THE
PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 78, NO. 5 JUNE 1, 1950

The Spallation Products of Antimony Irradiated with High Energy Particles*

M. LINDNER** AND I. PERLMAN
Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California
(Received February 11, 1950)

The distribution and yields have been determined for a considerable number of spallation products resulting from the irradiation of antimony with particles in the 100-Mev range. The highest yields were found for nuclei about five mass numbers removed from the target material showing that compound nucleus formation is not an important mechanism in such high energy reactions. Products in lower yields were found which probably resulted from nuclei which had been excited to the full energy of the projectile. A more detailed discussion of the types of reactions responsible for the observed products is given.

1. INTRODUCTION

THE acceleration of charged particles into the hundred million volt range by the Berkeley 184-inch cyclotron has made it possible to extend investigations of induced nuclear transformations into a new energy region. Preliminary results already reported[1–3] indicated that one of the predominant characteristics is the multiplicity of reactions. Reaction products covering a range from the region of the target nucleus down to nuclei about 20 mass units lighter were readily identified and good evidence was obtained for products even considerably further removed. To indicate these reactions in which excited nuclei are degraded by losing one or more nucleons, the term *spallation* has been suggested.

The present study consists of the determination of the radioactive products which result from the irradiation of antimony with 380-Mev helium ions and with deuterons of 190 Mev and lower. Particular attention was paid to the measurement of the yields of the many product nuclei as a function of projectile energy. Early in the course of these investigations a number of activities were encountered that had not been previously reported. It was necessary to characterize these nuclei and some of the results have been published separately.[4,5]

The distribution of reaction products to be discussed below is in keeping with the general features of a description by Serber[6] of the encounter between high energy particles and nuclei. In this high energy range it is no longer possible to assume compound nucleus formation at each nuclear encounter. Instead, the instantaneous reaction is considered to proceed through individual collisions between target and projectile nucleons, the incidence of which may be described in terms of the geometry of the encounter and the mean free path between collisions within the nucleus. As an example of one type of encounter, a deuteron of high energy may graze a nucleus in such a way that the neutron is sheared off by a single collision with a nucleon while the proton does not react with the nucleus. Even the neutron may leave the nucleus again giving up only part of its kinetic energy. In this type of reaction a relatively small amount of the kinetic energy of the deuteron is transferred as excitation energy to the nucleus and the product of the reaction will be within a few mass units of the target nucleus. On the other hand, it is also possible for the high energy deuteron to give up all of its energy to the nucleus. According to Serber, the mean free path of a nucleon of 100-Mev energy in nuclear matter is 4×10^{-13} cm. It gives up about 25 Mev in its first collision, and as its energy becomes degraded its mean free path decreases. As a result, with a moderately large nucleus such as antimony one might expect a measurable yield of reactions in which a 200-Mev deuteron gives up essentially all of its energy

* This work was performed under the auspices of the AEC.
** Now at the Department of Chemistry, State College of Washington, Pullman, Washington.
[1] G. T. Seaborg *et al.*, Phys. Rev. **72**, 739, 740 (1947).
[2] H. H. Hopkins, Jr. and B. B. Cunningham, Phys. Rev. **73**, 1406 (1948).
[3] Miller, Thompson, and Cunningham, Phys. Rev. **74**, 347 (1948).
[4] M. Lindner and I. Perlman, Phys. Rev. **73**, 1124 (1948).
[5] M. Lindner and I. Perlman, Phys. Rev. **73**, 1202 (1948).
[6] R. Serber, Phys. Rev. **72**, 1114 (1947).

resulting in a highly excited nucleus which then boils off a large number of nucleons in dissipating its energy.

In the present study reactions illustrative of the extremes cited were noted as well as intermediate types. It was the purpose of these investigations to obtain a survey of the yields of a wide range of reaction products and to determine how these yields vary with projectile energy.

2. PROCEDURE

Irradiations

The antimony targets consisted of strips of pure antimony metal 1 mm in thickness. When the strip is fastened to the end of a movable probe in the cyclotron tank normal to the direction of the deuteron beam, the deuterons strike a thin segment (1 to 2 mm) along the leading edge. The adjustment of the radial position of the probe allows a variation of particle energy to be obtained. Because deuterons in the energy range studied are not degraded in energy readily, antimony targets 1 millimeter thick could be considered as thin targets.

Radiation Counting Methods

After irradiation the targets were separated into chemical fractions by the procedures to be described. Along with half-life determination, the radioactive species present were identified principally by absorption methods using cylindrical end-window Geiger counter tubes with mica windows of approximately 3 mg/cm^2 thickness.

It is characteristic of irradiations with high energy particles that many neutron-deficient isotopes are formed and, in the region of the periodic table under investigation, many of these decay by orbital electron capture. As a result all chemical fractions contained considerable x-ray activity. In order to aid in identifying beta-particles and electrons in the presence of the many x-rays, beryllium rather than aluminum was used for determining the absorption end-points. Using sufficient beryllium to filter out electrons, the x-rays were usually characterized with the use of aluminum absorbers. Gamma-ray energies beyond the x-ray energy region were determined with lead absorbers.

A crude beta-ray spectrometer was used in distinguishing particles of different sign, and was found particularly useful in the cases in which one type was a minor component of the total activity. Used as a spectrometer, lines of electrons could usually be distinguished from continuous beta-spectra and approximate values for the energies were obtainable.

