ment of the instrument and the cross-section measurement program. They are also indebted to G. H. Goertzel and H. C. Schweinler for valuable discussions on various problems which have been encountered.

This document is based on work performed

PHYSICAL REVIEW

VOLUME 74, NUMBER 8

OCTOBER 15, 1948

# **Radioactive Br Isotopes**

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A radioactive isotope of 2.4-day half-life has been produced in bromine by deuteron bombardment of electromagnetically enriched Se<sup>76</sup>, and by alpha-particle bombardment of electromagnetically enriched Se<sup>74</sup>. Assignment of the isotope is made to Br<sup>77</sup>. A positron end point of 0.4 Mev is determined. In addition to annihilation radiation, gamma-rays and K-capture are observed. The ratio of K-capture to positron emission is determined to be 20. The cross-section ratio of the (d, n) to the (d, 2n) reaction producing the isotope is 0.3. The ratio of the sum of the cross sections for formation of Br<sup>77</sup> by Se<sup>78</sup>(d, n) and Se<sup>77</sup>(d, 2n)to the cross section for Br<sup>82</sup> by Se<sup>82</sup>(d, 2n) is 0.4. A radioactive isotope of 1.7-hour half-life has been produced in

**C**YCLOTRON bombardments have been made with alpha-particles, deuterons, and protons on electromagnetically enriched selenium.\*\*\* Samples in which the stable isotope Se<sup>74</sup> was enriched from 0.9 percent to 14.1 percent and samples in which the stable isotope Se<sup>76</sup> was enriched from 9.5 percent to 41.5 percent were used. For comparison purposes, bombardments were also made with Hilger selenium.

As a result of these bombardments, two previously unreported radioactive isotopes in bromine have been found. The location and characteristic radiations of these isotopes, and also results of investigations on the 4.4-hour Br<sup>80</sup> isotope, will be presented in this paper. bromine by deuteron bombardment and by proton bombardment of enriched Se<sup>74</sup>. Assignment of the isotope is made to Br<sup>75</sup>. A positron end point of 1.6 Mev is determined. K-capture is observed in the activity. No gammaray activity other than that due to annihilation is found. The ratio of K-capture to positron emission from the Se<sup>74</sup>(d, n) reaction is determined to be 4.4. The ratio of the cross section for formation of Br<sup>75</sup> by Se<sup>74</sup>(d, n) to that of Br<sup>82</sup> by Se<sup>82</sup>(d, 2n) is 2.1. The 4.4-hour Br<sup>80</sup> isotope has been produced by a Se( $\alpha$ , p) reaction and found to emit positrons with an end point of 0.8 Mev. In producing Br<sup>77</sup> by proton bombardment of Se the  $(p, \gamma)$  reaction is observed to be two-thirds as probable as the (p, n) reaction.

under Contract Number W-35-058, Eng. 71 for

the Atomic Energy Project, and the information

covered therein will appear in the National

Nuclear Energy Series (Manhattan Project Technical Section) as part of the contribution of

the Oak Ridge National Laboratory.

#### THE 2.4-DAY Br<sup>77</sup> ISOTOPE

Samples of enriched stable Se<sup>74</sup> and Se<sup>76</sup> isotopes were prepared for alpha-particle bombardment by pressing equal amounts by weight of the finely ground selenium into the bottom of aluminum target holders under approximately 5000 pounds pressure. The two targets were bombarded simultaneously in the cyclotron by means of a rotating probe.

Figure 1 shows the decay of total activity obtained in each of the samples from this bombardment. A new activity of 2.4-day half-life appeared in the Se<sup>74</sup> sample but not in the Se<sup>76</sup> sample, in which only 0.5 percent of stable Se<sup>74</sup> was present. It was concluded that this 2.4-day period was formed from Se<sup>74</sup> and must belong either to Kr<sup>77</sup> or Br<sup>77</sup>.

To locate the activity, samples of enriched Se<sup>74</sup> and Se<sup>76</sup> were bombarded simultaneously with deuterons. The samples were prepared for bombardment in a manner similar to that in the

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<sup>\*\*\*</sup> Supplied by the Y-12 plant, Carbin Carbon Chemicals Corporation, through the Isotopes Division, U. S. Atomic Energy Commission, Oak Ridge, Tennessee.

