, Phys. Rev. **75**, 221 (1949). ⁸ R. P. Metcalf, Report CC-2310 (1945); see also *National Nuclear Energy Series* (McGraw-Hill Book Company, Inc., New York, 1951), Vol. 9, Division IV, paper 125 (p. 891) and paper 127 (p. 898).

⁽p. 898).
⁹ Seren, Engelkemeier, Sturm, Friedlander, and Turkel, Phys. Rev. 71, 409 (1947); see also Paper 124, Vol. 9, Div. IV, National Nuclear Energy Series (McGraw-Hill Book Company, Inc., New York, 1951).
¹⁰ Gill, Mandeville, and Shapiro, Phys. Rev. 80, 284 (1950).

percent of the 43-day cadmium decays to 4.5-hour In^{115m}.

The fission yield (thermal neutrons on U²³⁵) has been reported to be 0.0008 percent.8

Although from nuclear shell theory it seems very probable that the 43-day cadmium is the upper isomeric state, this conclusion is not reached unambiguously from simple energy considerations.

4.5 hour In115m

The 4.5-hour indium has been produced by a variety of nuclear reactions.³ It decays principally by isomeric transition to the ground state, about 50 percent of the transitions going by internal conversion.¹³ About 6 percent of the 4.5-hour indium decays to stable Sn¹¹⁵ by emission of an 0.8-Mev beta-particle.^{5,14}

Martell and Libby¹⁵ have shown that the In¹¹⁵ ground state decays to tin with the emission of a 0.6-Mev beta-particle. The half-life is nearly 10¹⁵ years.

II. GENERAL PROCEDURES AND CALIBRATIONS

A. Purification of Fission-Product Cadmium

Cadmium activities formed in fission were purified by a method described by Glendenin.¹⁶ This method involves three cadmium sulfide precipitations from 0.2Macid, two palladium sulfide precipitations from 2Macid (Glendenin used 1.2M acid), two basic ferric acetate scavenging precipitations, one cadmium sulfide precipitation from ammonia, and one cadmium ammonium phosphate precipitation. Glendenin claims this procedure results in 43-day Cd¹¹⁵ of about 99 percent radiochemical purity as shown by decay curves. We have checked the purity of a 43-day Cd¹¹⁵ sample, obtained by the foregoing purification procedure from U²³⁵ which had been irradiated one week in the Los Alamos homogeneous reactor (water boiler), by carrying the purification through an additional cycle (one cadmium sulfide, one palladium sulfide, and one basic ferric acetate precipitation). The further purification did not measurably decrease the specific activity of the cadmium, so the radiochemical purity of the original sample must have been > 98 percent.

B. Sample Mounting and Chemical Yields

Cadmium was mounted for counting by precipitating CdNH₄PO₄·H₂O according to Glendenin's directions,¹⁶ filtering onto tared Whatman's No. 42 filter paper, drying in a 110°C oven for 15 minutes, cooling in a desiccator for 20 minutes and in the balance case for 10 minutes, weighing to determine the chemical yield, and mounting on $\frac{1}{16}$ -inch aluminum plates under Scotch

Tape $(9.2\pm0.1 \text{ mg/cm}^2)$. The weight of a precipitate was about 30 mg, and it was spread over an area of 3.5 cm².

The chemical yield for the over-all purification and mounting procedure was usually \sim 70 percent.

The composition of the precipitates resulting from the foregoing procedure was checked by comparing the weights of four precipitates containing tagged cadmium with the weights calculated from the radiochemical yields, which were determined from the activity of the tagged cadmium remaining in the filtrates. The activity of the cadmium recovered from the filtrates was measured on cover slips under Scotch Tape in the same manner as the activity of the original cadmium tracer solution had been measured. The weights of the cadmium ammonium phosphate precipitates averaged 2 to 3 percent lower than the calculated values. Whether this small discrepancy is real, indicating less than one molecule of water of hydration, or whether it is the result of incomplete (about 80 percent) recovery of the cadmium in the filtrates is not known. We have assumed the latter and have used the molecular weight corresponding to $CdNH_4PO_4 \cdot H_2O$ in our yield calculations.¹⁷ If the discrepancy is real, the error introduced because of the uncertainty in the composition is <3 percent.

Indium was mounted for counting by precipitating the hydroxide with ammonia from a chloride-free nitric acid solution, igniting to the oxide at 800°C for 15 minutes, powdering the sample, suspending it in water, filtering onto tared Whatman's No. 42 filter paper, and drying, cooling, weighing, and mounting as described for cadmium ammonium phosphate. The weight of a precipitate was 15 to 20 mg and covered an area of 3.5 cm^2 . We found that the weight of ignited indium oxide did not change when wet with water and dried by this procedure.

