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Long Period Activity of Indium Produced by Slow Neutrons

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Indium was irradiated with slow neutrons from a radium-beryllium source for a period of several months. A period of 45 ± 3 days was produced quite strongly. It is shown that this period is produced when In^{113} captures a neutron to form In^{114}. The ratio of the activity of the long period to that of 54 minutes, due to In^{116}, is approximately the same as the isotope ratio In^{113}/In^{115}. The capture cross sections for the two processes are given by the relation $\sigma_{113}=0.56\sigma_{115}$.

SINCE this laboratory had available a neutron source, consisting of 200 milligrams of radium and beryllium, it was thought worth while to see if long period radioactivities could be produced by slow neutron bombardment. The strength of a radium-beryllium source does not decrease appreciably with time, as does the activity of a radon beryllium source. It therefore provides an ideal tool for long period irradiations. Accordingly, a number of substances were placed in a large box suitably filled with paraffin, in the center of which was placed the neutron source. The irradiation commenced July 1, 1937 and has continued up to the present, except for short but rather frequent intervals, in which the neutron source was removed for other purposes. Of the substances so far investigated, the most active is indium, which shows a long period of 45 ± 3 days in addition to the well-known period of 54 minutes. While this work was in progress Lawson and Cork[1] reported the discovery of an activity of 50-day period in indium which they ascribed to In^{114}. The relation between this work and the present paper will be discussed in more detail below.

[1] J. L. Lawson and J. M. Cork, Phys. Rev. **52**, 531 (1937).

IRRADIATION AND MEASUREMENT OF PERIODS

The box used for irradiating the samples was 26 cm high, 24 cm long, and 12 cm wide. The radium-beryllium source was placed in a small lead container in the center of the box. Two indium sheets 6×10 cm^2 and of a thickness of 0.109 g/cm^2 were placed about 5 cm from the neutron source, the rest of the box being filled with paraffin.

The first indium sample was removed after 80 days intermittent irradiation. Its activity was measured by wrapping it around a Geiger-Müller tube counter attached to an amplifier and a thyratron recording mechanism. The sensitivity of the counter system was measured before each measurement of the activity of the indium with the help of a standard uranium sample. The activity of the indium was then corrected for slight changes in counter sensitivity. Measurements on the activity of the sample did not commence until 5 minutes after it had been removed from the neutron source, so that the 13-sec. period was not measured. The scale of two recording instruments used to measure the first sample would not follow at counting rates above 1000 per minute so that the early part of the run could not be followed accurately. This defect was rectified when the second sample was measured, and the results will be discussed below.

The activity of the sample was followed for 62 days. It was found to decay with a period of 42 days. The initial activity was 142 counts/min. above a background of 35 counts/min. so that it was easily measurable. The long period activity subtracted from the total, gave the short period activity a period of 50 ± 5 min., with an extrapolated initial rate of about 10,000/min. In this calculation only those points were considered for which the counting rate was less than 1000/min. The short period could be followed for seven hours after the end of irradiation and there was no evidence indicating the existence of any intermediate periods.

A second indium sample, identical with the first, was removed after 155 days irradiation. The activity of the sample was measured in the same manner as before, except that the recording mechanism was a "scale-of-eight" counter fed by an amplifier of short time constant. The system was reliable for counting rates of 5000 per min., and probably missed only a small fraction of the counts at counting rates of 10,000 per min. Measurements were taken at half-hour intervals for a period of twenty hours. After the twelfth hour the activity was relatively constant at about 370 counts/min. above the background. This activity, measured from time to time over a long period was shown to decay with a period of 47 days. Subtracting the activity of the long period from the total activity only the 54-min. period was found.[2] The 54-min. period was observable for 10 hours. No indication of an intermediate period was found. The initial activity of the 54-min. period was 1.75×10^4 counts/min. while that of the long period was 3.72×10^2 counts/min.

Beta- and Gamma-Rays

In order to see whether the radioactive indium of long period emitted positive or negative electrons, an active sample, several weeks old, was placed in a cloud chamber. The curvature of the tracks in a magnetic field showed that the emitted particles were negative electrons. Lawson and Cork performed a similar experiment in which they were able to show that the upper limit of the electron energy is 2.15 Mev.

