mental fact that the showers show a smaller latitude effect than the total radiation at sea level is that the latitude sensitive part of the radiation is much less effective in producing showers at sea level than the nonlatitude sensitive part. This behavior we have shown can be accounted for, at least in part, by assuming that the latitude sensitive component, which consists of incoming electrons, has degraded to such an extent by the time it has reached sea level as to be relatively ineffective in producing showers. The major part of the sea-level showers are therefore due to the nonlatitude sensitive radiation.

We wish to express our appreciation to the Dollar Steamship Company and to Captain Murphy of the President Van Buren, also to the K.P.M. Line and to Captain Blaauboer of the Niew Holland, and to the Canadian Australasian Line for their cooperation and assistance in the experimental work here reported. We also wish to make grateful acknowledgment to the Carnegie Corporation which provided funds that made this work possible.

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Radioactivity in Silver Induced by Fast Neutrons

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Radioactive silver, Ag^{112}, has been produced by fast neutron bombardment of Cd^{112} and In^{115}. A half-life period of 3.2 hours is observed with the upper limit of the electron beta-ray spectrum at 2.2 Mev. Gamma-rays are also emitted, the number per beta-ray is about four. The radioactive silver isomer, Ag^{106}, produced by fast neutron bombardment of Ag^{107}, has a positron beta-ray spectrum upper limit of 1.9 Mev and an electron beta-ray spectrum upper limit of 1.3 Mev. The half-life periods are respectively 24.5 min. and 8.2 days. The former emits no gamma-rays other than the annihilation radiation. The latter emits a complex gamma-ray spectrum. The number of gamma-rays per beta-ray is about 35. Nuclear K electron capture is offered to explain this anomalously high gamma to beta ratio. A total of twenty nuclear reactions all leading to the production of radioactive silver have been observed.

INTRODUCTION

THE work with fast neutron bombardment of silver has been continued and the evidence that Ag^{106} is isomeric,[1] yielding two radioactive periods, has been augmented with beta- and gamma-ray measurements. In addition a new period in silver has been found which can be obtained only from cadmium or indium.

There are now a total of six radioactive periods in silver. The finding of the four new periods was made possible by the availability of strong beams of high energy bombarding particles. The two well-known periods in silver, 2.3 min. and 22 sec., were found by Fermi[2] nearly four years ago. He used neutrons from a Ra-Be source and found that slow neutrons were particularly effective in producing the radioactivity.

In this paper it will be pointed out that very fast neutrons, with energies ranging up to 20 Mev, are very effective in producing certain other radioactive periods in silver. However, for the purpose of identification of the radioactive products a routine survey was made with alpha-particles, deuterons, gamma-rays, slow and fast neutrons as bombarding particles on rhodium, palladium, silver, cadmium and indium. In each case the silver precipitate from the chemical separation was measured for radioactivity. As a result, a total of twenty nuclear reactions leading to radioactive silver have been observed.

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[1] Pool, Cork and Thornton, Phys. Rev. **52**, 380 (1937).
[2] Fermi, Ricerca Scient. **5**, 330 (1934).

Apparatus

Protons, deuterons and alpha-particles of energies of about 3.2, 6.3 and 12 Mev, respectively, were produced by the cyclotron. The respective currents were about 4, 8 and 0.15 microamperes. The fast neutrons from the $Li+H^2$ reaction were obtained by deuteron bombardment of lithium metal fastened securely to a water-cooled copper plate. The 17 Mev gamma-rays were obtained by bombarding metallic lithium with protons. The intensities of the radioactive products were measured with a Wulf string electrometer equipped with an ionization chamber filled with difluorodichloromethane ("Freon") to a pressure of about forty pounds. The energy of the beta-rays and Compton recoil electrons were measured with a six-inch diameter Wilson cloud chamber filled with hydrogen and placed in a magnetic field.

The 3.2-Hour Period, Ag^{112}

Cadmium and indium in the metallic form were both bombarded with the high energy neutrons from the $Li+H^2$ reaction. The length of bombardment has been in some cases as long as four hours. At the termination of the bombardment the cadmium was dissolved in dilute nitric acid to which was added a small amount of palladium and silver. Hydrochloric acid was then added to bring down silver chloride. After coagulation, decantation and filtration the precipitate was dissolved in ammonium hydroxide and the silver chloride brought down again as before. The bombarded indium was treated in the same manner except a small quantity of cadmium and silver were added instead of palladium and silver.