Chemical Separations

The procedure followed in separating the irradiated target into the several chemical fractions was in general the same for the different bombardments. The fractions separated and examined covered the range from tellurium, atomic number 53, to yttrium, atomic number 39. It was not possible to separate all elements in sufficient degree of purity by rapid methods which would at the same time give good yields. However, pure fractions were obtained for tellurium, antimony, tin, indium, cadmium, silver, palladium, ruthenium, molybdenum, and yttrium. In order to determine the chemical yield of each fraction, a known amount of carrier for each element was added before separation and the final step in each purification was the precipitation of a compound suitable for weighing.

The following outline of the methods of separation indicates the reactions used but is not complete in that, in general, the procedures were repeated one or more times to insure adequate purity.

The irradiated antimony metal was dissolved by covering with concentrated hydrofluoric acid and the dropwise addition of concentrated nitric acid. The solution was diluted, carriers for the elements to be separated were added, and the solution was made 0.1M in HCl. The yttrium and silver were separated as yttrium fluoride and silver chloride. The silver was dissolved from the precipitate by treatment with ammonium hydroxide. The yttrium fluoride was dissolved in nitric and boric acids, removed as yttrium hydroxide upon addition of sodium hydroxide, and then dissolved in hydrochloric acid. The solution was buffered with sodium acetate and sulfide-insoluble activities removed by addition of indium and precipitation of indium sulfide with hydrogen sulfide. The hydrogen sulfide was expelled from the solution and yttrium fluoride precipitated by the addition of hydrofluoric acid after adding inactive elements to stabilize the activities from which the yttrium was to be separated.

The silver was purified by a series of precipitations of silver chloride from nitric acid solutions interspersed with scavenging precipitations of ferric hydroxide from ammoniacal solutions

The supernatant solution remaining from the precipitation of silver and yttrium was divided into two equal portions. From one of these, palladium was precipitated with dimethylglyoxime, after which addition of alpha-benzoinoxime precipitated molybdenum. The solution remaining from these precipitations was fumed with sulfuric acid in order to destroy the organic reagents. Perchloric acid was added and upon boiling, the ruthenium was distilled and trapped in dilute sodium hydroxide solution from which it was precipitated by the addition of alcohol and warming.

The second portion of the solution from the silver and yttrium removal was evaporated almost to dryness and taken up in 2M HCl. Tellurium was reduced to the elemental state by treatment of the hot solution with sulfur dioxide. The excess sulfur dioxide was expelled by heating the supernatant solution and the antimony precipitated with hydrogen sulfide while the solution was maintained near boiling. The hydrogen sulfide was driven off by heating, and the solution was made alkaline by addition of sodium hydroxide. This pre-

cipitated the cadmium and indium, and left the tin in solution. The indium and cadmium were separated by dissolution of the hydroxides in dilute acid and precipitation of the indium with ammonium hydroxide. The cadmium was then recovered by precipitation with hydrogen sulfide. The sodium hydroxide solution containing the tin was made 0.3*M* HCl and the tin removed by hydrogen sulfide treatment.

3. RADIOACTIVE NUCLIDES IDENTIFIED

The isotopic composition of each chemical fraction was determined by combinations of absorption and decay measurements, and in a number of cases through parent-daughter relationships. A brief description of the activities encountered in each fraction follows, with some remarks on the methods used in the resolution. Because of the large number of activities found, references are given only for those which have appeared in the literature recently. Most of the others will be found in the 1948 edition of "Table of Isotopes" by Seaborg and Perlman.[7]

Tellurium Fraction: 143-day Te^{121m}

This isotope appeared as the longest-lived component of the tellurium fraction when counted either without absorbers or through beryllium (electromagnetic radiation) and was followed in all cases through at least one half-life. Identification was made certain by complete absorption curves. The 17-day ground state,[8] Te^{121}, was undoubtedly formed independently, but it could not be resolved readily because of the concomitant presence in good yield of 4.5-day and 6.0-day tellurium periods as well as complications caused by the growth of Te^{121} from Te^{121m}.

4.5-day Te^{119} and 6.0-day Te^{118}

The characteristics and mass number assignments of these activities have been reported previously.[4] The 4.5-day period decays by orbital electron capture; it is characterized by conversion electrons and a hard gamma-ray of 1.6 Mev and is the parent of a 39-hour antimony activity which also decays by orbital electron capture. The 6.0-day tellurium assigned to Te^{118} decays by orbital electron capture and probably shows only x-rays in its decay. It is the parent of Sb^{118}, a 3.5-min. positron-emitter.

When this mixture of isotopes is produced with 190-Mev deuterons on antimony, the activity measured without absorbers is largely that of the 3.5-min. Sb^{118} which serves as an index of the amount of Te^{118} present. When counted through sufficient thickness of beryllium ($\sim$2 g/cm^2) to stop all particles, the electromagnetic radiation is approximately equally divided between the 4.5-day and 6.0-day periods. The yield of Te^{119} is more precisely determined by separating and measuring the amount of 39-hr. Sb^{119} which appears at equilibrium. For elements in this region the *K* x-rays are assumed to be counted with 1 percent efficiency under the conditions employed.

2.5-hr. Te

This activity reported previously[4] is a positron-emitter which has not been otherwise characterized. It probably has a mass number of 117 or less since it appears in measurable yield only when antimony is irradiated with deuterons of energy 75 Mev or higher.

Antimony Fraction: Short-lived Antimony

Activity variously resolved in the range 3 to 5 hours appeared in the antimony fraction following all irradiations using deuterons of 75 Mev or greater. This activity is probably a mixture of 5.2-hr. and 2.8-hr. periods reported by Coleman and Pool[9] and assigned by them to Sb^{118} and Sb^{117}, respectively. The present work demands that the mass numbers be in this region, but Sb^{118} has been assigned to a 3.5-min. activity (see 6.0-day Te^{118} above).

