Fission of Bismuth with 15- and 22-Mev Deuterons*

A. W. FAIRHALL
Department of Chemistry, University of Washington, Seattle, Washington
(Received February 27, 1956)

Radiochemical studies of bismuth bombarded with 15- and 22-Mev deuterons give evidence of fission product species, clearly distinguishable from species arising from activation of target impurities. The mass-yield curve of fission products from bismuth bombarded with 22-Mev deuterons is unlike the double-humped curves of the heavier elements ($Z \geqslant 90$) excited to comparable energies: it is single humped, symmetric about mass 103.5 and quite narrow. The width of the curve at half-maximum is only 17 mass units. The cross sections for fission of bismuth by 20-Mev deuterons is estimated to be about 1×10^{-29} cm^2, and for 15-Mev deuterons about 4×10^{-31} cm^2. The competition between fission and neutron evaporation in high-energy fission is discussed.

I. INTRODUCTION

ELEMENTS of atomic number $\geqslant 90$ (thorium) are observed to fission rather readily, either spontaneously or upon bombardment with very moderate energy particles. Little is known about the fission behavior of elements between thorium and bismuth ($Z=83$) because these elements are all radioactive, with short half-lives. With the exception of radium, which is unpleasant to handle, only the minutest quantities of these elements could be available for fission studies.

Elements from bismuth on down to quite light nuclei have been observed to fission when bombarded with high-energy particles, or quanta, of energy in excess of 50 Mev.[1–8] At the time the present studies were begun, the few fission excitation functions of bismuth which had been measured down to energies of the order of 50 Mev implied that the fission cross section is small for deuteron energies of about 20 Mev.[7,8] However, it was thought to be of interest to look for the fission of bismuth at this relatively low energy to see whether or not the mass distribution of fission products showed the asymmetry which is characteristic of the heavy elements ($Z \geqslant 90$) at comparable excitation energies. The fission of bismuth is predicted to be asymmetric by a theory of fission which shows considerable promise of interpreting mass distributions of the heaviest elements.[9]

A second reason for choosing to study the fission of bismuth at as low an energy as possible was the expectation that the fissioning species would be well defined, and interpretations therefore made easier. This would be in contrast with the experiments at high energy. There it is believed that the emission of various numbers of neutrons may precede the fission event,[7] so that those results may arise from a superposition of fissions of different nuclear species at different excitation energies.

The present paper gives the results of some experiments on the fission of bismuth with 15- and 22-Mev deuterons.

II. EXPERIMENTAL PROCEDURE

Highest purity (>99.99%) bismuth metal, spectroscopic standard material, was used as the target. The principal impurities detected spectroscopically were silver and copper, with a trace of silicon and lead. A host of elements were reported to be undetectable. Impurities in the target are very undesirable since they can be activated by the deuteron beam to give activities which might mask the fission product activities being sought. Uranium or thorium impurity would obviously be particularly undesirable.

Likewise the material that is used to hold the bismuth target during bombardment has to be especially pure. Activation analysis with lower energy deuterons indicated that the bismuth was free of thorium and uranium, and that the super-pure aluminum[10] foil and target holders used in the bombardments gave no interfering activities.

The bismuth was bombarded in the form of coarse powder held into grooves in a water-cooled pure aluminum target plate by thin aluminum foil. In addition to water cooling helium cooling was also necessary in order to prevent the bismuth particles from melting. In each experiment the targets, which were thick to the deuteron beam, were bombarded for one hour at a deuteron flux of 3 microamperes/cm^2. The first experiments were performed at the Massachusetts Institute of Technology Cyclotron using deuterons of incident energy 15.5 Mev, and a target of 3.0-cm^2 area. These