The rate of decay of the active silver separation from the irradiated indium is shown in Fig. 1. From irradiated cadmium the silver separation showed, in addition to the 3.2-hour period, the presence of a much longer period of about 7.5 days. This period has been attributed to Ag^{111} by Kraus and Cork; they obtained the activity by bombarding palladium with deuterons.[3] Fast neutron bombardment of indium produces a very strong two month period in the indium chemical separation.[4] However, in the chemical

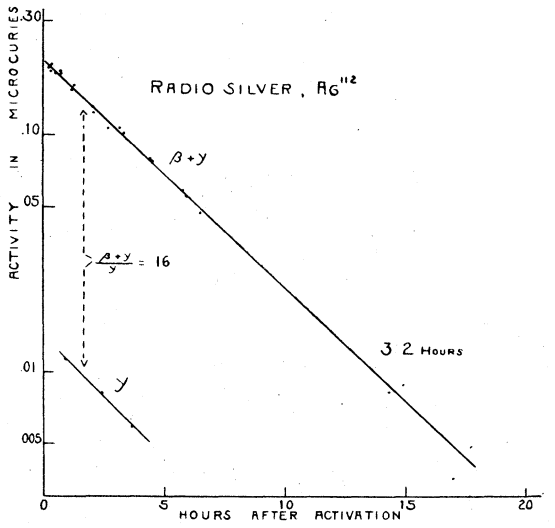

FIG. 1. Decay of Ag^{112} produced by fast neutron bombardment of indium. Period 3.2 ± 0.2 hours.

separation for silver, this strong long period was entirely absent and the 3.2-hour period appeared as in Fig. 1. The activity scale is arbitrary, but in order to give some idea of the order of magnitude of the intensity of the radioactivity, the scale was adjusted to read roughly in microcuries. The background was about 0.02 microcuries.

Other observers have detected the presence of a three to four hour period when indium was irradiated with neutrons. Amaldi *et al.* reported a weak long period of about three hours when indium was irradiated with Ra-Be neutrons.[5] A chemical check indicated the activity to be in indium. Szilard and Chalmers also using Ra-Be neutrons found a 3.5-hour period which was not water sensitive, but the intensity was too small to check chemically.[6] Bothe and Gentner were unable to obtain any period in the neighborhood of three hours by gamma-ray bombardment of indium.[7] Recently Lawson and Cork[8] have attributed a 4.1-hour period in indium to In^{114}.

Amaldi *et al.* found cadmium inactive when irradiated with Ra-Be neutrons. With deuteron bombardment of cadmium Cork and Thornton found a 4.3-hour period in the cadmium chemical separation but no activity in the silver separa-

[3] Kraus and Cork, Phys. Rev. **52**, 763 (1937).
[4] Pool, Cork and Thornton, Phys. Rev. **52**, 239 (1937).
[5] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti and Segrè, Proc. Roy. Soc. **149**, 522 (1935).
[6] Szilard and Chalmers, Nature **135**, 98 (1935).
[7] Bothe and Gentner, Zeits. f. Physik **106**, 236 (1937).
[8] Lawson and Cork, Phys. Rev. **52**, 531 (1937).