39-hr. Sb^{119}

This isotope, exhibiting only x-rays in its decay,[9] was also identified as the daughter of 4.5-day tellurium[4] as part of the present studies. Since this activity possesses no particles or gamma-rays, it could best be seen in decay curves taken through beryllium. Under these conditions the activity was prominent, and with deuteron energies in excess of 100 Mev, it was formed in much higher yield than its tellurium parent. When counted without absorber, the 2.8-day Sb^{122} negatrons completely overshadowed the less efficiently detected x-rays of Sb^{119} since both were formed in somewhat comparable yields.

5.7-day Sb^{120}

The electromagnetic radiation of the antimony fraction when followed either through beryllium ($\sim$2 g/cm^2) or through beryllium plus lead (130 mg/cm^2) showed a prominent 5.7-day period in all irradiations of antimony with deuterons in the energy range 50–190 Mev. It was assigned as an isomer of the 17-min. Sb^{120} positron-emitter on the basis of irradiations of separated Sn^{120} with deuterons of 19-Mev energy and from other considerations.[4] It was shown to undergo orbital electron-capture decay by critical absorption measurements of the x-rays and to have gamma-rays and conversion electrons.

2.8-day Sb^{122}

This negatron-emitter appeared as the most prominent activity a few days after irradiation when the

[7] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. **20**, 585 (1948).
[8] J. E. Edwards and M. L. Pool, Phys. Rev. **69**, 140 (1946).
[9] K. D. Coleman and M. L. Pool, Phys. Rev. **55**, 1070 (1947).

antimony fraction was counted without absorber. When only electromagnetic radiation was followed, the 2.8-day period of Sb^{122} was not in evidence because of the preponderance of 39-hr. Sb^{119} and 5.7-day Sb^{120} radiation. The identity of the Sb^{122} was confirmed by a beta-ray absorption curve.

60-day Sb^{124}

All antimony decay curves tailed out into this period which could be followed through several half-lives. The beta-ray absorption curve served further to identify the isotope.

Tin Fraction: 4.5-hr. $Sn^{108(?)}$

In the irradiation of antimony with highest energy particles (190-Mev deuterons and 380-Mev helium ions), a 4.5-hr. period was found in the tin fraction. This activity could not be detected following the irradiation of antimony with deuterons of 100 Mev or less. The 4.5-hr. tin decays by orbital electron capture and is the parent of a 70-min. indium positron-emitter of 2.2-Mev energy as determined by a beryllium absorption curve. It was not possible to make a mass assignment on the basis of this work. Recently Mallary and Pool[10] have produced this pair by bombardment of enriched Cd^{106} with 20-Mev alpha-particles and assigned the mass number 108 to it.

The high energy necessary to produce the 4.5-hr. Sn from antimony is consistent with its assignment to an isotope of low mass number, as is its mode of decay.

28-hr. Sn^{121}

This activity which appeared in all irradiations has been described previously.[4] The isotopic assignment was made by the irradiation of separated Sn^{120} with 19-Mev deuterons. Since there is no gamma-ray associated with this isotope, it could only be followed in decay curves taken without absorber.

14-day Sn^{117m}

This period was previously reported by Livingood and Seaborg[11] from the irradiation of cadmium with helium ions, setting the maximum mass number at 119. In the present study critical absorption measurements of the x-rays showed the decay process to be an isomeric transition; from yield considerations of deuterons of different energies on antimony to be discussed below, it seemed probable that the mass number lay in the range 118–122. Of the two possible assignments from these studies, 119 was chosen as the most probable. However, it has recently been shown[12,13] that the mass number more likely is 117.

The radiations from Sn^{117m} are K x-rays in high abundance, an electron of about 0.15 Mev as determined by absorption in beryllium and in a beta-ray spectrometer, and the corresponding gamma-ray of 0.175 Mev determined by absorption in lead.

The 14-day period for Sn^{117m} was resolved readily from the decay curves of the tin fraction from antimony irradiations when the electromagnetic radiation was followed through beryllium.

110-day Sn^{113}

This period could be followed after decay of the 14-day Sn^{117m} and could be determined earlier by removal of its 110-min. In^{113m} daughter.

Indium Fraction: Short-lived Isotopes

The indium fraction, like that of antimony, always contained a complex mixture of activities ranging in half-life from less than an hour to about five hours. Resolution and identification of the different components were unsatisfactory, so no yield data are presented. By following the positrons in the beta-ray spectrograph, the 20-min. period and the 65–70 min. period were found. However, it is fairly certain that 105-min. In^{113m}, 4.5-hr. In^{115m}, 54-min. In^{116}, and 117-min. In^{117} were also present, making quantitative determination of all isotopes difficult.

2.7-day In^{111}

In the irradiation of antimony with 100-Mev deuterons, this activity could barely be detected but became more prominent at higher energies.

48-day In^{114m}

This nuclide, along with its 72-sec. negatron-emitter daughter, was identified readily by following the decay curve through several half-lives and by confirming the radiation characteristics by absorption curves.

Cadmium Fraction: Short-lived Isotopes

At high energies at least one activity with a half-life in the range 1 to 2 hr. was detected in low yield. From the fact that 100-Mev deuterons failed to show this activity, it is deduced that the mass number is near or below the lightest stable cadmium isotope, Cd^{106}. These isotopes were not further characterized because of the low yield.