* Supported in part by the U. S. Atomic Energy Commission.
[1] A review of work prior to June, 1952 is given by R. W. Spence and G. P. Ford, in *Annual Reviews of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. II, p. 399.
[2] W. F. Biller, University of California Radiation Laboratory Report UCRL-2067, December, 1952 (unpublished).
[3] P. Kruger and N. Sugarman, Phys. Rev. **99**, 1459 (1955).
[4] L. G. Jodra and N. Sugarman, Phys. Rev. **99**, 1470 (1955).
[5] N. A. Perfilov and V. I. Ostroumov, Proc. Acad. Sci. U.S.S.R. **103**, 227 (1955).
[6] E. L. Kelly and C. Wiegand, Phys. Rev. **73**, 1135 (1948).
[7] R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).
[8] J. Jungerman, Phys. Rev. **79**, 623 (1950).
[9] P. Fong, Phys. Rev. **89**, 332 (1953); **102**, 434 (1956).
[10] We are very grateful to Mr. J. A. Nock, Jr., of the Aluminum Research Laboratories, Aluminum Company of America, for supplying the very high purity (>99.99%) aluminum used in the experiments at 22 Mev.

TABLE I. Yields of fission products of bismuth bombarded with 22-Mev deuterons.

Nuclide	Fission yield (%)
Sr^{91}	2.0
Sr^{92}	2.4 ±0.3[a]
	2.1[b]
Y^{93} [c]	3.6
Zr^{97}	6.5 ±1.4
Mo^{99}	10.2 ±0.6
Ru^{105}	10.0 ±1.2
Pd^{109}	6.4 ±0.6
Pd^{111m}	1.6 ±0.08
Pd^{112}	6.0 ±0.5
Ag^{111} [d]	5.1 ±1.9
Ag^{113}	4.5 ±0.08
Sb^{127}	<0.13
Ba^{139}	<0.06±0.02

[a] From observed amount of Y^{92} daughter.
[b] By direct observation.
[c] The fission yields of the other fission products are measured relative to this nuclide. The fission yields of all mass numbers integrate to 200%.
[d] From decay of Pd^{111} and independent formation.

experiments were later repeated at the University of Washington Cyclotron using deuterons of 22 Mev and a target of 1.1-cm² area. Following bombardment, the target was processed radiochemically for specific elements. The chemical separations which were used are outlined in the Appendix.

Each sample of separated element was mounted on Scotch tape in a 1-inch hole in a sample mounting card and covered with thin (2 mg/cm²) polystyrene. In the experiments at the higher energy the counting was done in a set of 4 end-window G-M counters with identical geometries. The detection efficiencies of the four counters differed by less than 5%. A set of 3 counters, having a slightly different shelf assembly but otherwise essentially identical with the counting equipment used in the higher energy runs, were used in the experiments at M.I.T. The decay curves were measured and analyzed in the usual way, and corresponding saturation counting rates were computed. Corrections were applied for chemical yield, for counter detection efficiency, for absorption of the radiations by the sample covering, air, and counter window,[11] and for self-scattering and self-absorption of the radiations within the sample.[12] The species 10-hr Y^{93} was found to be a convenient nuclide to use as a standard to which other yields were normalized. It was separated in each bombardment and assigned an arbitrary fission yield until the complete mass-yield curve could be defined. As is customary with mass-yield curves, the fission yield curve was then normalized to a total yield of 200% for all fission products.

III. EXPERIMENTAL RESULTS

A detailed survey of all the elements examined using 15-Mev deuterons is given in the appendix. Some showed activities most likely coming from activation of impurities. Others could be produced only by a fission process.

The results of experiments using 22-Mev deuterons are given in Table I, the limits of error being the standard deviations of the measurements in replicate runs. Each nuclide, except Sr^{91}, Sr^{92}, and Sb^{127}, which were measured quantitatively only once, was measured at least twice, and some were measured several times. No attempt has been made to assess systematic errors, although these are felt to be small where no chemical complications would be expected. The difficulty of measuring ruthenium fission yields is well known, and the fission yield of Ru^{105} might be in error on this account.