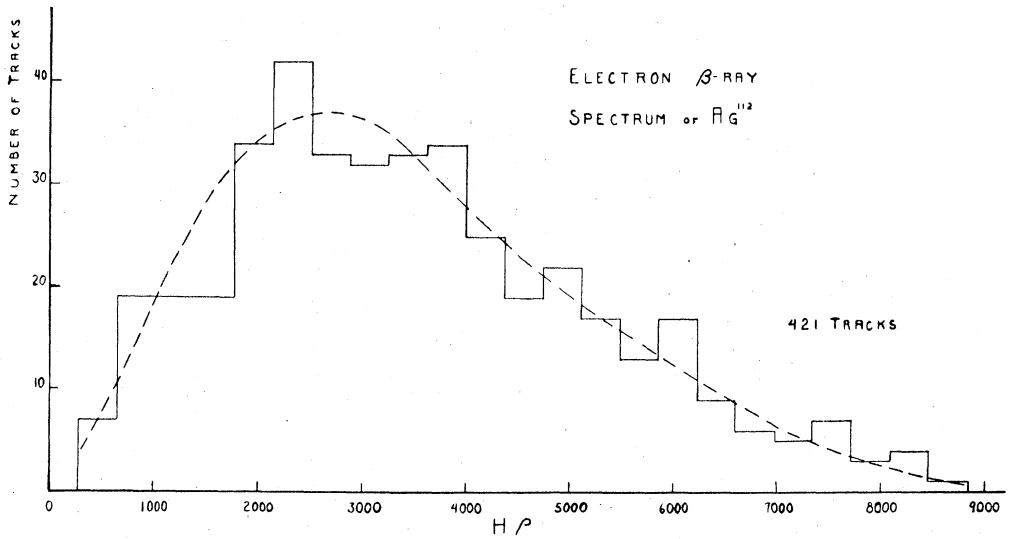

FIG. 2. Beta-ray spectrum of Ag^{112}. By inspection the upper limit is 8800 $H\rho$ (2.18 Mev).

tion.[9] Mitchell has verified the presence of a 4.3-hour period when cadmium is bombarded with slow neutrons; no chemistry was carried out. Heyn has observed a 3.3-hour period when cadmium is bombarded with high energy neutrons; again no chemistry was performed.[10]

Extended bombardment of palladium and silver by alpha-particles and by deuterons has failed to produce this 3.2-hour period. Since this period can be obtained only from indium and cadmium, it seems most probable that silver, Ag^{112}, is the carrier of the activity and the reaction equations are as follows:

$$_{49}In^{115}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{112}+{}_{2}\alpha^{4},$$
$$_{48}Cd^{112}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{112}+{}_{1}p^{1}.$$

Gamma-rays

In order to identify the presence of gamma-rays a piece of aluminum $\frac{1}{8}''$ or more in thickness was placed between the radioactive sample and the ionization chamber. The beta-rays were stopped while the gamma-rays passed through into the ionization chamber. The activity of the $(\beta+\gamma)$-rays divided by the activity of the gamma-rays alone was about 16. In order to interpret this value the ionization chamber was calibrated against radioactive nitrogen N^{13} as formed from C^{12} by deuteron bombardment. It is known that in this case two gamma-rays are emitted for each beta-ray emitted.[11] The calibration ratio was about 34 for similar geometrical conditions. The interpretation is, then, that Ag^{112} emits about 4 gamma-rays for each beta-ray.

Beta-rays

The radius of curvature of 421 beta-ray tracks were measured in a magnetic field of 372 oersteds, and the results are shown in Fig. 2. By inspection the upper limit of the beta-ray spectrum is 8800 $H\rho$ which corresponds to an energy of 2.2 Mev. This value of the energy together with the decay constant gives, on a Sargent plot, a point which falls on the second Sargent curve. The interpretation is that $_{47}Ag^{112}\rightarrow{}_{48}Cd^{112}+{}_{-1}\epsilon^{0}$ is a singly forbidden process.

THE 24.5-MIN. PERIOD, Ag^{106}

A 24-min. period was found by Bothe and Gentner when they bombarded silver by gamma-rays from the $Li+H^{1}$ reaction.[12] No chemical separations were made and the sign of the beta-particle was not reported. However, it appeared that the activity was probably due to Ag^{106}.

[9] Cork and Thornton, Phys. Rev. **51**, 608 (1937).
[10] Heyn, Nature **139**, 842 (1937).
[11] McMillan, Phys. Rev. **46**, 868 (1934).
[12] Bothe and Gentner, Naturwiss. **25**, 126 (1937).

Heyn shortly verified this period by bombarding silver with fast neutrons.[10] A 26-min. period in the silver separation from deuteron bombarded palladium was observed by Kraus and Cork.[13] The 24-min. period could also be obtained by bombarding silver with Ra-Be neutrons, and the intensity was great enough to show that chemically the activity was in silver.[14]

With the aid of the cyclotron much stronger radioactive sources could be prepared; the sign of the ejected particle was then found to be positive[15] which definitely placed the activity with Ag^{106}. For the purpose of verifying the identification Rh was bombarded with 12 Mev alpha-particles and cadmium was bombarded with fast neutrons. In each case the 24.5-min. period appeared faintly.