The uniformity of the Scotch Tape covering was checked three times during the course of this work by weighing three \sim 3-inch strips of 1-inch tape. All nine determinations agreed to within 0.2 mg/cm².

Carrier solutions were standardized by conventional analytical procedures. Silver was weighed as silver chloride dried at 110°C; cadmium was weighed as cadmium sulfate ignited at 500°C; indium was weighed as indium oxide ignited at 800°C.

C. Counting Efficiencies

The decay of samples mounted as described previously was followed on external beta-proportional methane-flow counters, a sample being 8 mm from a 2-inch diameter, 0.0007-inch (4.8 mg/cm²) Dural window. The counters were 2 inches inside diameter, and the window was in a side. The efficiency of this method

¹³ J. L. Lawson and J. M. Cork, Phys. Rev. 57, 982 (1940).

 ¹⁶ J. L. Lawson and J. M. Cork, Phys. Rev. 57, 982 (1940).
 ¹⁴ Bell, Ketelle, and Cassidy, Phys. Rev. 76, 574 (1949).
 ¹⁵ E. A. Martell and W. F. Libby, Phys. Rev. 80, 977 (1950).
 ¹⁶ L. E. Glendenin in Report CN-1312X, p. 121 (1945); see also National Nuclear Energy Series (McGraw-Hill Book Company, Inc., New York, 1951), paper 265, Vol. 9, Division IV (p. 1575).

¹⁷ Occasionally the precipitates failed to crystallize, or crystallized in some form of lower molecular weight so the specific activity of a sample was unreasonably high. In such cases the precipitate was dissolved and reprecipitated or the results were discarded.

of counting the 53-hour Cd¹¹⁵, 43-day Cd¹¹⁵, and 4.5-hour In^{115m} radiations was compared to the efficiency of counting these radiations from thin $(<0.5 \text{ mg/cm}^2)$ samples mounted¹⁸ on thick glass plates (a stack of microscope cover slips) on an internal beta-proportional methane-flow counter (a nucleometer). It was assumed that the efficiency of counting either the 53-hour or 43-day cadmium beta-particles in this internal geometry was the same as the radium E (Bi²¹⁰) counting efficiency, which we determined to be 70 percent (see following material). The approximate validity of this assumption seems reasonable because all of the beta-rays are at least fairly energetic, the decay schemes are reasonably simple, and Hudis¹⁹ has found the efficiency of this same geometry (on a different nucleometer) for I¹³¹ and P³², furnished by the Bureau of Standards, to be 72 and 68 percent, respectively.

Pure RaE was prepared from a RaD, RaE, RaF mixture in 25 ml of 0.1M HCl containing about 0.5 mg of lead carrier by depositing RaE and RaF on platinum over which hydrogen was bubbling, dissolving the RaE and RaF from the platinum in warm 6M HNO₃, converting them to the chlorides by several evaporations with HCl, and depositing the RaF on silver from a hot 1.5M HCl solution. Three cover-slip plates were prepared. The decay of the RaE was followed on the nucleometer, and the growth of RaF was followed on an alpha-proportional methane-flow counter. The efficiency of the internal beta-proportional counter for RaE radiation was calculated from the known 51 percent geometry of the alpha-counter, the half-life values of RaE and RaF, 5.0 days and 138.3 days, respectively, and the activities of the three samples. The result obtained was that 70 percent (69, 70, 72 percent) of the RaE disintegrations were counted. The purity of the RaE was demonstrated by the absence of alpha-activity in the freshly prepared samples (< 0.5 percent of the alpha-activity at maximum growth) and by the decay of a RaE sample under Scotch Tape on an external proportional counter with a 5.0-day half-life over a factor of 40. A small tail indicates that ~ 0.3 percent of the initial RaD came through the procedure with RaE.

A second method of determining the RaE counting efficiency involved comparison of the activities of a Bureau of Standards RaD, E, F standard mounted on a silver-palladium disk with aliquots of a RaD, E, F solution mounted on glass cover slips and on silverpalladium disks. All samples were measured under a 9.5 mg/cm² aluminum absorber in the internal nucleometer. The fraction of the RaE radiation absorbed in the aluminum absorber was determined empirically with samples prepared from the pure RaE solution mentioned before. 37 percent of the radiation emitted from a sample on glass and 38 percent of the radiation emitted from a sample on palladium was absorbed. (Extrapolation of absorption curves gave an incorrect value of 26 percent absorption.) The efficiency of RaE beta-counting on glass in the nucleometer, determined by this method, was 67 percent.