The fission yields of Table I represent cumulative yields of each mass chain up to the species separated radiochemically. They do not include species formed independently which lie closer to stability than the nuclides examined. The tabulated values therefore represent lower limits of the yields of all nuclides of that particular mass. However, it is likely that the measured yields are close to the total mass yields of each mass number listed if the charge distribution of primary fragments follows the pattern of equal charge displacement for light and heavy fragments observed with other kinds of fission.[13] On the reasonable assumption that two neutrons are lost from each primary fission fragment (see below), the primary species from which

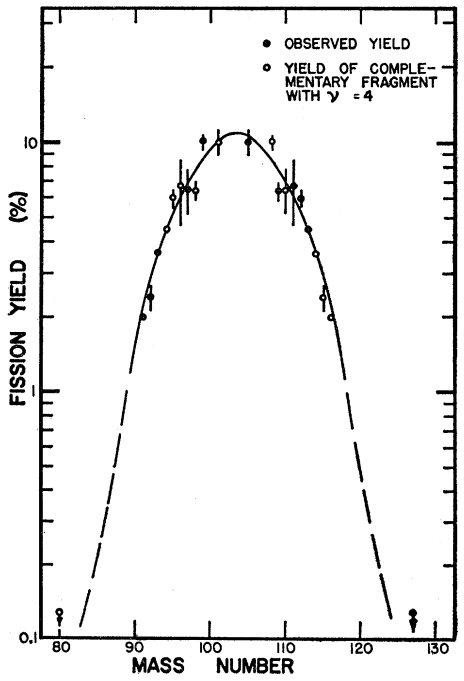

FIG. 1. Fission yield *vs* mass number for bismuth bombarded with 22-Mev deuterons. The fissioning species is probably Po^{211} excited to about 27 Mev.

[11] C. D. Coryell and N. Sugarman, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, Part I.
[12] W. E. Nervik and P. C. Stevenson, Nucleonics **10**, 18 (1952).
[13] See reference 11, Paper No. 52.

each nuclide in Table I comes all lie closer to stability than does the complementary fragment. The species listed will therefore come predominantly from the decay of precursors, and the formation of unobserved species lying closer to stability must be small.

An alternative assumption would be to postulate that fission takes place with unchanged charge distribution. Again this assumption would lead to the same conclusion. The fact that the yield of mass 92 calculated from the observed amount of Y^{92} is closely the same as the yield of Sr^{92} observed separately, and the absence of detectable independent formation of 3.2-hr Ag^{112} are considered good evidence that the yields given are close to total mass yield for all fragments having the observed mass.

The fission yields of Table I are plotted in Fig. 1, with reflected points being assigned on the assumptions that the energy involved is sufficient to evaporate four neutrons from the nascent fission fragments, and the fissioning species is Po^{211}. Bismuth 209 bombarded with deuterons[14] will give either Po^{211}, if the deuteron is captured whole, or Bi^{210} (or possibly Po^{210}) if the deuteron is stripped and only one nucleon enters the nucleus. Both reactions undoubtedly occur, with much more energy deposited in the compound nucleus in the former case than in the latter. Since the fission process is so energy-sensitive in this energy region, the fission of Po^{211} will undoubtedly predominate. The energy deposited in Po^{211} by a 20-Mev deuteron impinging on Bi^{209} may be readily calculated from the nuclear masses of the species involved.[15] The value obtained is found to be of the order of 27 Mev.

The mass distribution shown in Fig. 1 is evidently single-humped, symmetric about mass 103.5, and has no pronounced fine structure. The width of the curve at half the maximum yield is 17 mass units, and at about 1% of the maximum yield the width may be estimated to be about 40 mass units.

IV. CROSS SECTION FOR FISSION

The computation of the cross section for fission of bismuth by a deuteron of given energy is difficult to make using the data from these experiments because of the thick targets which were used. However, the 50-fold increase in observed counting rate in the runs at 22 Mev over those at 15.5 Mev indicated that the cross section for fission is varying very rapidly in this energy region. Assuming an average effective deuteron energy of 20 Mev for the experiments at the higher energy, an estimation of the total fission cross section gives a value of 1×10^{-29} cm^2. In the experiments at the lower energy if the average effective deuteron energy is taken to be 15 Mev, a value of 4×10^{-31} cm^2 is obtained for the total fission cross section. These values are probably reliable within a factor of 2 or 3. They are very much smaller than the value of 0.2×10^{-24} cm^2 observed for the fission of bismuth with 190-Mev deuterons.[7]

V. COMPARISON WITH HIGH ENERGY FISSION

The mass distribution displayed in Fig. 1 may be compared with the distribution observed by Goeckermann and Perlman for fission induced by high energy deuterons.[7] Using 190-Mev deuterons they also obtained a single-humped curve, but their distribution includes a much wider range of masses than is observed here. The width of their curve at half-maximum is 40 mass units, and at 1% of the maximum yield covers a range of 80 mass units.