6.7-hr. Cd^{107}

Decaying by orbital electron capture, this isotope was definitely identified by separating its daughter, 40-sec. Ag^{107m}, as well as by its prominence in the decay curve taken through beryllium. The Cd^{107} was detected with difficulty by decay curves taken without absorber since it was masked by the beta-activity of the shorter- and longer-lived isotopes.

[10] E. C. Mallary and M. L. Pool, Phys. Rev. **76**, 1454 (1949).
[11] J. J. Livingood and G. T. Seaborg, Phys. Rev. **55**, 667 (1939).
[12] J. W. Mihelich and R. D. Hill, Phys. Rev. **77**, 743 (1950).
[13] E. C. Mallary and M. L. Pool, Phys. Rev. **77**, 75 (1950).

2.3-day and 42-day Isomers of Cd^{115}

The 2.3-day Cd^{115} was prominent in decay followed without absorber, but was almost undetectable in the decay curve taken through beryllium. Identification and yield determination were also aided by determining its 4.2-hr. In^{115m} daughter. Since the 42-day Cd^{115} has no prominent electromagnetic radiation, it could only be measured in the decay curve taken without absorber.

330-day Cd^{109}

This activity was apparent in the decay curve taken through beryllium soon after the 6.7-hr. Cd^{107} had disappeared. Ultimately it could be seen in the curve taken without absorber after the 42-day Cd^{115} had decayed. Identification was made certain by removal of its daughter, 40-sec. Ag^{109m}.

From the above account of the cadmium fraction it may be visualized that the decay curves taken with and without absorbers were quite different. Without absorber the short-lived activities and the Cd^{115} isomers were most prominent, while decay curves taken through beryllium showed largely the 6.7-hr. Cd^{107} and 330-day Cd^{109}.

Silver Fraction: 24-min. Ag^{106}

This positron-emitting isomer of Ag^{106} was detected with the beta-ray spectrometer as the shortest-lived positron component but it could not be analyzed satisfactorily in the decay curve followed on the Geiger counter and no yield data are presented. The 8.2-day isomer of Ag^{106} is discussed below.

70-min. Ag

This positron-emitter, probably that reported by Enns,[14] could be identified in the same manner as that described for Ag^{106}. It could not be resolved in the Geiger counter decay curve taken without absorber but was prominent in that determined through beryllium. This indicates considerable orbital electron capture branching in the decay of this isotope.

7.5-day Ag^{111} and 8.2-day Ag^{106}

It was possible to show that both of these isotopes are formed in antimony spallation and to determine the yield of each. The 7.5-day Ag^{111} is a negatron emitter with no associated gamma-rays, as a result of which it could be detected without absorber but would not be counted through a sufficient amount of beryllium. On the other hand, 8.2-day Ag^{106} decays by orbital electron capture and has gamma-rays which Feather and Dunworth[15] showed to be but slightly converted. As a result, nearly all of the 8-day activity followed without absorber was due to Ag^{111} while all of the 8-day activity followed through beryllium was due to Ag^{106}. Further proof that the two isotopes were present could be found in the ratio of the 8-day activity measured in these two ways when the projectile energy was changed.

45-day Ag^{105}

This activity could be detected only after irradiation of antimony with deuterons of 190 Mev or helium ions of 380 Mev. It was more prominent in decay curves taken through beryllium than without absorber but, even so, the resolution was difficult from 225-day Ag^{110} in the relatively short period of time the samples were followed.

225-day Ag^{110}

As mentioned above the accuracy in resolution from 45-day Ag^{105} was not as high as for most of the nuclides for which yields were calculated.

3.2-hr. Ag^{112}

This activity was observed in all irradiations of antimony with deuterons from 50 to 190 Mev. It showed up better at the lower energies because of the absence of the other short-lived isotopes of silver which only appeared at higher energies. The isotope was identified by absorption curves which showed the presence of a very hard beta-particle.

Palladium Fraction

The palladium fraction contained so many activities of similar half-life that they could not be resolved directly from decay curves. However, three activities whose half-lives lie in the range 10 to 20 hrs. have radioactive daughters which can serve as indices of the amounts of the parent nuclides present. For palladium and lower elements measurable yields were obtained only with highest energy deuterons and helium ions.

21-hr. Pd^{112}

This activity could be measured by its 3.2-hr. Ag^{112} daughter. Decrease in yield of the Ag^{112} removed from the palladium fraction at intervals defined a 21-hr. half-life and served to determine the yield of Pd^{112}.

26-min. Pd^{111}

When the palladium separation was performed sufficiently rapidly, the 26-min. negatron emitter was noted and after decay the 7.5-day Ag^{111} could be removed from the palladium fraction.

12-hr. Pd^{109}

This negatron emitter was positively identified by separation of its daughter, the 40-sec. Ag^{109m}. Its yield was determined by resolution of the decay curve taken without absorber correcting for the 21-hr. Pd^{112} present which was uniquely determined through its daughter, 3.2-hr. Ag^{112}.

[14] T. Enns, Phys. Rev. **56**, 872 (1939).
[15] N. Feather and I. V. Dunworth, Proc. Roy. Soc. **A168**, 578 (1938).

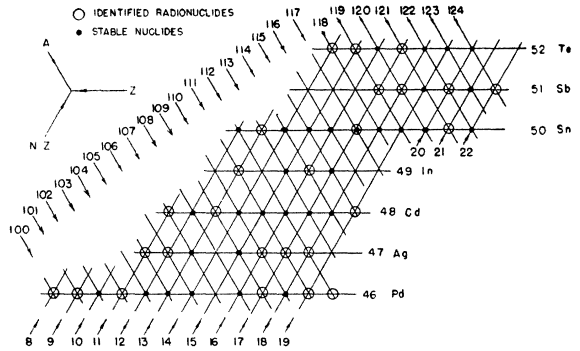

FIG. 1. Distribution of spallation products of antimony.