experiment above. Figure 2 shows the decay curves of Br activity from the two samples. Measurements were taken on a Wulf electrometer attached to a Freon-filled ionization chamber. To obtain the  $\gamma$ -activity curves, a  $\frac{1}{4}$ -inch aluminum absorber was inserted between the sample and the ionization chamber. This amount of absorber was sufficient to stop all  $\beta$ -radiation and x-ray radiation present. In both samples, the established 34-hour Br<sup>82</sup> activity and the new 2.4-day activity appeared. The 34-hour Br<sup>82</sup> activity appeared in nearly equal intensities in both samples since the parent Se<sup>82</sup> isotope was present in approximately equal percentages in both cases and since equal amounts by weight of the two samples were bombarded. The 2.4-day activity appeared in the Se<sup>76</sup> sample approximately 28 times as strong as in the Se<sup>74</sup> sample. Since stable Se<sup>76</sup> was 28 times as abundant in the Se<sup>76</sup> sample as in the Se<sup>74</sup> sample, it was concluded that the 2.4-day activity was that of a Br isotope formed by deuteron bombardment of Se<sup>76</sup>. Since bombardment of Se with alpha-particles had previously located the activity as either Kr<sup>77</sup> or Br<sup>77</sup>, assignment of the activity was made to Br77.

To determine the character of beta-activity in the 2.4-day period, a sample of Br activity from deuteron bombardment of enriched Se<sup>76</sup> was placed in an electromagnetic field, with a Geiger counter tube located so as to intercept either  $\beta^{-}$ or  $\beta^{+}$ -activity separately, according to the direction of the magnetic field. Figure 3 shows the decay curves obtained from these measurements. In addition to the gamma- and  $\beta^{-}$ -decay curves of the 34-hour Br<sup>82</sup> period, a 2.4-day positron activity appeared. From the decay curve of this activity it was determined that the Br<sup>77</sup> isotope emits positrons.

Figure 4 shows the results of aluminum absorption measurements of Br activity from deuteron bombardment of enriched Se<sup>76</sup>. Measurements of activity were taken with various thicknesses of aluminum absorber placed between the sample and an ionization chamber. The curve of  $\beta$ activity was obtained by a subtraction of the curve representing electromagnetic activity from the curve representing total activity. A beta-end point of 0.097 g/cm<sup>2</sup>, corresponding to 0.36 Mev by the Sargent range-energy relation, and a betaend point of 0.172 g/cm<sup>2</sup>, corresponding to 0.50 Mev, were determined. The former was attributed to the 2.4-day Br<sup>77</sup> activity, and the latter to the 34-hour Br<sup>82</sup> activity. The end point of the 2.4-day Br<sup>77</sup> period was confirmed by betaray spectrometer measurements.

Figure 5 shows the decay of x-ray activity in the 2.4-day Br<sup>77</sup> period. The activity was obtained from simultaneous  $Se^{76}+d$  and  $Se^{74}+d$ bombardments of equal amounts of enriched selenium. Ionization measurements of  $(x+\gamma)$ activity were taken with a magnetic field so situated as to prevent all beta-radiation from entering the ionization chamber. The curves of  $\gamma$ -activity were obtained by inserting a  $\frac{1}{4}$ -inch aluminum absorber between the sample and the ionization chamber. The difference between the curve of  $(x+\gamma)$ -activity and the curve of  $\gamma$ activity then represented the activity from the sample due to x-ray radiation. Curves representing this x-ray activity from both bombardments are shown in Fig. 5. In both samples a 2.4-day half-life was determined as the period of the x-ray activity. In the Se<sup>76</sup> sample this activity appeared with approximately 4.4 times the intensity of the x-ray activity in the Se<sup>74</sup> sample. Since stable Se<sup>76</sup> was 4.4 times as abundant in the Se<sup>76</sup> sample as in the Se<sup>74</sup> sample, it was concluded



FIG. 1. A comparison of total activity decay from alphaparticle bombardment of enriched Se<sup>74</sup> and Se<sup>76</sup>. The new 2.4-dav Br<sup>77</sup> period is shown in the Se<sup>74</sup> sample.



FIG. 2. A comparison of Br activity decay curves from deuteron bombardment of enriched Se<sup>74</sup> and Se<sup>76</sup>. The 2.4-day Br<sup>77</sup> period is found more intense in the Se<sup>76</sup> sample.

ACTIVITY IN ARBITRARY UNITS

that the Br<sup>77</sup> isotope emits x-ray radiation. From energy considerations it was determined that this radiation was not due to the "bremsstrahlung" effect. It was thus determined that the K-capture process occurs in the Br<sup>77</sup> isotope.



The ratio of K-capture processes to positron emissions in Br<sup>77</sup> is computed to be 20. This ratio was obtained by a determination of saturation intensities and by corrections made for the relative ionization produced by beta radiation and x-ray radiation in the energy ranges involved. The ratio of reaction cross sections for the

0.36 MEV B

ALUMINUM ABSORPTION OF BR ACTIVITY

FROM A SE<sup>76</sup>+ d BOMBARDMENT

ELECTROMAGNETIC ACTI

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FIG. 4. Aluminum absorption measurements showing the 0.36-Mev positron end point of the 2.4-day  $Br^{77}$  period. The 0.50-Mev end point of the 34-hour  $Br^{s2}$  period is also

.156 .208 G/CM<sup>2</sup> OF ALUMINUM

0.50 MEV B

shown.