Their distribution peaks at a lower mass number than that of Fig. 1, being centered at about mass 99.

Using lower energy deuterons, Goeckermann and Perlman concluded that the mass-yield curve becomes somewhat narrower at lower energies. A similar observation was made by Sugarman[16] in experiments on the photofission of bismuth using 85-Mev bremsstrahlung.

The mass distribution of fission fragments from the heaviest elements resembles that of bismuth fission when the excitation energies are large. That is, the curves are single-humped and broad.[17–19] With lower bombarding energies, and in particular at excitation energies of 25 to 30 Mev, there is a well-defined asymmetry to the mass-yield curves of these elements.[20–24] Bismuth is up to now unique in having such a narrow symmetric mass distribution at moderate excitation energies.

VI. MODELS OF HIGH-ENERGY FISSION

Fission of Bismuth

The results observed by Goeckermann and Perlman were interpreted by them as representing the fission of the species Po^{199}. They postulated that this species was produced by the reaction $Bi^{209}(d,12n)$, with almost all of the excitation energy brought into the nucleus by

[14] The possibility that the fission products observed in bombardments with deuterons from the University of Washington Cyclotron arise from 44-Mev helium ions contaminating the deuteron beam was eliminated in experiments in which bismuth was bombarded with 44-Mev helium ions. In these bombardments, the yields are about a factor of 10 higher. The contamination of helium ions in the deuteron beam is known to be less than 1%, so that activation by helium ions cannot be responsible for the activities being reported here. In the experiments at M.I.T., no helium ions had been accelerated for many months prior to the bismuth fission studies.

[15] K. T. Bainbridge, *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley and Sons, Inc., New York, 1953), Vol. I, p. 559.

[16] N. Sugarman, Phys. Rev. **79**, 532 (1950).

[17] R. R. O'Connor and G. T. Seaborg, Phys. Rev. **74**, 1189 (1948).

[18] Folger, Stevenson, and Seaborg, Phys. Rev. **98**, 107 (1955).

[19] Vinogradov, Alimarin, Baranov, Lavrukhina, Baranova, Pavlotskaya, Bragina, and Yakovlev, *Conference of the Academy of Sciences of the U.S.S.R. on Peaceful Uses of Atomic Energy, July 1–5, 1955*, Session of the Division of Chemical Sciences (translated by Consultants Bureau, Inc., New York, 1955), Vol. 2, p. 65.

[20] A. S. Newton, Phys. Rev. **75**, 17 (1949).

[21] Fowler, Jones, and Paehler, Phys. Rev. **87**, 174 (1952).

[22] H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).

[23] Turkevich, Niday, and Tompkins, Phys. Rev. **89**, 552 (1953).

[24] R. A. Schmitt and N. Sugarman, Phys. Rev. **95**, 1260 (1954).

the deuteron being dissipated in the evaporation of neutrons. The species Po^{199} has a value of Z^2/A comparable with that of U^{238}, and presumably fissions rather readily at low excitation energies. In order to account for the observed fission products it was assumed that the Po^{199} fissioned symmetrically and with unchanged charge distribution. They pointed out, however, that the same experimental results would be observed if symmetric fission took place in the highly excited compound nucleus, preceding or in competition with neutron evaporation, and neutrons were subsequently lost from each of the highly excited fission fragments.

The possibility that the present results represent the fission of species like Po^{208} or Po^{209}, formed with necessarily low excitation energy in neutron boil-off from Po^{211}, seems very unlikely. The slight increase in fissionability of Po^{208} over Po^{211}, corresponding to a difference in Z^2/A of only 1.5%, could scarcely be comparable with the effect of about 20 Mev more excitation energy in a species only three neutrons heavier.