To test for gamma-rays a counter surrounded by lead of thickness 1.44 grams/cm^2 was used. Two days after the end of the irradiation the more active indium sample was examined with the gamma-ray counter. The counting rate (background) with no sample present was 23 ± 2 per minute, while that with the sample wrapped around the counter was also 23 ± 2. If there had been as many as 10 counts per min. due to gamma-rays these would have been detected. The count due to beta-rays from the sample at the time of measurement was 370 per min. Thus the gamma-ray effect is certainly less than 1/40 the beta-ray effect.

It is interesting to compare the above result with results of a similar experiment by Mitchell and Langer[3] on the relative number of counts due to beta- and gamma-rays associated with the 54-min. period of indium. Using the same apparatus as was used in the present experiment, these investigators found that the total count (beta-rays+gamma-rays) was 3700 per minute, while that for the gamma-rays alone was 700 per minute, both for the 54-min. period. Hence the ratio

$$\frac{I_0(\gamma)}{I_0(\gamma+\beta)}=\frac{700}{3700}=0.189. \quad (1)$$

Now the gamma-rays from the 54-min. period have an energy of 1.40 Mev.[4] If the 45-day activity had emitted γ-rays of like energy and in a corresponding amount, one should have obtained a count of 70 per min. with the gamma-ray counter. Since there was no observed count due to gamma-rays, one must conclude either that there are no gamma-rays associated with the long period or that their energy is quite small. The first assumption appears to be the more plausible.

The Origin of the 50-Day Activity

In order to show that the long indium period is not due to a fast neutron reaction with In^{115} the following experiments were tried. A silver

[2] Measurements were commenced too late to observe the 13-sec. period.

[3] A. C. G. Mitchell and L. M. Langer, to appear shortly.

[4] A. C. G. Mitchell and L. M. Langer, Phys. Rev. **52**, 137 (1937).

foil, the same size as the indium foil was given the same length of irradiation as the indium. Aside from a weak activity of very long period (probably about 3 months), the only other periods found were the well-known ones of 22 sec. and 2.3 min. There was no trace of the 26-min. period known to be produced in silver by fast neutrons.

A second experiment was performed in which an Ag detector was placed in the box in the same position as the indium foil and was irradiated for 1 hour. On measurement, this detector likewise showed only the 22-sec. and 2.3-min. periods. The latter period was followed until it disappeared into the background at the end of 35 min., there being no trace of the 26-min. period. Finally, the radium was removed from the paraffin and was allowed to irradiate for 1 hour an Ag foil, bent into the form of a cylinder around the source. In this case the 26-min. period was present and began to show its influence as early as the tenth minute of counting. Since there was no observable activity due to the 26-min. period of Ag, when this substance was irradiated in the box containing paraffin, we estimate that the contribution to the activity due to fast neutrons, under these conditions of irradiation, is less than 0.1 percent. The long period activity of the indium must therefore be due to slow neutrons.

In view of the above experiments it seems certain that the activity of 45-day period is due to In^{114} produced through the capture of a slow neutron by In^{113} according to the reaction.

$$In^{113}+n^1=In^{114},\quad In^{114}=Sn^{114}+e^-. \tag{2}$$

The two normal indium isotopes occur in the following percentages: In^{113}, 4.5 percent; In^{115}, 95.5 percent. Since the activational process has been shown to be due to the capture of slow neutrons, one can get some information as to the nature of the process by comparing the initial rates of decay for the 54-min. period and the 42-day period.

The 54-min. activity needs no correction for time of irradiation since it was radiated for a period very long compared to the half-life. In comparing the activities one should use only the beta-activity of the short period, since the long period gave no gamma-activity. The correction for gamma-rays is, however, negligible since a thin walled glass counter was used in which the gamma-rays would not produce many secondaries.

The initial activity of the long period radioactivity must be corrected for irradiation time. The time of irradiation is somewhat indefinite but lies between 2 and 3 half-lives. The observed initial activity of 3.72×10^2 counts per min. becomes 4.9×10^2 or 4.2×10^2 counts per min. when corrections are made for irradiation time of 2 or 3 half-lives, respectively. Now, the 54-min. period is certainly due to In^{116} produced from In^{115} by neutron capture. Assuming that the capture cross sections are the same for the two isotopes we can calculate the percentage abundance of the two isotopes. Taking the data given above we have for the abundance ratio

$$\frac{N_{113}}{N_{113}+N_{115}}=\frac{4.6\times10^2}{175+4.6}\times10^2 =\frac{I_0\,(45\text{ d.})}{I_0\,(45\text{ d.})+I_0\,(54\text{ min.})}=2.6\text{ percent.}$$

In view of the uncertainties involved, the figure is in satisfactory agreement with the value 4.5 percent for In^{113} obtained from mass spectra data. It, therefore, appears certain that the 45-day activity is due to In^{114}. Conversely if one assumes that activities are in the ratio of the relative abundance N_{113}/N_{115} one can calculate the relative cross section for the activation of In^{113} to In^{115} by neutrons. By use of the above figures one obtains $\sigma_{113}=0.56\sigma_{115}$.