FIG. 5. A comparison of x-ray decay in Br activity from simultaneous deuteron bombardment of enriched Se<sup>74</sup> and Se<sup>76</sup>. X-ray activity from the 2.4-day Br<sup>77</sup> period is seen to be stronger in the Se<sup>76</sup> sample.

Se<sup>76</sup>(d, n) and Se<sup>77</sup>(d, 2n) processes forming the Br<sup>77</sup> isotope is approximately 0.3. The rate at which the 2.4-day Br<sup>77</sup> period is produced compared to that of the 34-hour Br<sup>82</sup> period by deuteron bombardment of selenium is approximately 0.4.

Gamma-ray activity in addition to that due to annihilation is observed in the Br<sup>77</sup> period.

# THE 1.7-HOUR Br<sup>75</sup> ISOTOPE

A 1.7-hour half-life has been observed in Br

activity from simultaneous deuteron bombardment of equal amounts of enriched Se<sup>74</sup> and Se<sup>76</sup>. Figure 6 shows a comparison of this 1.7-hour activity in the Se<sup>74</sup> and Se<sup>76</sup> samples. A Br<sup>80</sup>  $\beta^{-}$ -activity of 4.4-hour half-life appeared in both samples. The activity was somewhat stronger in the Se<sup>74</sup> sample than in the Se<sup>76</sup> sample due to the fact that the Se<sup>74</sup> sample contained 40.4 percent of the parent Se<sup>80</sup>, while the Se<sup>76</sup> sample contained 30.2 percent of Se<sup>80</sup>. Gamma-ray activity, due to 34-hour Br<sup>82</sup>, was also found in both samples. In addition to these periods, a  $\beta^+$ activity of 1.7-hour half-life appeared in both samples. The intensity of this activity in the Se<sup>74</sup> sample was greater than that in the Se<sup>76</sup> sample by a factor equal to the enrichment of stable Se<sup>74</sup> in the two samples. It was then concluded that the 1.7-hour positron activity was produced by deuteron bombardment of stable Se<sup>74</sup>.

Simultaneous proton bombardments of equal amounts of enriched Se<sup>74</sup> and Se<sup>76</sup> were also made. Figure 7 shows the decay of activity from the Se<sup>74</sup>+p bombardment, and Fig. 8 the decay of activity from the Se<sup>76</sup>+p bombardment. Measurements were made with the samples in an electromagnetic field to separate  $\beta^{-}$ - and  $\beta^{+}$ activity. Activities of the 4.4-hour Br<sup>80</sup> and 34-hour Br<sup>82</sup> periods appeared in both samples in intensities proportional to the abundance of stable parent isotopes present. The increased



FIG. 6. A comparison of  $\beta^{-}$  and  $\beta^{+}$ -activity in Br from simultaneous deuteron bombardment of enriched Se<sup>74</sup> and Se<sup>76</sup>.  $\beta^{+}$ -activity in the 1.7-hour Br<sup>75</sup> period is seen to be stronger in the Se<sup>74</sup> sample than in the Se<sup>76</sup> sample.



FIG. 7. Decay of activity from proton bombardment of enriched Se<sup>74</sup>. Strong  $\beta^+$ -activity in the 1.7-hour Br<sup>76</sup> period is shown.  $\beta^+$ -activity in the 4.4-hour Br<sup>80</sup> period is also shown. Currents used in producing the electromagnetic field for separating  $\beta^-$ - and  $\beta^+$ -activity are shown for the various decay curves.

intensity of the 1.7-hour half-life in the  $Se^{74}$  sample over that in the  $Se^{76}$  sample indicated that the activity was produced by proton bombardment of stable  $Se^{74}$ .

As a guide to the likelihood of a  $(p, \gamma)$  reaction taking place to produce the 1.7-hour Br activity, the relative probability of the (p, n) to the  $(p, \gamma)$ reactions in forming 2.4-day Br<sup>77</sup> was estimated. The observed intensities of Br<sup>77</sup> activity, from simultaneous proton bombardments of enriched Se<sup>74</sup> and Se<sup>76</sup>, were compared with known abundances of Se<sup>76</sup> and Se<sup>77</sup> present in the two samples. From these comparisons it was determined that in producing Br<sup>77</sup> by proton bombardment of selenium the ratio of the (p, n) to  $(p, \gamma)$  reactions is 1.5. It is therefore quite possible that the 1.7-hour Br activity is formed by the Se<sup>74</sup> $(p, \gamma)$ reaction.