17-day Pd^{103}

This activity appeared as the longest-lived component of the palladium fraction and its yield was determined by measuring the x-rays.

10-hr. Pd^{101}

This isotope, which decays both by electron capture and positron-emission to its 4.3-day Rh^{101} daughter, was described in an earlier report.[5] Its yield was determined by measuring the x-rays of the Rh^{101} to which it decayed.

4.0-day Pd^{100}

Also described previously,[5] this isotope decays by orbital electron capture with the emission of gamma-rays to a 19.4-hr. daughter, Rh^{100}, which in turn decays predominantly by orbital electron capture with about 5 percent branching through positron-emission. The yield was determined best by measuring the x-rays of the Rh^{100}.

Ruthenium Fraction: 4.5-hr. Ru^{105}

This negatron emitter was the only short-lived isotope in the ruthenium fraction. The yield was checked by distilling the ruthenium from perchloric acid after decay of the Ru^{105} leaving its 36-hr. Rh^{105} daughter which could then be measured.

2.8-day Ru^{97}

This activity could be resolved out of the decay curves taken through beryllium and without absorber. It showed up well in the ruthenium distilled from the 36-hr. Rh^{105} after decay of the 4.5-hr. Ru^{105}.

42-day Ru^{103}

Detected in the decay curve taken without absorber, this isotope also appeared in the ruthenium distilled from the Rh^{105}.

1-yr. Ru^{106}

This period was found after decay of 42-day Ru^{103} and could be detected earlier through the hard beta-rays of its daughter, 30-sec. Rh^{106}. The yields determined by the two methods were in essential agreement.

Molybdenum Fraction: 67-hr. Mo^{99}

No short-lived molybdenum activities were looked for and this activity was the only one found in the molybdenum fraction. The beta-ray absorption curve was in agreement with those reported. This period was followed through about 6 half-lives following irradiation of antimony with 200-Mev deuterons.

Yttrium Fraction: 80-hr. Y^{87}

No yttrium activities of half-life less than one day were sought. Initial growth in the yttrium fraction suggested the presence of 80-hr. Y^{87} producing its daughter 2.7-hr. Sr^{87m}. This was confirmed by removal of the strontium. The decay curve of the yttrium was less than 80 hrs. which suggests that some 62-hr. Y^{90} was present but that the two yttrium activities were not resolved.

57-day Y^{91}

This period which was followed through three half-lives appeared after Y^{87} had decayed.

4. DISCUSSION OF RESULTS

In Fig. 1 is shown the distribution of identified spallation products of antimony with 190-Mev deuterons as far down as isotopes of palladium, in relation to the stable nuclides in this region. Table I gives the yields of these and other products listed in columns according to the energy and type of projectile which produced them.

In general, there is a continuous distribution of reaction products from the neighborhood of the target nuclides down to those lower by about 12 units of atomic number and 35 mass number units. For almost every element, radioactive isotopes are found on both sides of the region of beta-stability, that is, those with excess of neutrons and those that are neutron-deficient. Because of the complexity of chemical separations and the difficulty of resolving many of the radioactive mixtures, many short-lived activities which were undoubtedly present were either missed through decay or were not used in the yield calculations for lack of positive identification. As an example, there was evidence for the presence of six indium activities of a few hours half-life or less, but the yields of none could be calculated.

As pointed out in Section 1, the wide distribution of products is in keeping with the description by Serber[6] of nuclear encounters with high energy particles. According to this, if antimony is irradiated with 190-Mev deuterons, it is not to be expected that a compound nucleus will be formed in each nuclear encounter; instead, the present picture demands that all degrees of excitation up to the maximum be possible.

With respect to its reaction with a nucleus, a deuteron of 190 Mev is fairly accurately described as an independent proton and neutron each carrying approximately one-half of the kinetic energy. The principal significance of the existence of the proton and neutron as a deuteron is their spatial relation as they approach a nucleus. Another consequence of the high velocity of the neutron (or proton) is that its reaction with the struck nucleus may be described in terms of collisions with individual nucleons. This follows when the velocity of the approaching particle is greater than that of a particle within the struck nucleus, making it improbable that the kinetic energy can be distributed during the period of nucleon-nucleon interaction. For similar reasons, nuclear matter begins to be transparent at these energies, making the mean free path between collisions comparable with the nuclear dimensions.

From the above picture it is possible to visualize the multiplicity of nuclear reactions resulting from the irradiation of antimony with high energy deuterons. On one extreme a deuteron may approach a nucleus near an edge in such a way that the neutron of the deuteron, for example, penetrates and makes a single collision with a nucleon while the proton is stripped off in the process. According to Serber, the neutron would lose an *average* of about 25 Mev in the collision and, since it is near the edge of the nucleus, it would leave, carrying off most of its initial kinetic energy. The struck nucleon would, in general, give up its energy to excite the nucleus. Products from reactions of this type will be a few mass numbers lighter than the target nucleus and it is seen from Table I that representatives of this type, Sb^{119} and Sb^{120}, are formed in highest yield.

On the other extreme of energy transfer, one can visualize that both parts of the deuteron will strike the antimony nucleus near the center and in rare instances, each will undergo a sufficient number of collisions to give up its entire energy to the nucleus. This case is

TABLE I. Cross sections in millibarns for spallation products of antimony at various energies.

