

reaction for proton energies ranging from 6.0 to 6.3 Mev is  $0.95 \times 10^{-25} \text{ cm}^2$  (cf. Table I).

A long lived activity is observed in copper samples which have been subjected to long bombardments. The decay curve (cf. Fig. 8) shows a single half-life of  $235 \pm 20$  days. A cloud-chamber investigation made by Mr. George Valley<sup>12</sup> shows that the radioactive decay takes place either by positron emission or  $K$  electron capture. The negative electrons which are observed are identified as internal conversion electrons. This same activity has been obtained by bombarding zinc with deuterons<sup>13</sup> and consequently must be assigned to  $\text{Zn}^{65}$ .

<sup>12</sup> A preliminary report was made by S. W. Barnes and George Valley, *Phys. Rev.* **53**, 946 (1938). A more detailed report will be published soon.

<sup>13</sup> J. J. Livingood and G. T. Seaborg, *Phys. Rev.* **54**, 239 (1938).

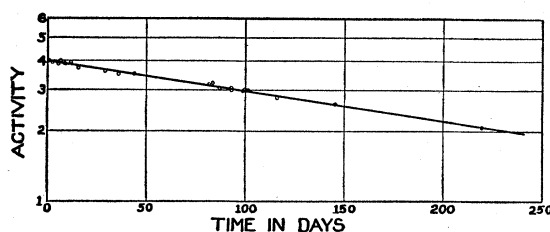


FIG. 8. Decay curve for  $\text{Zn}^{65}$ .

I wish to express my thanks to Professor L. A. DuBridge and Professor S. W. Barnes for advice given during the investigations, to Dr. J. H. Buck for help in bombarding samples and taking data, and to Mr. George Valley for the cloud-chamber photographs. The work has been supported in part by a grant from the Research Corporation.

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## Proton Induced Radioactivities

### III. Zinc and Selenium Targets

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Zinc bombarded with 6.5-Mev protons shows activities of half-lives  $18.0 \pm 0.5$  min.,  $72 \pm 4$  min.,  $9.4 \pm 0.2$  hr., and  $84.4 \pm 2.0$  hr., corresponding to the known periods of  $\text{Ga}^{70}$ ,  $\text{Ga}^{68}$ ,  $\text{Ga}^{66}$ , and  $\text{Ga}^{67}$ , respectively. In addition, a new period of  $48 \pm 2$ -min. half-life is observed and assigned to  $\text{Ga}^{64}$ . Thick target excitation curves are given for  $\text{Ga}^{64}$ ,  $\text{Ga}^{68}$ , and  $\text{Ga}^{70}$ . Selenium bombarded with 6.3-Mev protons shows activities of half-lives  $6.3 \pm 0.2$  min.,  $17.4 \pm 0.5$  min.,  $4.4 \pm 0.3$  hr., and  $33 \pm 1$  hr., corresponding to the known periods of  $\text{Br}^{78}$ ,  $\text{Br}^{80}$  (2) and  $\text{Br}^{82}$ , respec-

tively. The previously observed discrepancy between the thresholds for the two  $\text{Br}^{80}$  activities and their maximum  $\beta$ -ray energies has been confirmed. Thick and thin targets excitation curves were obtained. The cross section for the production of the  $\text{Br}^{80}$  isomers by the  $\text{Se}^{80}$  ( $p-n$ ) reaction are  $0.82$  and  $0.22 \times 10^{-25} \text{ cm}^2$  for the short and long periods, respectively, at a proton energy of 6.3 Mev. The ratio of these two cross sections rises from 3.6 at 6.3 Mev to over 200 at 3.2 Mev.

#### INTRODUCTION

THIS work is a continuation of the studies<sup>1</sup> made in this laboratory of the nuclear reactions produced by high energy protons. The present paper concerns the results obtained when targets of zinc and selenium are bombarded by protons of energies up to 6.5 Mev. Results with

these targets have already been reported<sup>1</sup> for proton energies up to 3.8 Mev. Much larger yields are obtained with the higher energy beam and additional periods are produced. Further studies of the excitation functions have therefore been made, particularly for the  $\text{Br}^{80}$  isomers produced from Se.

#### RADIOACTIVE Ga ISOTOPES FROM Zn

The complete decay curves for the radioactivity produced by 6.3-Mev protons in a pure

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<sup>1</sup> DuBridge, Barnes, Buck and Strain, *Phys. Rev.* **53**, 447 (1938).