To get a strong sample of radiosilver, Ag^{106}, fast neutron bombardment of stable silver proves to be by far the best method. The next best method is by deuteron bombardment of palladium. 17 Mev gamma-ray bombardment gave a very feeble activity. Apparently, high energy proton bombardment of lithium does not enhance the emission of the 17 Mev gamma-rays from the $Li+H^1$ reaction as the high energy deuteron bombardment does the yield of neutrons from the $Li+H^2$ reaction.

[13] Kraus and Cork, Phys. Rev. **51**, 382 (1937).
[14] Reddeman and Strassmann, Naturwiss. **25**, 458 (1937).
[15] Pool, Cork and Thornton, Phys. Rev. **51**, 890 (1937).

Beta- and gamma-rays

Figure 3 shows the decay curve of a sample of silver bombarded for 22-min. with fast neutrons. The $(\beta+\gamma)/\gamma$ ratio is about 32. This value is sufficiently close to the calibration value 34, to indicate that no gamma-rays are emitted by this period in excess of the annihilation radiation due to the presence of the positrons. Fig. 4 shows the positron beta-ray histogram made from 814 tracks. By inspection the upper limit of the beta-ray spectrum is 7750 $H\rho$ which corresponds to 1.9 Mev. This value and the decay constant gives a point which falls on the first Sargent curve. The interpretation is that $_{47}Ag^{106} \rightarrow {}_{46}Pd^{106} + {}_{+1}\epsilon^0$ is an allowed transition.

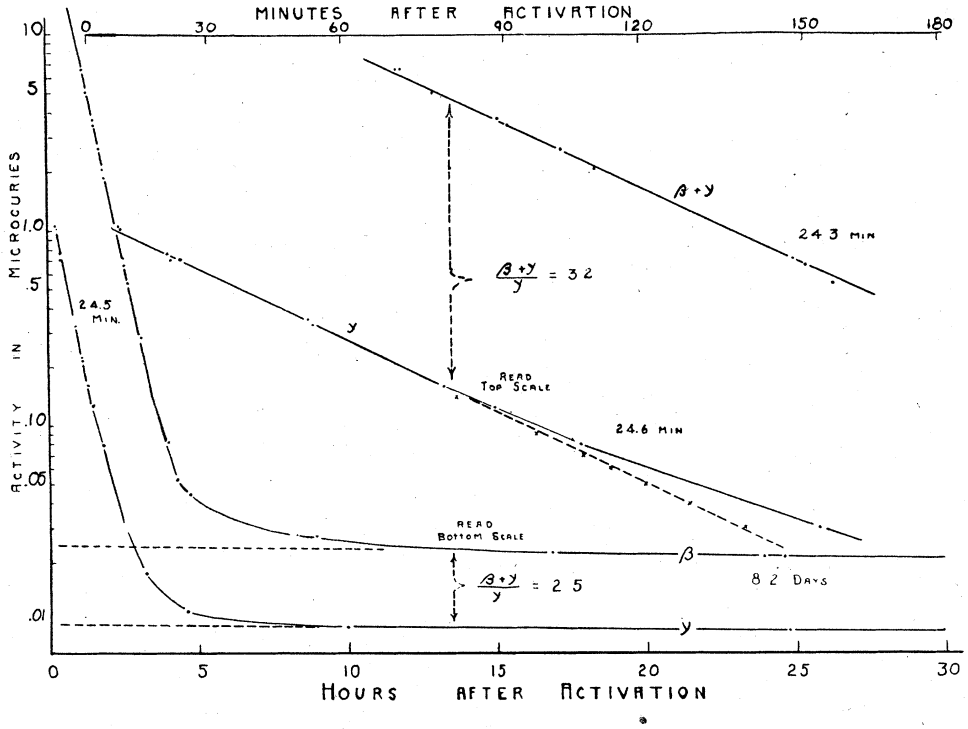

FIG. 3. Decay of Ag^{106} produced by fast neutron bombardment of silver. Period 24.5±0.5 min.