4.5-hour In^{115m} is always present in 53-hour Cd¹¹⁵ samples, being essentially in transient equilibrium in all samples more than one day old. Thus it was necessary to determine what fraction of the radiation from old 53-hour Cd¹¹⁵ samples detected by the nucleometer was caused by In¹¹⁵ decay. Two runs were made in which indium was separated from cadmium samples by several cadmium sulfide precipitations from 0.3M acid and several indium hydroxide precipitations from ammonia. Two small aliquots of both the cadmium and indium fractions were mounted on cover slips, and the growth and/or decay followed on the nucleometer. Chemical yields were determined by weighing cadmium sulfate or indium oxide from large aliquots. Extrapolation of the indium decay curves and the sum of cadmium and indium curves, corrected for chemical vield, to separation time showed, after subtraction of the 43-day tail from the cadmium plus indium curves, that 39 percent (39.1 percent, 39.5 percent) of the total radiation detected from old 53-hour Cd¹¹⁵ samples on glass is from In^{115m} .

The final results of the calibration are that in the standard mounting under Scotch Tape on our external beta-proportional counter the 43-day Cd¹¹⁵ is counted with 43 percent efficiency, the 53-hour Cd¹¹⁵ (in equilibrium with In^{115m}) is counted with 60 percent efficiency, and the 4.5-hour In^{115m} is counted with 27 percent efficiency, assuming all of the 53-hour cadmium decays to the 4.5-hour indium, which seems probable.

The counting rates ordinarily measured were in the range of 1000 to 100,000 counts per minute. (With the counters used, the coincidence correction was about 2 percent at a counting rate of 100,000 per minute.) The initial activities of decay curves were usually in the range of 10,000 to 100,000 counts per minute, and the 43-day tail was usually reached at counting rates of about 1000 counts per minute. Background ran about 80 counts per minute.

III. EXPERIMENTS AND RESULTS

A. Mass Number Assignments

At the time this work was done the assignment of mass 115 to the 43-day cadmium seemed fairly certain, but since no direct evidence of its genetic relationship with the rest of the 115 chain was available, it seemed worthwhile to check the mass assignment. Since then, however, the mass assignment has been verified by means of the n,γ reaction on enriched Cd¹¹⁴ samples.⁴

¹⁸ Acid solutions were evaporated to dryness on a microscope cover slip under an infrared heat lamp, the residue taken up in water, precipitated as sulfide or hydroxide, and evaporated to dryness. Cadmium was precipitated as the sulfide by blowing H_2S over the drop. Indium was precipitated as sulfide in the same manner or as the hydroxide by inverting a beaker wet with concentrated ammonia over the sample.

¹⁹ J. Hudis (private communication from the Department of Chemistry, Washington University, St. Louis, Missouri).

We bombarded in the 20-Mev betatron beam a sandwich consisting of an ~ 0.002 -inch cadmium foil enriched in Cd¹¹⁶²⁰ inserted between two ~ 0.002 -inch foils of normal cadmium. The whole sandwich was then wrapped in normal cadmium to stop any stray neutrons, which would confuse the issue by producing Cd¹¹⁵ by the n,γ reaction on Cd¹¹⁴. Both the 53-hour and 43-day cadmiums were produced by the reaction Cd¹¹⁶(γ,n)Cd¹¹⁵. Table I gives the specific activities obtained, expressed in counts per minute per milligram of Cd¹¹⁶, and corrected for small differences in self-absorption of the samples. No chemistry was done on the samples—the foils were merely mounted under Cellophane and counted.

The constancy of the specific activities confirms the assignment of both the 53-hour and the 43-day cadmium activities to mass 115.

From the foregoing data and the previously discussed counting efficiencies it was calculated that the ratio of γ,n cross sections for the formation of the two isomers in a 20-Mev betatron beam is about four in favor of the 53-hour isomer.

B. Nuclear Properties

21-minute Ag¹¹⁵

We have determined the half-life of Ag¹¹⁵ by observing the growth of cadmium as a function of time (see the following section on genetic relationships). Our value, 21 minutes, is in agreement with the values of 22 minutes determined by Turkevich¹ and 20 minutes determined by Duffield and Knight.² We made no studies of the radiation from the silver.

53-hour Cd115

On the basis of ten determinations, usually valid over about five half-lives, we obtained a best value of 53.0 ± 1 hours for the half-life. The samples used were obtained from n,γ and γ,n reactions and from thermal and 14-Mev fission. In each case it was necessary to resolve the decay curve by subtracting the 43-day tail from the total observed activity.