The observation of the fission of polonium nuclei excited to about 27 Mev, formed by bombardment of bismuth with about 20-Mev deuterons, suggests that the results of Goeckermann and Perlman may indeed represent fission taking place at high energy in competition with neutron emission. The observed behavior at higher energies suggests, as one possibility, that the probability of fission of nuclei in the region of bismuth increases with increasing excitation energy; and that the mass distribution of fission products becomes increasingly broad as the energy of the fissioning nucleus increases.

Another possibility which could account for the breadth of the fission yield curve for fission induced by 190-Mev deuterons would be to postulate that fission taking place at high energy is always symmetric with a rather narrow mass distribution; and that there is an admixture of asymmetric fission of the residual Po^{199} nuclei formed from the evaporation of neutrons in competition with high-energy fission. Approximately equal proportions of symmetric high-energy fissions and asymmetric low-energy fissions would be sufficient to account for the shape of the mass-yield curve of Goeckermann and Perlman. With decreasing excitation energy the asymmetric fissions would be expected to disappear, since fewer neutrons could be evaporated and the residual nuclei would not be so readily fissile with small excitation energy. Thus the mass distribution would become narrower and single-humped at lower energies.

The possibility that the curve obtained by Goeckermann and Perlman represents the superposition of many narrow curves like the one observed here can be eliminated by a consideration of the energetics involved. To account for the lightest fission product species they observed on this basis, it would be necessary to assume a curve centered about mass 80. This would involve the emission of about 50 neutrons, which is energetically impossible. These light fragments must therefore represent very asymmetric fissions. The symmetry of the light and heavy wings of the observed curve suggests that these represent complementary fragments. Since the wings are centered about mass 99, as are the most probable fission fragments, the asymmetric fissions and the symmetric fissions must be associated in some way with the same highly excited compound nucleus, whether or not fission precedes neutron emission.

The observation of Jungerman and Wright that the fission fragments from fission of bismuth induced by 90-Mev neutrons have an energy expected for low-energy fission[25] is not inconsistent with a picture of fission taking place at high energy. The velocity of fission fragments probably arises from Coulomb repulsion and does not reflect the internal energy of the fissioning system to any marked degree.

Fission of the Heaviest Elements

If highly excited nuclear species in the mass region of bismuth fission with a symmetric mass distribution, one would expect a similar behavior for nuclei of the heaviest elements. The increasing probability of symmetric fission in the heaviest elements as the energy of the fission-inducing particle is increased has already been mentioned. At moderate energies, however, asymmetric fission is still the predominant fission mode. With these heavy elements, however, there is an added complication. While the energy of the bombarding particle may give nuclei excited to 25 or 30 Mev, if neutron emission competes with fission, which is the case for excitation energies of 15 to 20 Mev,[22] the resulting species may have excitation energies ranging from 0 to about 20–25 Mev. For these nuclei, the probability of fission at excitation energies greater than about 5 Mev is quite high. Nuclei in this mass region with excitation energies of a few Mev in excess of this amount are observed to give asymmetric mass distributions.[26,27] It may be that all highly excited nuclei that undergo fission have a symmetric mass distribution, but in the case of the heaviest elements the admixture of low-energy asymmetric fissions following neutron evaporation gives the observed shallow asymmetric mass-yield curves. In this view of fission, asymmetric fission is to be associated only with nuclei excited to a few Mev in excess of the fission threshold energy.

The recent observation that radium fissioned with 11-Mev protons gives a triple-humped mass-yield curve,[28] with symmetric fission being much more probable than would be expected on the basis of the systematics pointed out by Fowler, Jones, and Paehler,[21]

[25] J. Jungerman and S. C. Wright, Phys. Rev. **76**, 1112 (1949).
[26] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 52 (1951).
[27] H. G. Richter and C. D. Coryell, Phys. Rev. **95**, 1550 (1954).
[28] R. C. Jensen and A. W. Fairhall (to be published).

is consistent with the above interpretation. The rather narrow central peak of this mass yield curve may be ascribed to the fission of Ac^{227}, formed with about 16 Mev of excitation energy in the capture of an 11-Mev proton in Ra^{226}. The two peaks on either side are ascribed to the fission of Ac^{226}, formed with lower excitation energy in the $Ra^{226}(p,n)$ reaction. The location of these asymmetric peaks fits the prediction of the empirical curve recently noted by Swiatecki.[29] The lower proportion of asymmetric fission, relative to symmetric fission, would reflect the decreasing fissionability of Ac^{226} at low excitation energy compared with uranium nuclei.