Nuclide		$T_{1/2}$	380-Mev alpha-particles	190-Mev deuterons*	100-Mev deuterons	75-Mev deuterons	50-Mev deuterons
52 Te	121	140 d *IT*	18	11	17	15	33
	119	4.5 d *K*	67	22	66	65	77
	118	6.0 d *K*	29	18	53	31	23
	?	2.6 hr. β^+	20	13	21	4.4	<0.07
51 Sb	124	60 d β^-	15	1.4	2.5	1.5	3.6
	122	2.8 d β^-	72	44	65	28	31
	119?	5.7 d *K*	180	110	160	40	47
	118?	3.9 hr. *K*	100	160	180	63	29
50 Sn	121	28 hr. β^-	2.2	——	0.81	0.32	0.14
	117*m*	14 d *IT*	28	17	17	5.6	2.2
	113	110 d *K*	30	17	6.2	0.7	<0.03
	108	4.5 hr. *K*	8	——	<0.01	——	——
49 In	114*m*	48 d *IT*	23	16	5.1	0.86	<0.002
	111	2.7 d *K*	160	72	0.95	<0.01	——
48 Cd	115	2.3 d β^-	0.35	0.15	0.026	0.0041	<0.0017
	115	42 d β^-	1.1	0.68	0.11	0.0088	0.0041
	109	330 d *K*	14	3.2	<0.14	——	——
	107	6.7 hr. *K*	43	5	<0.005	——	——
47 Ag	112	3.2 hr. β^-	0.57	0.28	0.038	0.0029	<0.00042
	111	7.5 d β^-	2.3	0.63	0.080	0.0035	<0.00017
	110	225 d *K*	——	4.8	0.18	——	——
	106	8.2 d *K*	3.5	11	<0.01	——	——
	105	45 d *K*	1.7	5	<0.02	——	——
46 Pd	112	21 hr. β^-	0.0043	0.0008	<0.0001		
	111	26 min. β^-	——	0.03			
	109	13 hr. β^-	0.12	0.027			
	103	17 d *K*	4.7	1.0			
	101	8 hr. β^+	1.1				
	100	4 d *K*	1.0	0.013			
44 Ru	106	1 yr. β^-		0.002			
	105	4 hr. β^-		0.008			
	103	40 d β^-		0.004			
	97	2.7 d *K*	0.32	0.01			
42 Mo	99	67 hr. β^-		0.0023			
39 Y	91	57 d β^-		0.0009			
	87	80 hr. *K*		0.0003			

* Most of the values in this column represent averages of three measurements.

TABLE II. Yields of silver isotopes from 190-Mev deuterons on antimony.

Mass	Cross section (millibarns)
Ag 112	0.28
111	0.63
110	4.8
106	11
105 (?)	1.7

identical with compound nucleus formation at lower energies in that the incident particles are captured, after which nuclear evaporation considerations explain the results. With 190-Mev deuterons it would be predicted that such collisions would cause the evaporation of 20 or more nucleons, depending upon whether they are lost as individual neutrons and protons or as more complex fragments such as alpha-particles. Representative products of such reactions are indeed found, as shown in Table I.

Formation of Antimony Isotopes

As mentioned above, the formation of the antimony isotopes measured corresponds to the transfer of comparatively small amounts of energy to the nucleus, hence the loss of only a few neutrons, or none at all, as evidenced by the formation of Sb^{124}. It is of interest to compare the yields of Sb^{122} and Sb^{124}. The latter can be formed only by the reaction: $Sb^{123}(d,p)Sb^{124}$ or its equivalent. The former, however, can be formed by two reactions: $Sb^{121}(d,p)Sb^{122}$, and $Sb^{123}(d,p2n)Sb^{122}$.

Since the yield of Sb^{124} is an order of magnitude less than that of Sb^{122}, it is apparent that Sb^{122} must be formed principally by the $(d,p2n)$ reaction. For 190-Mev deuterons, the $(d,p2n)$ reaction appears about thirty times as probable as the (d,p) reaction. This ratio appears to have decreased to about twenty for 75-Mev deuterons and ten for 50-Mev deuterons, and is in the direction which would be anticipated since the lower energy available would tend to make the (d,p) reaction more probable relative to the $(d,p2n)$ reaction.

Of further interest is the relatively large yield of Sb^{124} resulting from irradiation with 380-Mev alpha-particles as compared with that from irradiation with 190-Mev deuterons. Both can be looked upon as neutron addition reactions; however, the neutrons in the alpha-particle have a greater kinetic energy and momentum than the neutron in a deuteron. If at the instant of collision with the nucleon in the target nucleus, the momentum of the neutron is in the reverse direction from that of the center of mass, the effect will be to decrease the energy to be transferred to a nucleon struck by this neutron. In this way it is possible to capture a neutron from an alpha-particle with less energy to be radiated than if the neutron is a component of a deuteron.

TABLE III. Energy requirement for particle removal.

Particle	Binding energy	Coulomb barrier	Total necessary for expulsion
n	8	0	8
p	8	8	16
α	3	16	19

Pattern for Yields of Nuclides Formed with High Energy Particles

Reference to Table I shows that at the highest bombardment energies, yields of neutron-deficient isotopes of any product element generally are larger than yields of isotopes containing an excess of neutrons. Table II, for example, illustrates this situation for silver.

It will be seen that the yield generally increases as the mass number of the isotope decreases. The isotope Ag^{105} appears not to conform to this pattern; however, this may be due to difficulties in estimating counting efficiencies of the radiations associated with this nuclide and with resolution of the Ag^{105} from the Ag^{110}. Furthermore, the isotopic assignment of this period is still in doubt, and if its mass number is actually lower than 105, the low yield could be due to approach to the energy threshold.