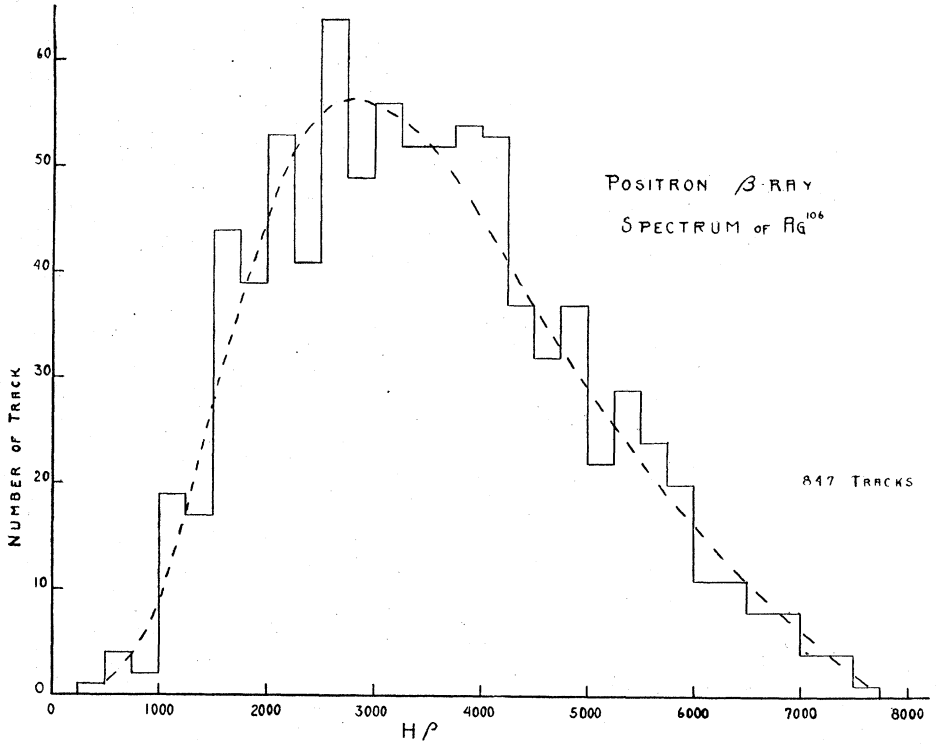

FIG. 4. Beta-ray spectrum of 24.5-min. Ag^{106}. By inspection the upper limit is 7750 $H\rho$ (1.86 Mev).

THE 8.2-DAY PERIOD, Ag^{106}

As may be seen in Fig. 3 there is, in addition to the 24.5-min. activity, an indication of a much longer period. This long period activity is produced with considerable intensity only by fast neutron bombardment of silver. All other methods, including alpha-particle bombardment of rhodium, deuteron bombardment of palladium and fast neutron bombardment of cadmium, produce this period in the silver separation very feebly. Fast neutron bombardment of indium, slow neutron bombardment of silver, and deuteron bombardment of silver definitely fails to produce the activity. As has been pointed[1] out, this activity which is in the silver chemical separation is also attributed to Ag^{106}.

Beta-rays

In Fig. 5 is shown the decay curve for two silver samples, one of which was bombarded six hours with fast neutrons. Fig. 6 shows the electron beta-ray histogram made from 447 tracks. By inspection the upper limit of the beta-ray spectrum is 5800 $H\rho$ which corresponds to an energy of 1.3 Mev.

Gamma-rays

As is seen in Fig. 5 the $(\beta+\gamma)/\gamma$ ratio is 1.37 for one silver sample and 1.62 for the other. This low ratio, which signifies a large number of gamma-rays per beta-ray, is quite remarkable because in a survey of the elements for radioactivity induced by fast neutron bombardment[4] no other induced activity had a ratio anywhere in the neighborhood of this value. Since the beta-rays are very strongly absorbed in silver metal, the $(\beta+\gamma)/\gamma$ ratio was observed for various thicknesses of silver and the value of the ratio for zero thickness of sample was obtained by extrapolation. This value is 2.9 and means that for each beta-particle emitted about 35 gamma-rays are emitted.