Using two scintillation counters in coincidence,²¹ we were able to observe beta-gamma coincidences and to measure absorption curves of the beta-ray and gamma-ray involved. The gamma-ray had a half-thickness in lead of 5.1 g/cm², corresponding to an energy of ~ 0.55 Mev. (A small amount of a softer component, probably x-rays and gamma-rays from the indium daughter, was also present but was not coincident with a beta-ray.) The coincident beta-ray had an end point of ~ 150 mg/cm² of Al, corresponding to an energy of

about 0.5 Mev. This end point was difficult to determine with any degree of accuracy because of the presence of the large amount of the harder, noncoincident beta-ray.

A rough calibration of the instrument, using the known radiations of Sc^{46} , indicated that about 15 percent of the decay occurs by way of the soft beta-ray followed by the gamma-ray, the other 85 percent going by way of the 1.10-Mev beta-ray.

The measurements were made on fission-product cadmium which contained only a few percent (in terms of activity) of the 43-day cadmium. Corrections were made for the presence of this isomer and of the 4.5-hour indium daughter.

A measurement made on a sample of the indium daughter showed that none of the coincidences observed were caused by its decay.

These coincidence counting results are in agreement with the results of Mandeville, Scherb, and Keighton.⁷

43-day Cd115

We have not yet determined a precise value for the half-life of this isomer, since our decay curves show a

TABLE I. Specific activities^a of cadmium foils bombarded with 20-Mev betatron x-rays.

Sample	53-hour Cd	43-day Cd
1 (normal)	830	8.8
2 (enriched)	850	8.2
3 (normal)	850	8.9

 $^{\rm a}$ Counts per minute per mg of Cd^{116} at the end of an intermittent bombardment conducted during a 7.0-hour interval.

long-lived component (see Sec. IIID) and have not been resolved. The initial slopes are consistent with the previously reported 43-day and 44-day values,³ and we have used the generally accepted value of 43 days in all calculations.

An absorption curve indicates that if there is a gamma-component to the radiation, it is extremely weak. Using a beta-proportional counter, the betacounting rate decreased by a factor of 4000 before the gamma-ray tail was reached. Assuming a relative counting efficiency of about 1 percent for the gammaradiation, it appears that no more than a few percent of the beta-particles are accompanied by a gamma-ray. This result is consistent with data, published after the completion of our work, that showed that \sim 1 percent or less of the 43-day cadmium beta-particles are accompanied by gamma-rays.^{10,12} The beta-end point is \sim 700 mg/cm² in Al, corresponding to an energy of \sim 1.5 Mev.

4.5-hour In115m

Our decay curves show a half-life of 4.5 hours, in agreement with other workers.^{3,11}

Since the counting efficiency of the indium is 65 percent of the efficiency of the 53-hour cadmium (in an

 $^{^{20}}$ The enriched Cd^{,16} used in this investigation was supplied by Carbide and Carbon Chemicals Corporation, Y-12 Plant, Oak Ridge, Tennessee, and was obtained on allocation from the Isotopes Division of the AEC. The Cd¹¹⁶ content was 71.6 percent as compared to 7.6 percent for normal cadmium.

²¹ We thank Dr. Arthur Freedman, without whose assistance and equipment these measurements could not have been made.

internal beta-proportional counter), it appears that over half of the indium decays are accompanied by either an internal conversion electron or a beta-particle. This is consistent with the reported facts that about 6 percent of the indium decays by beta-emission, and that about half of the isomeric transitions are by internal conversion.

C. Genetic Relationships

Growth of 53-hour and 43-day cadmium from 21-minute silver

A sample of about one gram of U^{235} (as uranyl nitrate) contained in a dilute nitric acid solution was irradiated in the Los Alamos homogeneous reactor (water boiler) for 20 minutes. After irradiation 20 mg of silver and 20 mg of cadmium were added and AgCl was precipitated by addition of HCl plus HNO₃. The precipitate was filtered, washed with 0.25*M* NH₄NO₃, and dissolved in ammonia. Twenty milligrams of cadmium were added to the ammonia solution, which was then acidified with HNO₃ plus HCl. The resulting AgCl precipitate was filtered and washed with NH₄NO₃. (Total time from end of bombardment to end of second filtration plus wash was 16.3 minutes, including a 10-minute wait to allow some of the short-lived fission products to decay.)

A previous tracer experiment had shown that less than 0.01 percent of the cadmium originally present was carried through this procedure. The AgCl precipitate at this stage was, therefore, free of cadmium.