Calculations of the energetics of bismuth fission by the theory of Fong[9] predict asymmetry in the mass distribution, with the peaks occurring in the mass regions 70 and 140. It has not been possible, so far, to resolve the discrepancy between theory and experiment.

ACKNOWLEDGMENTS

The author wishes to thank Mr. Earl White and the crew of the M.I.T. Cyclotron and Mr. Ted Morgan and the crew of the University of Washington Cyclotron for assistance in making the numerous bombardments. He is indebted to Professor I. Halpern for many helpful suggestions and stimulating discussions of this work.

APPENDIX

The following elements were examined for fission product activity produced in bombardments of bismuth with 15-Mev deuterons. The chemical separations used are briefly described where they differ from the usual radiochemical procedures.[30] These modifications were introduced because of the presence of a relatively high concentration of bismuth in the solution of the target material. The quantity of each element added as carrier was usually 10 mg.

The activities observed in each element are given, and the most probable source of the activity noted, whether produced by activation of impurities or by a fission process.

In the experiments with 22-Mev deuterons, only those elements were examined (Sr, Y, Zr, Mo, Ru, Pd, Ag, Te from Sb precursors, and Ba) which showed no clear evidence of admixture of activities from activation of impurities. Some of these could come from (d,p) reactions on impurities, but the much higher yields observed at the higher energy, and the fact that the observed yields give so smooth a fission yield curve makes it probable that all the activities tabulated in Table I are fission-produced.

Zinc

Most of the bismuth was precipitated by hydrolysis. Zinc was purified by repeated scavengings with $Fe(OH)_3$ from NaOH solution. The decay curve showed abundant 60-minute and 14-hour species, probably 14-hr Zn^{69m} and 52-min Zn^{69} produced from $Zn^{68}(d,p)$ reactions.

Gallium

Extraction of gallium from 8*M* HCl by dichloroethyl ether removed most of the bismuth. Purification was by bismuth scavengings from acid solution with H_2S, with a final extraction into dichloroethyl ether. The decay curve showed abundant 70-minute, 9.7-hour, and 14-hour components, presumably respectively Ga^{68} and Ga^{66} from $(d,2n)$ reactions on zinc isotopes, and Ga^{72} from (d,p) reaction on Ga^{71} impurity.

Germanium

Germanium was isolated from the same solution as bromine, being distilled from strong HCl solution after the removal of bromine by distillation. Bismuth was scavenged from alkaline solution with H_2S and the germanium was redistilled before final precipitation as the sulfide. The decay curve was mostly 40-hour species, presumably 39.6-hr Ge^{69} from $(d,2n)$ activation of Ga^{69} impurity, with some 80-minute period, presumably 82-min Ge^{75} from (d,p) reaction on germanium impurity. The absence of complementary mass 132 rules out the possibility of this species being a fission product.

Selenium

Selenium was distilled from solution with HBr and precipitated with SO_2. Repeated precipitations with SO_2 from acid solution gave a sample decaying with 57-minute half-life, presumably Se^{81m}. It had a yield about 30-fold greater than Y^{93}. The absence of a fission fragment of mass 127 indicates (d,p) activation of a trace of selenium impurity.