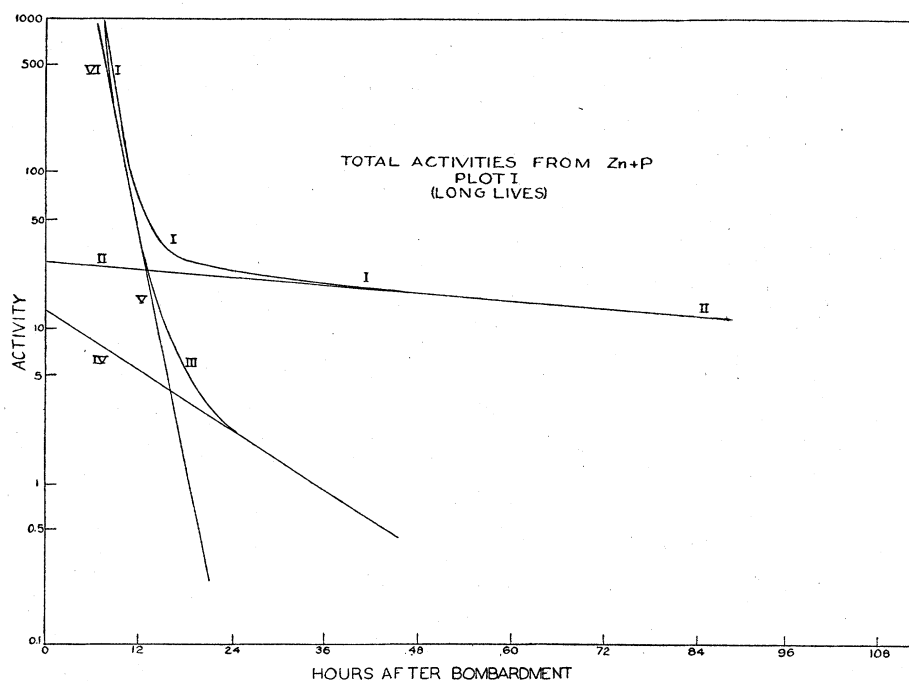


FIG. 1A. Decay of radioactivity of Zn target (long periods). I. Composite curve; II. 84.2-hr. activity; IV. 9.4-hr. activity; VI. 72-min. activity.

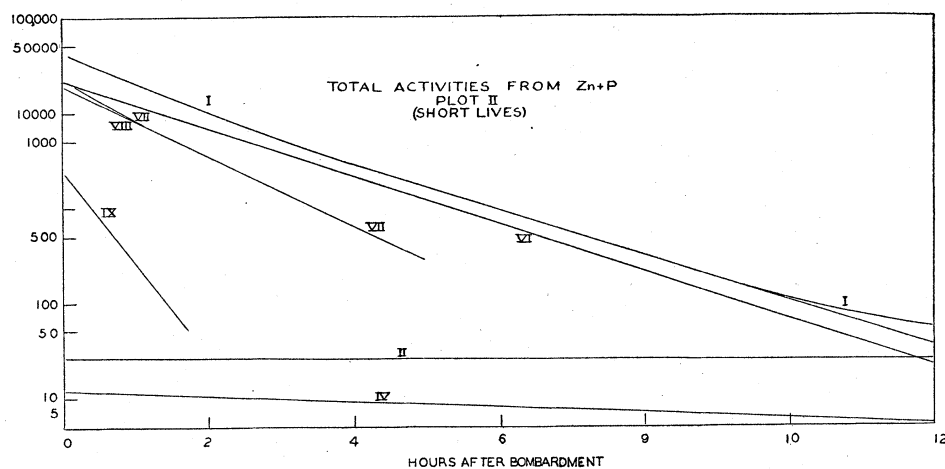


FIG. 1B. (Short periods.) VIII. 48-min. activity; IX. 18-min. activity.

Zn target are shown in Fig. 1. The curves are at once analyzed into five half-lives, all of which are found to occur only in Ga precipitates. The periods and the corresponding isotope assignments are listed in Table I.

The  $\text{Ga}^{68}$  and  $\text{Ga}^{70}$  periods were previously reported.<sup>1, 2</sup> The 84-hr. period was also reported

but was too weak to be chemically identified. Later tests showed definitely that this was due to a Ga isotope. In the meantime Mann<sup>3</sup> found the same period in the Ga precipitate from Zn bombarded by alpha-particles, and assigned the activity to  $\text{Ga}^{67}$ . Alvarez<sup>4</sup> has shown this to be

<sup>2</sup> Livingston and Bethe, Rev. Mod. Phys. 9, 245 (1937).

<sup>3</sup> Mann, Phys. Rev. 53, 212 (1938).