Figure 7 shows the gamma-ray spectrum as measured by means of Compton recoil electrons from a strip of mica. In accordance with the method outlined by Richardson and Kurie a

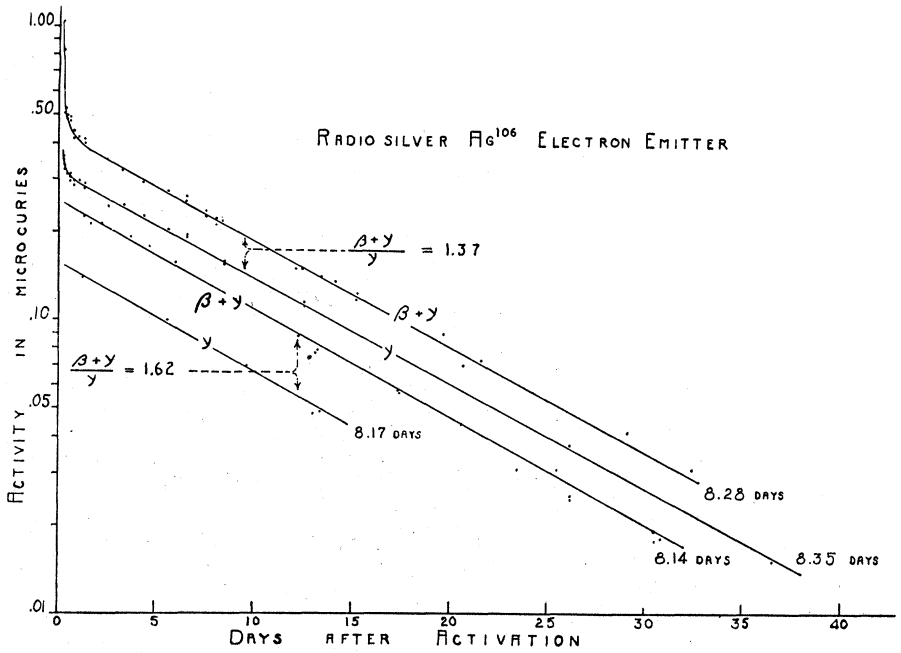

FIG. 5. Decay of Ag^{106} produced by fast neutron bombardment of silver. Period 8.2±0.3 days.

mica strip was placed across the cloud chamber.[16] Preliminary observations indicated that the gamma-rays were complex; consequently, a 20 mg/cm² strip was used instead of a thicker one because as good a resolution as possible was desired. In order to satisfy the requirements in the above reference in regard to the selection of recoil electron tracks only one track in about 150 pictures was good enough to measure. To satisfactorily measure the gamma-ray spectrum of this 8.2 day silver activity a very much stronger radioactive source is desired. Nevertheless, a spectrum line at about 0.95 Mev is evident. Spectra lines at about 0.68 and 0.28 Mev are also apparently present. The intensity of the 0.95 Mev line is about 1/15 the intensity of the two longer wave-length radiations.

K electron capture

If a nucleus Z emits a beta-ray and becomes a nucleus $Z+1$ in an excited state a gamma-ray may then be emitted leaving the nucleus $Z+1$ in the ground state. If there are two or more excited states above the ground state of the nucleus $Z+1$, a correspondingly larger number of gamma-rays per beta-ray could be emitted. However, in the case of Ag^{106} it is difficult to expect 35 such excited states in order to account for the observed gamma-rays.

Consequently, a totally different process may be responsible for the gamma-rays, such as nuclear K electron capture. According to the Fermi theory for the emission of beta-particles a nucleus can, instead of emitting a positron, absorb an electron from the K shell of the atom. The K radiation of the element formed might

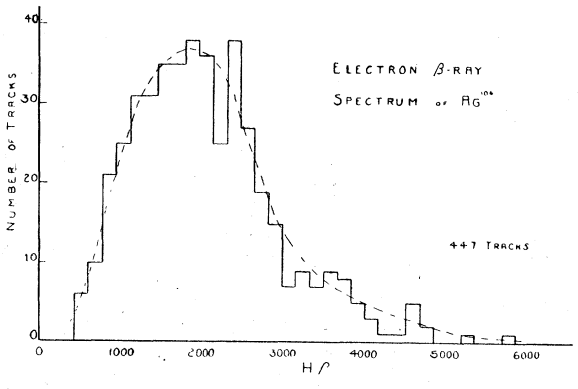

FIG. 6. Beta-ray spectrum of 8.2-day Ag^{106}. By inspection the upper limit is 5800 $H\rho$ (1.3 Mev).