In the decay curve of activity from  $Se^{74}+p$  bombardment, a  $125\pm5$ -day half-life appeared



FIG. 8. Decay of activity from proton bombardment of enriched Se<sup>78</sup>. Weak  $\beta^+$ -activity in the 1.7 hour Br<sup>76</sup> period is shown.  $\beta^+$ -activity in the 4.4-hour Br<sup>80</sup> period is also shown. Currents used in producing the electromagnetic field for separating  $\beta^-$ - and  $\beta^+$ -activity are shown for the various decay curves.

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FIG. 9. Aluminum absorption measurements showing the 1.6-Mev positron end point of the 1.7-hour Br<sup>75</sup> period.

which was presumed to be that of Se<sup>75</sup>. The ratio of reaction cross sections for production of the 1.7-hour Br activity compared to the 127-day Se<sup>75</sup> activity by proton bombardment was determined to be approximately one. This indicated that the 1.7-hour Br activity decays into the 127-day Se<sup>75</sup> period. From this data, assignment of the 1.7-hour activity was made to Br<sup>75</sup> as a Se<sup>74</sup>(p,  $\gamma$ ) reaction.

Figure 9 shows the results of aluminum absorption measurements of Br<sup>75</sup> activity from deuteron bombardment of enriched Se<sup>74</sup>. The end-point energy of Br<sup>75</sup> positron emission was determined to be 0.75 g/cm<sup>2</sup>, corresponding to 1.6 Mev.

The presence of x-ray emission due to Kcapture in the Br<sup>75</sup> period was determined in the manner described in the preceding section. The ratio of K-capture processes to positron emissions in Br<sup>75</sup> from deuteron bombardment of selenium is approximately 4.4. The rate at which 1.7-hour Br<sup>75</sup> is produced by the Se<sup>74</sup>(d, n) reaction compared to the rate at which Br<sup>82</sup> is produced by Se<sup>82</sup>(d, 2n) is approximately 2.1.

No gamma-radiation, other than that due to positron annihilation, was found in the 1.7-hour Br<sup>75</sup> activity.

### THE 4.4-HOUR Br<sup>80</sup> ISOTOPE

A 4-hour  $Br^{80}$  period was first reported<sup>1</sup> as a result of slow neutron bombardment of bromine. Emission of negatively charged beta-particles in the  $Br^{80}$  period was reported<sup>2</sup> as a result of neutron and deuteron bombardments of bromine, and also<sup>8</sup> as a result of proton bombardment of

875 MASS NUMBERS 74 75 76 77 78 80 79 81 82 17.5 ,₄Se 8.3 % 19 1 49 41 "Br

FIG. 10. Se and Br section of the periodic table. The new Br<sup>77</sup> and Br<sup>75</sup> isotopes and new reactions reported are indicated by heavy lines.

selenium. It has also been reported<sup>4</sup> that most of the nuclei in the upper 4.4-hour isomeric state of Br<sup>80</sup> decay by falling into the lower 18-minute state from which observed disintegration electrons are emitted. Existence of 49 kev and 37 kev internally converted gamma-rays from the 4.4hour Br<sup>80</sup> period has been shown.<sup>5</sup>

Figures 7 and 8 show a 4.4-hour bromine  $\beta^+$ -activity resulting from proton bombardment of enriched Se<sup>74</sup> and Se<sup>76</sup>. By comparing the activities of this period with the abundance of Se<sup>80</sup> present in the two samples, it was determined that the 4.4-hour  $\beta^+$ -activity was due to the Br<sup>80</sup> period. By spectrometer measurements the end point of the 4.4-hour Br<sup>80</sup>  $\beta^+$ -activity was estimated to be approximately 0.8 Mev.

Figure 3 shows  $\beta^+$ -activity of the 4.4-hour Br<sup>80</sup> period resulting from deuteron bombardment of selenium.

The 4.4-hour Br<sup>80</sup> period was also obtained by alpha-particle bombardments of selenium.

Figure 10 shows the Se and Br section of the periodic table. The new Br<sup>77</sup> and Br<sup>75</sup> isotopes and new reactions reported are indicated by heavy lines.

#### ACKNOWLEDGMENTS

The support received from the Ohio State University Development Fund and the Graduate School is gratefully acknowledged. The significant chemical separations which were necessary in this work were very ably performed by Mr. H. L. Finston and Mr. R. M. Dyer.

<sup>&</sup>lt;sup>1</sup> M. L. Pool, J. M. Cork, and R. L. Thornton, Phys. Rev. 52, 239 (1937). <sup>2</sup> A. H. Snell, Phys. Rev. 52, 1007 (1937).

<sup>&</sup>lt;sup>3</sup> J. H. Buck, Phys. Rev. **54**, 1025 (1937).

<sup>&</sup>lt;sup>4</sup> E. Segrè, R. S. Halford, and G. T. Seaborg, Phys. Rev. 55, 321 (1939).
<sup>5</sup> G. E. Valley and R. L. McCreary, Phys. Rev. 56, 863

<sup>(1939).</sup>