The pattern illustrated above is rather general for all nuclides at these energies. Hence in Fig. 1, those nuclides lying far to the left (neutron-deficient) predominate over those lying on the right.

When the bombardment energy is decreased, it will be noticed that the yields of the neutron-deficient isotopes (except for those of Sb and Te) drop off much more rapidly than do the yields of those with neutron excess. This trend follows from energy considerations and will be discussed below.

These features of the spallation process may be explained qualitatively by considering energy requirements on the one hand and, on the other, the predilection for loss of neutrons rather than charged particles from highly excited nuclei. In discussing spallation

TABLE IV. Cross sections for isobars produced with 190-Mev deuterons.

Atomic number	Nuclide	Cross section (millibarns)
52	Te^{119}	22
51	Sb^{119}	160
47	Ag^{112}	0.28
46	Pd^{112}	0.0008
49	In^{111}	72
47	Ag^{111}	0.63
46	Pd^{111}	0.03
48	Cd^{109}	3.2
46	Pd^{109}	0.027
47	Ag^{106}	11
44	Ru^{106}	0.002
46	Pd^{103}	0.1
43	Ru^{103}	0.004

reactions with high energy particles, the inability to write a unique reaction to describe each of the products is an immediate obstacle. As an example, in the formation of Ag^{112} from the irradiation of antimony (let us say Sb^{121}) with 190-Mev deuterons, it is neither possible to define an intermediate excited nucleus nor to tell by what combination of neutron and proton evaporation Ag^{112} is reached. For purposes of discussion, let us assume that Sb^{121} itself is the primary excited nucleus, and that protons are lost as alpha-particles whenever possible. Then the Ag^{112} would arise from the loss of "$2\alpha n$" using the short-hand expression. It is seen from Table II that with 190-Mev deuterons a greater disparity between neutrons and protons is favored even though the loss of more nucleons is demanded. Thus, Ag^{106} from the loss of $2\alpha 7n$ is formed with 11 mb cross section, as compared with $Ag^{112}(2\alpha n)$ with 0.28 mb, and $Ag^{111}(2\alpha 2n)$ with 0.63 mb. From Table I it may be seen that cadmium isotopes show the same behavior.

All of these reactions require considerable nuclear excitation; in fact, 100 Mev is probably not sufficient to form Ag^{106}. From Table I it is seen that Ag^{106} and Ag^{105} are no longer measurable using 100-Mev deuterons; and although the cross sections for formation of Ag^{110}, Ag^{111}, and Ag^{112} have decreased from what they were at 190 Mev, these nuclides are now the predominant silver isotopes. Table III shows[16] roughly the energies required to expel nucleons and alpha-particles in this region. For the loss of $2\alpha 7n$ to form Ag^{106}, just about 100 Mev would be required, and it is therefore not surprising that the yield should fall off at least 1000-fold between 190 Mev and 100 Mev. On the other hand, $Ag^{110}(2\alpha 3n)$ would require 62 Mev excitation at threshold, and the yield only falls off by a factor of 25 between deuterons of 190 Mev and 100 Mev. It may further be noted that for reactions of this type, the protons are indeed most likely lost as alpha-particles since at 100-Mev excitation there is not sufficient energy to expel the protons and neutrons individually. Conceivably, in some cases less energy would be required in reaching lower atomic numbers from the target nucleus by the ejection of an even greater ratio of neutrons to protons followed by positron-emission, but in the present case Ag^{110} would be blocked by stable Cd^{110}. For that matter, Ag^{106} is shielded from positron-emission by Cd^{106} and therefore must be hit by the emission of the required amount of positive charge in the form of nucleons. In this same mass number region it may be noted that In^{114}, a completely shielded nuclide, is found in good yield.

The same pattern as that just discussed may be extended to a higher energy region. In the irradiation of antimony with 380-Mev alpha-particles, palladium isotopes could be reached in easily measurable yields; of these, the neutron-deficient isotopes Pd^{100}, Pd^{101}, and Pd^{103} were found more abundant than the heavier isotopes Pd^{109} and Pd^{112}.

[16] W. Horning, private communication.

Isobar Yields

There were a number of cases in which independent yields of isobars could be measured. Table IV summarizes the data for isobars formed with deuterons of 190 Mev.

There is, in every case except the first, a significantly larger yield of the isobar of higher atomic number. This may be taken as some measure of the ease with which a neutron is emitted from some excited nucleus in preference to the emission of charged particles.

The fact that the yield of Sb^{119} exceeds that of Te^{119} may be explained by the restrictions imposed upon the formation of the tellurium isobar. This can come about only by capture of a proton and loss of neutrons, whereas the antimony isobar may result from capture of either the proton or neutron of the projectile or neither.

Relative Yields of Isomers

There is no reason to believe that the relative amounts of two isomers produced in spallation reactions should change particularly with bombardment energy, since in all cases, after the last nucleon has left, the nucleus is left in an excited state well above the level of the highest lying isomer. Table I shows cross sections for the formation of Cd^{115} isomers using 380-Mev alpha-particles and deuterons from 190 Mev to 75 Mev. The ratio varies irregularly in this energy range and on the average, the upper state (43 day) is formed in three times the yield of the lower state.