The precipitate was dissolved in ammonia, and a known amount of cadmium was added. A known time after this last precipitation, AgCl was again precipitated by addition of HCl plus HNO₃. The precipitate was filtered and washed with NH_4NO_3 . The time between filtrations (measured from the times at which the last drops of filtrate went through the funnel) was 20.5 minutes.

The procedure of dissolving the precipitate, adding cadmium, and reprecipitating AgCl was repeated, this time with a lapse of 136 minutes between filtrations. In 136 minutes only 1 percent of the original amount of 21-minute silver would be left.

The cadmium in the two filtrates containing the active cadmium which had grown from silver during the known time intervals was carried through the standard purification and mounting procedures. Both 53-hour and 43-day cadmium activities were present in each sample in amounts consistent with a half-life of

TABLE II. Thermal neutron fission yields of the Cd¹¹⁵ isomers.

Fission yield, percent		Ratio	
Aliquot	53-hour	43-day	53-hour/43-day
1	9.8×10 ⁻³	7.1×10^{-4}	
$\tilde{2}$	9.8×10 ⁻³	7.2×10^{-4}	
Av	9.8×10^{-3}	7.1×10^{-4}	13.8

21 minutes for the silver parent. (The actual half-life values calculated were 20.5 minutes from the 53-hour activities in the two samples and 20.6 minutes from the 43-day activities.)

The relative amounts of 53-hour and 43-day cadmium in each sample show that 9.2 percent of the silver decays to the 43-day state and 90.8 percent decays to the 53-hour state.

Growth of 4.5-hour In^{115m} from 53-hour cadmium

The 4.5-hour indium is known to be the product of at least some of the 53-hour cadmium disintegrations.^{5,6} The energies^{7,13} involved in the two known decay paths of 53-hour Cd¹¹⁵ indicate that both of these principal paths lead to the same state of In¹¹⁵. That this state is the 4.5-hour excited isomeric state is shown by the comparable counting efficiency of this isomer and its 53-hour cadmium parent.

Growth of In^{115m} from 43-day Cd¹¹⁵

The growth of 4.5-hour indium from the 43-day cadmium had not been reported at the time the following work was done. Since then it has been reported¹² that approximately 0.007 percent of the 43-day cadmium decays to In^{115m} .

By means of a chemical separation involving three precipitations of cadmium sulfide from 0.3M HCl, alternating with three precipitations of indium hydroxide from ammoniacal solution, cadmium holdback carrier being added after each cadmium sulfide step, it was possible to separate indium from large amounts ($\sim 10^6$ disintegrations per minute) of 43-day cadmium. The final indium fraction was essentially inactive $(\sim 10 \text{ c/m} \text{ above a background of } 80)$ and contained 79 percent of the original indium carrier added. The indium carrier had been added to an acid solution of cadmium tracer the day before the separation was carried out in order to prevent the loss of carrier-free indium by adsorption on the walls of the vessel, radiocolloid formation, etc. The cadmium used had been produced by fission of U²³⁵ and carefully purified.

Calculations involving the counting data and counting efficiencies show that less than 0.02 percent of the 43-day cadmium decays to the excited state of indium, either directly by beta-particle emission or indirectly by isomeric transition to the 53-hour cadmium. Considering all the evidence, it appears that the decay of the 43-day cadmium involves principally a single betaray, the product being the ground state of In^{115} .

D. Total Fission Yields; Thermal Neutrons

The thermal neutron fission yields of the Cd¹¹⁵ isomers were determined by purifying two aliquots of a U²³⁵ solution that had been irradiated in the water boiler for two hours and following the decay of the samples on an external beta-proportional counter. The number of fissions that occurred in the solution was

calculated from the Mo⁹⁹ activity determined²² in an aliquot of the solution, the relationship between the Mo⁹⁹ activity and the number of fissions having been previously determined by irradiation of U²³⁵ samples in a double fission chamber in the thermal columns of the water boiler and fast reactor.23 Results are summarized in Table II.

The decay of a sample of 43-day cadmium formed by thermal-neutron fission of U235 has been followed for eleven months. The decay curve shows definite curvature. A year after the irradiation, about 35 percent of the activity of the sample was caused by a component of half-life greater than 43 days. This estimate was made by drawing a line of 43-day half-time through the early points of the curve. The long-lived component was shown to be an isotope of cadmium by repurifying the sample by the Glendenin procedure¹⁶ and finding the specific activity had not changed. It seems probable that this long-lived cadmium isotope is the 3.5-year Cd^{113m}.^{24,25} If it is, its yield in U²³⁵ thermal-neutron fission is approximately 5×10^{-5} percent. Formation by neutron capture in cadmium impurity would have required about 75 mg of cadmium in the uranium (~ 2 percent impurity) calculated from Cassidy's²⁴ activation cross section of 0.02 barn. This amount of cadmium would have shown up in a chemical yield greater than 100 percent, whereas a normal 80 percent yield was obtained based on 20 mg of added cadmium carrier.