<sup>4</sup> Alvarez, Phys. Rev. 53, 606 (1938).

an isotope which decays by capture of a  $K$  electron, the soft electrons emitted being  $K$  and  $L$  conversion electrons from a 100-kv gamma-ray emitted in the process. These results and the assignment to  $\text{Ga}^{67}$  are consistent with our observations.

The 48-min. and 9.4-hr. periods are not formed at energies below about 4 Mev. The latter has been identified as due to  $\text{Ga}^{66}$  by Ridenour and Henderson.<sup>5</sup> The former period is new. If it were isomeric with any of the others it should certainly have been observed in previous work. We therefore assign it to  $\text{Ga}^{64}$  produced from the abundant  $\text{Zn}^{64}$  by  $p$ - $n$  reaction. This isotope cannot be formed by any other known type of process. If this assignment is correct then all of the five Zn isotopes have been converted to corresponding Ga isotopes by the proton beam. It is probable that the  $p$ - $n$  reaction is responsible in each case since results in this laboratory show that  $p$ - $\gamma$  reactions for elements in this range are improbable.

Thick target excitation curves for  $\text{Ga}^{64}$ ,  $\text{Ga}^{68}$  and  $\text{Ga}^{70}$  are given in Fig. 2 showing thresholds of 4.1, 3.7 and 1.6 Mev, respectively. Excitation curves were not obtained for  $\text{Ga}^{66}$  or  $\text{Ga}^{67}$  because of the long bombardments required. However, the  $\text{Ga}^{67}$  activity (84.2 hr.) was detectable at energies below 3.5 Mev while the  $\text{Ga}^{66}$  activity was found only above 4.1 Mev.

#### Br ISOTOPES FROM Se

The complete decay curve for the activity of a Se target bombarded by protons is shown in Fig. 3. Chemical tests show all the activity to be due to Br isotopes. All the periods have been previously reported<sup>1</sup> and identified and are

TABLE I. Radioactive Ga isotopes from Zn.

| ISOTOPE          | PERIOD             | EMITTED PARTICLE | MAX. ENERGY MEV | PROTON THRESHOLD MEV | YIELD*<br>RAD. ATOMS<br>PER 10 <sup>6</sup><br>PROTONS |
|------------------|--------------------|------------------|-----------------|----------------------|--------------------------------------------------------|
| $\text{Ga}^{64}$ | 48 $\pm$ 2 min.    | +                | (2.3)**         | 4.1                  | 5.0                                                    |
| $\text{Ga}^{66}$ | 9.4 $\pm$ 0.2 hr.  | +                | (>2.3)**        | >4.1                 | 1.1                                                    |
| $\text{Ga}^{67}$ | 84.4 $\pm$ 2.0 hr. | -(K)             |                 | <3.5                 | —                                                      |
| $\text{Ga}^{68}$ | 72 $\pm$ 4 min.    | +                | 1.85            | 3.7                  | 61.0                                                   |
| $\text{Ga}^{70}$ | 18 $\pm$ 5 min.    | —                |                 | ~1.6                 | 64.0                                                   |

\* At 6.5 Mev for a thick target of the pure Zn isotope.

\*\* Computed from the threshold.

<sup>5</sup> Ridenour and Henderson, Phys. Rev. 52, 869 (1937).

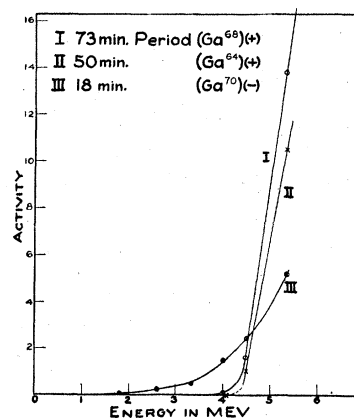


FIG. 2. Thick target excitation curves for zinc target.

formed by the  $p$ - $n$  reaction. The results are summarized in Table II.

The formation of the isomeric  $\text{Br}^{80}$  periods from  $\text{Se}^{80}$  is of particular interest. As was previously pointed out<sup>1</sup> Snell's<sup>6</sup> data on the  $\beta$ -ray energies for these two periods appear inconsistent with our threshold determinations. Thus Snell finds the 17.4-min. electron group to have a maximum energy of 2.2 Mev while for the 4.4-hr. group it is 2.0 Mev. In addition he found a soft gamma-ray ( $\sim 0.4$  Mev) accompanying the short period. This would indicate that the 17.4-min. isomer of the  $\text{Br}^{80}$  nucleus has an energy about 0.6 Mev above that of the 4.4-hr. isomer. Hence the long period would be expected to appear at lower proton energies. On the other hand, we found the threshold for the longer period to be *above* that of the shorter period by about 0.2 Mev.