In comparison with this ratio of Cd^{115} isomers formed in spallation, the ratios arising from slow neutron fission of uranium[17] and fission of thorium with 38-Mev alpha-particles[18] are both about 0.07. This is presumably the ratio at which these isomers arise from β^--decay of Ag^{115}, making it accordingly likely that Cd^{115} is not a primary fission product in these types of fission. On the other hand, in the fission of bismuth with 190-Mev deuterons[19] and uranium with 380-Mev alpha-particles,[20] the ratio of the upper to lower state is about 0.6, indicating that part arises from Ag^{115} decay but most as primary fission products. From slow neutron capture in cadmium, the ratio of 43-day to 2.5-day isomers was reported[21] to be approximately 0.1.

Yields of Nuclides of Low Atomic Number

The nuclide of lowest atomic weight and number which was definitely detected was $_{39}Y^{87}$. As may be inferred from the low cross section shown in Table I, the positive identification of nuclides in this region demanded extensive purification of the chemical frac-

[17] R. P. Metcalfe, reported in "Nuclei Formed in Fission," J. Am. Chem. Soc. **68**, 2411 (1946).
[18] A. S. Newton, Phys. Rev. **75**, 17 (1949).
[19] R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).
[20] P. R. O'Connor and G. T. Seaborg, Phys. Rev. **74**, 1189 (1948).
[21] Seren, Engelkemeir, Sturm, Friedlander, and Turkel, Phys. Rev. **71**, 409 (1947).

tions. Consequently, only isotopes with half-lives of the order of a day were detected for elements in this lower region. This region has not, therefore, been very thoroughly investigated for short-lived activities.

If such nuclides do indeed arise from the antimony rather than from some impurity, the energy threshold is very close to the deuteron energy (190 Mev) even if one assumes that the maximum number of alpha-particles are ejected. It may be that such a mechanism is not possible, and that these nuclides arise from the ejection of fragments heavier than alpha-particles perhaps approaching a reaction which might more properly be termed fission.

To shed some light on the possibility that these nuclides arise from impurities in the antimony, a spectrographic analysis was made in which no foreign lines were detected, and upper limits for the amounts of In, Cd, Pd, Rh, Ru, and Zr could be set at about 0.01 percent. Similar low limits can be set for molybdenum because of the extremely small amount of Mo^{99} found. In general, the limits of impurities are such that it is somewhat unlikely that the nuclides in question could come from this source.

ACKNOWLEDGMENTS

We wish to thank Dr. Duane Sewell and members of the 184-inch cyclotron staff for the bombardments which made this work possible.

PHYSICAL REVIEW VOLUME 78, NUMBER 5 JUNE 1, 1950

Total Cross Section of C, O, Mg, Si, and S for Fast Neutrons*

G. FREIER, M. FULK, E. E. LAMPI, AND J. H. WILLIAMS
University of Minnesota, Minneapolis, Minnesota

(Received February 20, 1950)

The total cross sections of light nuclei containing an integral number of alpha-particles was found by measuring the transmission of C, Mg, Si, S and SiO_2 samples for fast neutrons in the energy range 0.60 Mev to 1.90 Mev and in the energy range 2.5 Mev to 5.0 Mev. The cross sections have many resolved resonances, and the number of resonances per unit energy range increases rapidly with atomic number.

I. INTRODUCTION

THE experimental determinations of nuclear energy levels have as their prime objective the acquiring of sufficient information to enable one to construct detailed models of nuclei. A convenient way of finding the energy levels in a nucleus is to bombard it with neutrons of known energy and find at what energies the bombarded nucleus readily accepts the incident neutrons to form a compound nucleus in some excited energy state. If one measures the transmission of a beam of neutrons through some absorber as a function of the energy, the transmission will decrease markedly for certain incident neutron energies. The energy at which this occurs corresponds to a characteristic state of the compound nucleus, and by finding all these states one can construct an energy level diagram for the nucleus. In this experiment it was hoped that one might find some similarity in the energy levels of compound nuclei formed by bombarding light nuclei containing an integral number of alpha-particles with neutrons, and thus lend support to the hypothesis that the relatively stable alpha-particle remains an entity in the nucleus.

To make the measurements of transmission independent of the physical size and shape of the absorber, they are expressed in terms of cross section by the relation $T=e^{-nx\sigma}$ where T is the transmission, σ is the total cross section, and nx is the number of scattering centers per cm^2 presented to the incident neutron beam. Energy levels of the compound nucleus then appear as resonances in the curves for the total cross section as a function of energy. The method of measuring the total cross section as a function of energy was similar to that described previously.[1]

II. EXPERIMENTAL PROCEDURE

The $Li^7(p,n)Be^7$ reaction was used to obtain neutrons up to 1.90 Mev. Protons accelerated by the Minnesota Van de Graaff generator bombarded a thin layer of Li which had been evaporated onto a 0.010-inch thick tantalum backing. The Li target was of such thickness that incident protons of 1.88 Mev lost 30 kev energy in passing through the target. The energy of the incident protons was determined with an electrostatic analyzer, and all measured energy values are relative to 1.882 Mev, the threshold of the $Li^7(p,n)Be^7$ reaction.[2] The stability of the machine was such as to make the measured proton energy uncertain to ± 10 kev. The

* This research was done with the partial support of the ONR, Contract N5ori-147, T.O. III.

[1] Bailey, Bennett, Bergstrahl, Nuckolls, Richards, and Williams, Phys. Rev. **70**, 583 (1946); D. H. Frisch, Phys. Rev. **70**, 589 (1946); Nuckolls, Bailey, Bennett, Bergstrahl, Richards, and Williams, Phys. Rev. **70**, 805 (1946); L. W. Seagondollar and H. H. Barschall, Phys. Rev. **72**, 439 (1947).

[2] Herb, Snowden, and Sala, Phys. Rev. **75**, 246 (1949).