E. Independent Fission Yields; Thermal Neutrons

The fission yield of the 21-minute Ag115 and the independent fission yields of the Cd115 isomers were determined in duplicate runs performed as follows. Crystals of U^{235} nitrate were irradiated 10.1 minutes (timed from half-power) in the thermal column of the fast reactor and quickly dissolved in 10 ml of 0.7MHNO₃ containing 40 mg each of silver, cadmium, and indium²⁶ carriers as nitrates. The solution was split approximately in half. One-half was added to 7 ml of hot 1.7M HNO₃ and allowed to stand in hot water for 30 seconds, 1 ml of 6M HCl was added, and the silver chloride filtered onto a fine fritted funnel and washed with 5 ml of 0.25M ammonium nitrate. The total time required for the precipitation, filtration, and wash was one minute; the separation time was taken as the end of the filtration, which was about midway between the start of the precipitation and the end of the wash. The time interval between the end of the irradiation and

TABLE III. Thermal neutron fission yield of 21-minute Ag115 and the "independent" fission yields of the Cd115 isomers.

Run	Total fission yield	"Independ	lent" fission
	(percent) of	yields ((percent)
	21-minute Ag ¹¹⁵	53-hr Cd ¹¹⁵	43-day Cd ¹¹⁵
I	7.7×10^{-3}	2.8×10^{-3}	${<}^{2 imes10^{-5}}_{<2 imes10^{-5}}$
II	7.8×10^{-3}	2.6×10^{-3}	
Av	7.8×10^{-3}	2.7×10^{-3}	

the silver-cadmium separation was 5.1 minutes in run I and 6.2 minutes in run II. The efficiency of this silvercadmium separation procedure had been tested previously with cadmium tracer, and it was found that only ~ 0.2 percent of the cadmium remained with the silver chloride. Later, the filtrate was worked up for cadmium, the activities of the cadmium isomers in the resulting sample, corrected for growth from the 21-minute silver before the separation, being a measure of their independent fission yields.

The silver chloride precipitate was dissolved in 6Mammonia containing 20 mg of cadmium carrier. This solution was allowed to stand three hours to allow all the 21-minute Ag¹¹⁵ to decay, then silver chloride was reprecipitated, dried, and weighed for the chemical vield determination, and the cadmium in the filtrate was purified by the standard cadmium purification procedure. The activity of the 53-hour cadmium in the resulting sample was a measure of the fission yield of the 21-minute silver. The ratio of the 53-hour and 43-day activities gave a 9.0 percent value for the fraction of the 21-minute silver decaying to the 43-day cadmium, in good agreement with the 9.2 percent value we had determined previously.

The other half of the irradiated U²³⁵ solution was treated with cold, saturated H₂S solution, and the silver and cadmium sulfides filtered onto a fine fritted funnel containing Celite filter aid. (This step was taken to separate cadmium and silver from indium.²⁶) The sulfides were dissolved in concentrated HCl, and the solution allowed to stand three hours to allow silver to decay. Then the solution was evaporated to dryness, and the residue leached with 0.2M HCl to remove the cadmium from the silver chloride. The cadmium was purified by the usual procedure. The 53-hour activity, corrected for chemical yield, was used to calculate the number of fissions that had occurred in the sample, the 53-hour cadmium total fission yield having been previously determined (see preceding section).

Use of the counting and chemical yield data, the irradiation and separation times, and the decay constants of the 21-minute Ag¹¹⁵, 53-hour Cd¹¹⁵, and 43-day Cd¹¹⁵ in the standard growth and decay equations along with the branching ratio of the Ag¹¹⁵ and the total fission yield of the 53-hour Cd¹¹⁵ enabled us to calculate the fission yield of 21-minute Ag¹¹⁵ and the independent fission yields of both cadmium isomers. The results of the calculations are summarized in Table III.

²² We thank C. O. Minkkinen for determining the Mo⁹⁹ activity

for us. ²³ We thank R. W. Spence and members of his group for making this information available to us. ²⁴ J. M. Cassidy, Phys. Rev. 83, 483 (1951). ²⁵ Carss, Gum, and Pool, Phys. Rev. 80, 1028 (1950).