Bromine

$KBrO_3$ was used as bromine carrier, being reduced to Br^- during solution of bismuth metal in dilute HNO_3. The bromine was oxidized to Br_2 with $KMnO_4$ in HNO_3 solution and distilled from solution. Extractions into CCl_4 gave further purification. The decay curve showed very large amounts of 18-minute, 4.5-hour, and 36-hour species, undoubtedly Br^{80}, Br^{80m}, and Br^{82} respectively, all produced by (d,p) reactions on bromine impurity,

Strontium

Strontium and barium were precipitated with fuming nitric acid from bismuth solution, and separated and purified in the usual way, but with an added Bi_2S_3 scavenging step. The decay curve of the strontium fraction had a trace of 5-day period, presumably Bi^{210} contamination, the remaining activity being compatible with a 9.7-hour (Sr^{91}) component and a 2.7-hour parent decaying to a 3.5-hour daughter (Sr^{92}—Y^{92}). About 8 counts/min of 9.7-hour species were observed in replicate runs. Yttrium separated from strontium after a suitable growth period showed the 3.5-hour daughter of 2.7-hr Sr^{92}. There is no conceivable way these species could be produced except by a fission reaction. The possibility that they could be produced from activation of zirconium impurity was ruled out by bombarding some zirconium oxide with deuterons and isolating strontium. From the observed amount of 17-hr Zr^{97} in bombarded bismuth the strontium activity to be expected from this source would be unobservable.

Yttrium

Bismuth was removed by repeated H_2S scavengings and further purification was accomplished by repeated YF_3 precipitations. The decay curve showed two prominent species of 3.5-hour (Y^{92}) and 10-hour (Y^{93}) half-life when a trace of longer-lived species, of half-life indistinguishable from 5 days (Bi^{210}), was subtracted. Activation of zirconium impurity was ruled out as the source of these activities by activation of zirconium with deuterons. Yttrium isolated from bombarded zirconium had only 3.5-hour and 61-hour components. Less than 10% of the observed 3.5-hour yttrium fission activity could come from zirconium impurity.

Zirconium

Zirconium was isolated from the HF solution following YF_3 precipitation, by precipitation of the insoluble barium fluozir-

[29] W. J. Swiatecki, Phys. Rev. **100**, 936 (1955).
[30] See reference 11, Part VI.

conate. Further manipulations followed the usual procedure for zirconium. The decay curve showed a very pure 17-hour species (Zr^{97}) with an initial increase in activity corresponding to the growth of 72-min Nb^{97} daughter. This zirconium species could be a fission product or come from (d,p) activation of zirconium impurity.

Molybdenum

No very good evidence was obtained for any 67-hr Mo^{99} in the runs at 15 Mev, the yield from a 1-hour bombardment being too low to be observable above the ubiquitous background of a few counts per minute of 5-day Bi^{210}. With 22-Mev deuterons, where the yields were much higher, the decay curve indicated impurities in the molybdenum fraction. This difficulty was finally overcome by the following chemical procedure. Molybdenum was extracted from the mixed sulfides, obtained on gassing the original acid solution of bismuth with H_2S, by means of sodium sulfide solution. Molybdenum was precipitated from the basic solution upon acidification. Repeated sulfide scavengings were made, interspersed with precipitations of molybdenum from acid solution with *a*-benzoin oxime reagent. An extraction into ether of the thiocyanate complex from dilute HCl containing stannous chloride served to purify molybdenum completely, and a pure 67-hour period was obtained (Mo^{99}). This could be a fission product or could come from (d,p) reaction on molybdenum impurity.

Ruthenium

The standard perchlorate distillation was used to separate ruthenium. No activity was observed in the ruthenium sample when the bismuth target was dissolved in nitric acid alone. However, when aqua regia was used to dissolve bismuth and the carrier solution added after the bismuth solution had been treated repeatedly with aqua regia there was satisfactory evidence of 4.5-hr Ru^{105}. The decay curve in the 22-Mev runs had a small amount of very long-lived species, compatible with a mixture of 40-day Ru^{103} and 1-yr Ru^{106} (evidence from absorption data of a high-energy β ray), in yields about equal to Ru^{105}. When this background was subtracted the counting data fitted exactly on the curve expected for a 4.5-hour parent decaying to a 36-hour daughter with relative counting efficiencies taken into account. Ru^{105} can be fission-produced or come from (d,p) reaction on ruthenium impurity. Ru^{106} can only be fission-produced.