[16] Richardson and Kurie, Phys. Rev. **50**, 999 (1936).

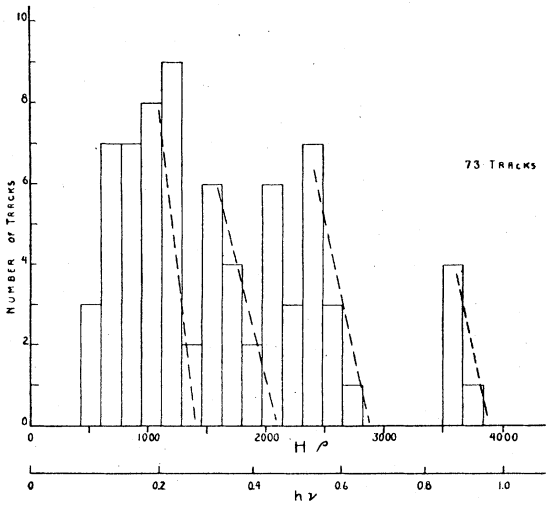

FIG. 7. Gamma-ray spectrum of 8.2-day Ag^{106}.

then be observed. Jacobsen,[17] searched without success for the K radiation from the positron emitting nucleus Sc^{43}. Alvarez,[18] however, has recently observed the K radiation from the positron emitting nucleus V^{48}.

The nuclear capture of the orbital K electron might be detected by another method. If the nucleus, which captures the K electron, is left in an excited state, gamma-rays could be emitted. It seems plausible that in the case of the 8.2-day period of Ag^{106} the very large γ/β ratio is due to this process. That is, Ag^{106} may either emit an electron and become Cd^{106} or absorb an orbital electron and become Pd^{106} in an excited state which immediately emits a gamma-ray. Preliminary calculations on silver in accordance with the method of Möller show that such a process is quite possible.[19]

Branching ratio

When the length of time of the bombardment, the half-lives of the radioactive substances formed and the initial intensities are taken into consideration, the branching ratio of the two members of the Ag^{106} isomer may be calculated. From three bombarded samples a mean determination indicated that about 20 times as many of 8.2-day Ag^{106} nuclei were formed as of the 24.5-min. Ag^{106} nuclei. *A priori* there is no reason to believe that each nucleus of an isomeric pair should be formed in equal numbers during a $n-2n$ reaction.

OTHER PERIODS

The 7.5-day period in silver, attributed to Ag^{111}, and mentioned above in connection with

[17] Jacobsen, Nature **139**, 879 (1937).
[18] Alvarez, Phys. Rev. **52**, 134 (1937).
[19] Möller, Physik. Zeits. Sowjetunion **11**, 9 (1937).