²⁶ It was originally intended to determine the independent fission yield of In^{115m}. However, the 4.5-hour period was masked in the purified samples by the 117-minute In¹¹⁷ and other fission products not removed in the purification.



FIG. 1. Genetic relationships of the mass-115 decay chain.

It should be pointed out that the difference between the 53-hour cadmium total fission yield and the 21minute silver total fission yield (corrected for branching) is 2.8×10^{-3} percent, in agreement with the directly determined value for the independent fission yield of 53-day cadmium listed in Table III. A similar calculation for the 43-day cadmium indicates that the independent fission yield is essentially zero, in agreement with the value in Table III.

F. Total Fission Yields; 14-Mev Neutrons

The yields of the cadmium isomers from U^{235} fission induced by 14-Mev neutrons were determined by purifying and counting cadmium samples from two aliquots of a solution made by dissolving a U^{235} button that had been irradiated directly behind the tritium target of the Cockroft-Walton machine. The number of fissions that occurred was calculated from the Mo^{99} activity determined²² in an aliquot of the solution. It was assumed that the 14-Mev neutron fission yield of Mo^{99} is 90 percent of the thermal neutron fission yield. This assumption was made in order to compensate for rise

TABLE IV. 14-Mev neutron fission yields of the Cd¹¹⁵ isomers.

Fission yields (percent)			Ratio
Aliquot	53-hour Cd115	43-day Cd115	53-hr/43-day
1	0.99	0.070	
2	0.97	0.068	
$\mathbf{A}\mathbf{v}$	0.98	0.069	14.3

of the valleys of the fission-yield curve with the increase in neutron energy.²³ Results are summarized in Table IV.

G. Relative (n,γ) Yields

A cadmium nitrate solution was irradiated in the water boiler and the decay of a sample followed.

From the relative activities observed, combined with the measured counting efficiencies, it was calculated that the relative cross sections for the formation of 53-hour and 43-day cadmium were in the ratio of 7.5/1 in favor of the 53-hour isomer.

IV. DISCUSSION

Present knowledge of the genetic relationships within the mass-115 decay chain, including contributions presented in this paper, are summarized in Fig. 1. The results of our determinations of the fission yields for members of this chain are summarized in Table V.

Since it is not entirely certain which of the two cadmium isomers is the upper state, it is necessary to

TABLE V. U²³⁵ fission yields of members of the mass-115 decay chain.

	Thermal neutron fission yield, percent ^a		14-Mev neutron fission yield, percenta	Ratio 14-Mev to thermal
Nuclide	total	independent	(total)	yield
21-min Ag ¹¹⁵ 53-hr Cd ¹¹⁵ 43-day Cd ¹¹⁵	7.8×10^{-3} 9.8×10^{-3} 7.1×10^{-4}	2.7×10^{-3} < 2×10^{-5}	0.98 0.069	100 97

^a The estimated error because of inaccuracies in sample mounting and counting and chemical yield determinations is about 3 percent. This is the error in the relative values of the thermal neutron fission yields of the 21-minute Ag¹¹⁵ and the 53-hour Cd¹¹⁵, both total and independent, since all three values were obtained from activity measurements of the 53-hour Cd¹¹⁵. The error in the absolute values of the fission yields is larger, perhaps about 10 percent, because the absolute beta-counting calibration is involved.

consider each of the two isomeric transition possibilities. The fact that the 4.5-hour In^{115m} grows from the 53-hour but not the 43-day isomer clearly shows that the 43-day state does not decay to the 53-hour state. The evidence against the occurrence of the reverse transition is less direct and has to do with the independent fission formation of 28 percent of the 53-hour cadmium. If some of the 53-hour cadmium decayed to the 43-day isomer, some of the independent fission yield of the former would be reflected as an apparent independent yield of the latter. Since the ratio of the experimentally determined independent fission yields of the two cadmium isomers is >100, <1 percent of the 53-hour decays to the 43-day state.

It seems unlikely that the relatively large independent fission yield of the 53-hour Cd¹¹⁵ is caused by its formation as a primary fission product. It seems more reasonable to postulate the existence of an earlier member of the chain, such as a short-lived Ag¹¹⁵ isomer, which decays to 53-hour cadmium. From the constancy of the ratio of the 53-hour to 43-day cadmium activities in the two fractions milked from the 21-minute Ag¹¹⁵ for the branching ratio determination, an upper limit of 3 minutes can be placed on the half-life of this postulated short-lived Ag¹¹⁵ isomer.