Palladium

A separation of palladium from bismuth by plating the former on copper powder removed it from most of the bismuth. Iron scavengings from ammoniacal solution gave high purity of the final palladium dimethylglyoxime precipitate. In the experiments using 15-Mev deuterons a few counts per minute of activity were observed to decay with a half-life of about 20 hours (Pd^{112}). The statistical errors for such low counting rates were so large that no breakdown of the decay curve into possible 13.4-hour (Pd^{109}) and 5.5-hour (Pd^{111m}) components could be made. Since Pd^{112} is detected via its 3.2-hr Ag^{112} daughter, failure to observe an initial increase in counting rate implies that some shorter-lived species must be present, or that the observed activity was due to contamination. In the experiments using 22-Mev deuterons, the decay curve was resolved into 21-hour (Pd^{112}), 13.4-hour, and 5.5-hour components. While the latter two can come from (d,p) activation of palladium impurity, Pd^{112} can only be fission-produced. The 5.5-hr Pd^{111m} species is not observed in fission of U^{235} with thermal neutrons. The decay of the Rh^{111} precursor is therefore to the 22-min Pd^{111} ground state. The presently-observed 5.5-hr Pd^{111m} therefore represents independent formation or activation of palladium impurity.

Silver

The separation was by standard methods. The decay curve showed 5.3-hour (Ag^{113}) and 7.5-day (Ag^{111}) components. Rapid separation of silver gave a 22-minute component, but this appeared to be largely β^+ activity, presumably 24-min Ag^{106} from (d,t) reaction on Ag impurity, the principal impurity known to be present in the bismuth target. Deuteron bombardment of cadmium gave principally 3.2-hr Ag^{112} so the observed Ag^{113} is certainly fission-produced.

In the experiments using 22-Mev deuterons, the amount of 3.2-hr Ag^{112} formed from decay of 21-hr Pd^{112}, during bombardment and during the time before silver was separated from the target, was calculated to be about 6.5% of the observed 5.3-hr Ag^{113} activity. This amount of 3.2-hour species would be detected with difficulty in a decay curve consisting mainly of 5.3-hour species. An admixture of 15% of 3.2-hour species would be readily apparent in the decay curve, however, even after 8 hours of decay. Since the decay data, with 7.5-day species subtracted, fell exactly on a 5.3-hour curve over a period of 18 hours, beginning 8 hours after the end of the bombardment, the admixture of 3.2-hour species is certainly less than 15% of the 5.3-hour species. The independent formation of Ag^{112} does not appear to be detectable in these experiments.

Tin

Extraction of mixed sulfides with NaOH removed tin, which was purified by bismuth scavengings from alkaline solution. The tin sample showed abundant 10-day (Sn^{125}), 27-hour (Sn^{121}) and 40-minute (Sn^{123}) periods, consistent with activation by (d,p) reactions of tin impurity.

Antimony

No satisfactory separation of a small amount of antimony from one gram of bismuth was found. A search for the tellurium daughters of 95-hr Sb^{127} and 4.2-hr Sb^{129} showed these two species to be absent in detectable amount.

Tellurium

Tellurium was separated from the bismuth solution remaining after selenium was distilled with HBr. Precipitation with SO_2 accomplished the separation, and the purification of the tellurium was by standard methods. Components of 30-hour (Te^{131}), 9-hour (Te^{127}) and 70-minute (Te^{129}) half-lives, present in copious amount, indicate (d,p) activation of tellurium impurity. Iodine isolated from the purified tellurium showed only the 8-day daughter of Te^{131} and no 2.4-hour daughter of 77-hr Te^{132}.

Barium

The barium fraction separated in the standard way showed a trace of activity which decayed with a half-life of a few days, probably 5-day Bi^{210} contamination. The absence of 85-min Ba^{139} is the best evidence that uranium and thorium are absent in detectable amount, and that $Sr^{91,92}$, Y^{93}, Pd^{112}, and Ag^{113} are bismuth fission products.

Several experiments were tried at the beginning to observe the 23.3-min Th^{233} and 23.5-min U^{239} (d,p) products of Th^{232} and U^{238}, respectively, if these elements were present as impurities. The branching ratio between these products and fission were determined separately. No short period was observed in either thorium or uranium separated from bismuth, although it was difficult to complete the chemical separation in less than an hour. It was concluded that thorium and uranium were absent in detectable amounts from these experiments.