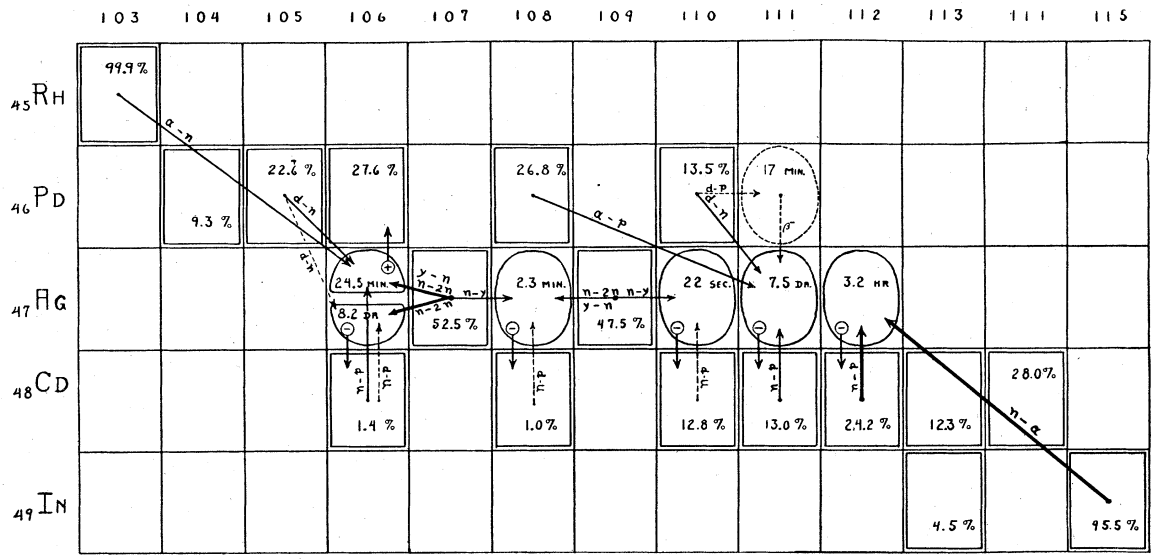

FIG. 8. Relative abundance of the stable isotopes and the positions of radioactive isotopes in silver. Arrows indicate method of production.

fast neutron bombardment of cadmium, is best formed by deuteron bombardment of palladium. Since a very small amount of the 8.2-day silver is also formed by the deuteron bombardment, it is necessary to bombard palladium with alpha-particles in order to get a pure 7.5-day activity. No gamma-rays are observed from this electron emitting substance. A four-hour bombardment with 0.15μA of 12 Mev alpha-particles on palladium placed inside the cyclotron tank gave a satisfactory activity.

The two short periods in silver obtained readily by slow neutron bombardment of silver, were also obtained by fast neutron bombardment of cadmium.

Summary of Reactions

Figure 8 shows a portion of the periodic table in the neighborhood of silver. The radioactive nuclei are shown and the arrows indicate how these nuclei may be formed. The reactions which were particularly studied in this report and which go strongly with fast neutron bombardment are indicated by heavy arrows. The dotted arrows represent reactions that were observed but not checked carefully. The twenty reactions which have been observed to produce radioactivity in silver are shown in Table I.

Table I

$_{45}Rh^{103}+{}_{2}\alpha^{4}\rightarrow{}_{47}Ag^{106}+{}_{0}n^{1}$ $_{46}Pd^{105}+{}_{1}d^{2}\rightarrow{}_{47}Ag^{106}+{}_{0}n^{1}$ $_{47}Ag^{107}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{106}+{}_{0}n^{1}+{}_{0}n^{1}$ $_{47}Ag^{107}+{}_{0}\gamma^{0}\rightarrow{}_{47}Ag^{106}+{}_{0}n^{1}$ $_{48}Cd^{106}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{106}+{}_{1}p^{1}$	24.5 min. period (positrons)
$_{46}Pd^{105}+{}_{1}d^{2}\rightarrow{}_{47}Ag^{106}+{}_{0}n^{1}$ $_{47}Ag^{107}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{106}+{}_{0}n^{1}+{}_{0}n^{1}$ $_{48}Cd^{106}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{106}+{}_{1}p^{1}$	8.2 day period (electrons, gammas)
$_{47}Ag^{107}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{108}+{}_{0}\gamma^{0}$ $_{47}Ag^{109}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{108}+{}_{0}n^{1}+{}_{0}n^{1}$ $_{47}Ag^{109}+{}_{0}\gamma^{0}\rightarrow{}_{47}Ag^{108}+{}_{0}n^{1}$ $_{47}Cd^{108}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{108}+{}_{1}p^{1}$	2.3 min. period (electrons)
$_{47}Ag^{109}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{110}+{}_{0}\gamma^{0}$ $_{48}Cd^{110}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{110}+{}_{1}p^{1}$	22 sec. period (electrons, gamma)
$_{46}Pd^{108}+{}_{2}\alpha^{4}\rightarrow{}_{47}Ag^{111}+{}_{1}p^{1}$ $_{46}Pd^{110}+{}_{1}d^{2}\rightarrow{}_{47}Ag^{111}+{}_{0}n^{1}$ $_{46}Pd^{111}\rightarrow{}_{47}Ag^{111}+{}_{-1}\epsilon^{0}$ $_{48}Cd^{111}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{111}+{}_{1}p^{1}$	7.5 day period (electrons)
$_{48}Cd^{112}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{112}+{}_{1}p^{1}$ $_{49}In^{115}+{}_{0}n^{1}\rightarrow{}_{47}Ag^{112}+{}_{2}\alpha^{4}$	3.2 hour period (electrons, gamma)

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