Lawson and Cork[1] have found a number of periods due to indium when this element was bombarded by deuterons, neutrons (fast or slow), and when cadmium was bombarded by deuterons. The indium periods produced by deuterons on cadmium are: 2.3 hours (In^{117}); 20 minutes (In^{111}); and 4.1 hours ascribed to In^{114}. With fast neutrons on indium they found the periods 72 sec. (In^{112}), 4.1 hours (In^{114}), 50 days In^{114} and the well-known 13-sec. and 54-min. periods ascribed to In^{116}. With slow neutrons they obtained the 13-sec. and 54-min. periods and the 4.1-hour period, which apparently appeared only weakly.

The present work substantiates the conclusion of Lawson and Cork that the 45–50-day period is to be attributed to In^{114}, since it has been shown here to go with slow neutrons on indium. Furthermore, the relative activity of the 50-day period compared with the 54-min. period, produced by slow neutrons, is about what one would expect from the relative abundances of In^{113} and In^{115}.

From the evidence presented by Lawson and Cork it seems very probable that the 4.1-hour period is due to In^{114} since it was obtained with deuterons on Cd and quite strongly with fast neutrons on In. It is isomeric with the 50-day period. In the present investigation no 4.1-hour period was observed when slow neutrons bombarded indium. Lawson and Cork found this period to be produced only weakly with slow neutrons. It is not impossible that the weak activity observed by them was due to residual fast neutrons not slowed down by the paraffin. From the above, it appears that the 4.1-hour period is due to In^{114}, that it is produced by fast neutrons on In^{115}, but that it is not readily produced by slow neutrons on In^{113}. Since the two periods 4.1 hours and 50 days are isomeric, one must conclude, from the nonappearance of the 4.1-hour period with slow neutrons that the formation of this state is forbidden due to energy considerations or to the action of a selection rule.

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Note on *K* Electron Capture and Isomerism in Radiosilver

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From radioactivity data a tentative energy-level diagram is constructed for the isomeric nucleus Ag^{106}. These data indicate that, for the 8.2-day period, the relative probabilities of K electron capture: negative β-emission: positive β-emission $=640:40:1$. The 24.5-min. Ag^{106} level is metastable and 0.3 Mev above the 8.2-day Ag^{106} ground state. Theory requires that the difference in angular momentum associated with these two levels be at least 5 nuclear units.

Introduction

The capture of orbital electrons as an alternative to positron emission by radioactive nuclei has been considered theoretically by several investigators.[1] For heavy radioactive nuclei the probability for K electron capture has been calculated to be comparable to, or even a thousand times greater than that of the corresponding positron emission. It is also probable that some radioactive nuclei which are stable with respect to positron emission may be unstable with respect to K electron capture.[2]

The x-rays following K electron capture have been found by Alvarez[3] in the comparatively light element V^{48}, which emits positrons and has a half-life of 16 days. The possibility of K electron capture in potassium has been discussed by Weizsäcker[4] and Bramley.[5] Various observations on radiosilver Ag^{106} have been made which suggest that K electron capture plays an important role in the radioactive transformation of this nucleus.[6]

Recapitulation of Data

The 8.2-day period in silver was determined by the γ as well as by the $\beta+\gamma$ activity; 447

[1] Möller, Physik. Zeits. Sowjetunion **11**, 9 (1937); Uhlenbeck and Kuiper, Physica **4**, 601 (1937); Yukawa and Sakata, Phys. Rev. **51**, 677 (1937); Hoyle, Nature **140**, 235 (1937).
[2] Sizoo, Physica **4**, 467 (1937).
[3] Alvarez, Phys. Rev. **52**, 134 (1937).
[4] Weizsäcker, Physik. Zeits. **38**, 623 (1937).
[5] Bramley, Science **86**, 424 (1937).
[6] Pool, Phys. Rev. **53**, 116 (1938).