The fact that the 14-Mev neutron fission yield of

cadmium isomer. The common ancestor could also be a short-lived palladium isotope which branch decays to the Ag¹¹⁵ isomers.

V. ACKNOWLEDGMENTS

each of the cadmium isomers is one hundred times the yield from thermal neutrons indicates that both isomers It is a pleasure to thank Dr. R. W. Spence and the have a common ancestor whose yield increases the members of his group for their hospitality and assistance hundred-fold. This common ancestor could be the shortduring the summer of 1950 when this work was carried lived Ag¹¹⁵ isomer (postulated in the previous paraout. We also wish to express our appreciation to the graph), which branches, 75 percent undergoing isomeric groups responsible for the operation of the water boiler, transition to the 21-minute silver and 25 percent fast reactor, Cockroft-Walton, and betatron for making decaying by beta-emission directly to the 53-hour their facilities available to us.

PHYSICAL REVIEW

VOLUME 85, NUMBER 4

FEBRUARY 15, 1952

Neutron Yield from the Nuclear Photoeffect

J. S. LEVINGER Louisiana State University, Baton Rouge, Louisiana

AND

H. A. BETHE Cornell University, Ithaca, New York (Received October 30, 1951)

The yields of neutrons produced in photonuclear reactions by a bremsstrahlung-photon-spectrum are analyzed in terms of the multiplicity of neutron production. Approximating the neutron multiplicity as proportional to the photon energy, we derive the integrated photonuclear cross section from the experimental neutron yields. The integrated cross section from copper to bismuth is $\int_0^{\infty} \sigma dW = 0.14 N Z/A$. (Uranium has a neutron yield 35 percent higher than given by this relation, probably due to photofission.) This relation has the correct form, but a somewhat higher absolute value than the theoretical relation $\int_0^\infty \sigma dW = 0.060 (NZ/A) (1+0.8x)$, where x is the fraction of exchange force.

I. INTRODUCTION

N a previous paper,¹ we calculated the integrated photonuclear cross section, and found satisfactory agreement between our theoretical result and preliminary experiments.²⁻⁵ Since that time very many new experimental results have appeared, some of which are in apparent contradiction with theory.

First, let us summarize the theoretical results of our previous paper,¹ and the assumptions behind them. The summed oscillator strength $\sum_{n} f_{0n}$ for electric dipole transitions by a nucleus containing N neutrons and Z protons is

$$\sum_{n} f_{0n} = (NZ/A)(1+0.8x). \tag{1}$$

Here x is the fraction of the neutron-proton force that has an exchange character. High energy neutron-proton scattering experiments⁶ indicate the value $x=\frac{1}{2}$. The

J. L. Lawson and M. L. Perlman, Phys. Rev. 74, 1190 (1948).
 G. C. Baldwin and G. S. Klaiber, Phys. Rev. 73, 1156 (1948).
 M. L. Perlman and G. Friedlander, Phys. Rev. 74, 442 (1948);

and Phys. Rev. 75, 988 (1949). ⁵ E. R. Gaerttner and M. L. Yeater, Phys. Rev. 77, 714 (1950).

cross section σ for photon absorption is proportional to the oscillator strength f. The cross section for photon absorption integrated over all photon energies W is given by

$$\int_{0}^{\infty} \sigma dW = (2\pi^{2}e^{2}\hbar/Mc)\sum_{n} f_{0n}$$

= 0.060(NZ/A)(1+0.8x)
= 0.015A(1+0.8x) Mev-barns. (2)

The last numerical result is for the case N = Z = A/2.

The sum rule $\sum_{n} f_{0n} = NZ/A$ is completely independent of any nuclear model. The modification shown by the 0.8x term in Eq. (1) occurs for any potential that does not commute with the position, such as an exchange potential or a velocity dependent potential. This modification was first found by Feenberg.⁷ Recently Austern and Sachs⁸ have considered the general problem of modifications for all multipole transitions due to potentials that do not commute with position. The coefficient 0.8 in Eq. (1) is based on the

¹ J. S. Levinger and H. A. Bethe, Phys. Rev. 78, 115 (1950).

⁶ Hadley, Kelly, Leith, Segrè, Wiegand, and York, Phys. Rev. **75**, 351 (1949); R. S. Christian and E. W. Hart, Phys. Rev. **77**, 441 (1950); Kelly, Leith, Segrè, and Wiegand, Phys. Rev. 79, 96 (1950).

⁷ E. Feenberg, Phys. Rev. 49, 328 (1936).

⁸ N. Austern and R. G. Sachs, Phys. Rev. 81, 710 (1951).