More accurate and complete data on the excitation of all four Br activities have now been obtained and are plotted in Fig. 4. Curves I and II are for the two periods of  $\text{Br}^{80}$  and it is evident that the short period becomes observable at a proton energy of 2.9 Mev while the longer period first comes in at 3.1 Mev. The ratio of the observed activities (after 10 min. bombardments) is also plotted and rises from about 78 at 5.3 Mev to 2000 at 3.2 Mev. For an infinite bombardment the ratios would be 6.7 at 5.3 Mev and 182 at 3.2 Mev. A 5-hour bombardment at 3.0 Mev yielded no trace (i.e., < 1 mm) of the 4.4-hr. period but there was an initial

<sup>6</sup> Snell, Phys. Rev. 52, 1007 (1937).

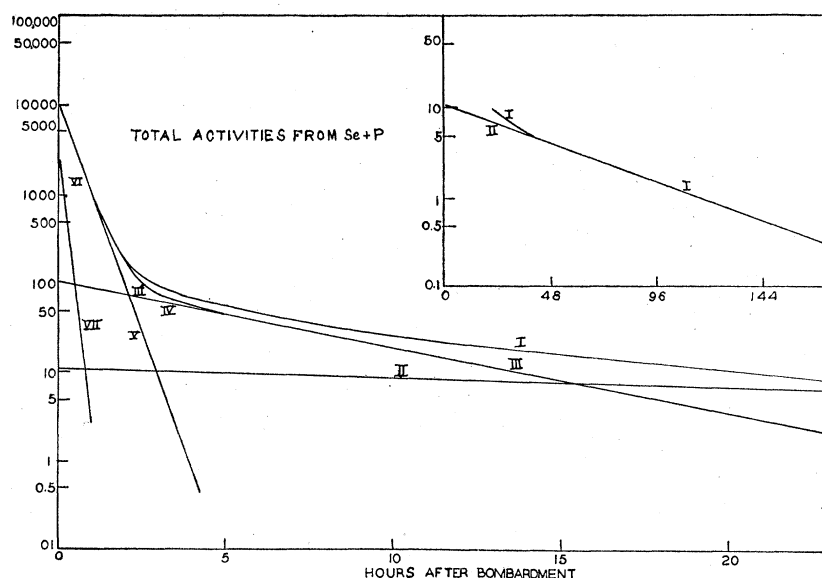


FIG. 3. Decay of radioactivity of Se target. I. Composite curve; II. 33-hr. activity; IV. 4.4-hr. activity; V. 17.4-min. activity; VII. 6.3-min. activity. (10-min. bombardment.)

activity of 20-cm deflection for the 17.4-min. period. There is thus no question but that the short period becomes observable at energies below those at which the long period can be detected.

The maximum  $\beta$ -ray energies of the isomeric activities have also been redetermined both by absorption in aluminum and from cloud chamber data. The absorption data gave 2.2 Mev and 2.0 Mev for these activities in exact agreement with Snell's values. The histograms for the distribution in curvature of cloud chamber tracks are shown in Fig. 5. The upper limits are at about 2.07 and 1.94 Mev for the short and long periods, respectively, in good agreement with the absorption data.

Snell has reported gamma-radiation with the 17.4-min. period but none with the 4.4-hr. period. In order to test this result a gamma-ray

activity curve was taken for a sample of selenium bombarded for about 1 hour with 6.3-Mev protons. About 7 mm of Al was placed between the sample and ionization chamber to absorb all  $\beta$ -rays. The resulting decay curve for the gamma-rays only is shown in Fig. 6. It is evident that the 4.4-hr. period is represented so that penetrating radiation accompanies this activity. In view of Snell's work this radiation is certainly softer than the 17.4-min. gamma-rays, so that one is still forced to conclude that the short period level of  $\text{Br}^{80}$  lies 0.2 to 0.6 Mev above the long period. The threshold for the long period would be expected then to lie this much lower than for the short period.

A possible explanation for this apparent discrepancy has been suggested by Dr. M. Goldhaber.<sup>7</sup> It is necessary to assume in accounting

<sup>7</sup> Goldhaber, private communication.

TABLE II. Br isotopes from Se.

| ISOTOPE          | PERIOD              | EMITTED PARTICLE | MAX. ENERGY MEV | PROTON THRESHOLD MEV | YIELD* (RAD. ATOMS PER $10^6$ PROTONS) | CROSS SECTION AT 6.3 MEV $\times 10^{25}$ CM <sup>2</sup> |
|------------------|---------------------|------------------|-----------------|----------------------|----------------------------------------|-----------------------------------------------------------|
| $\text{Br}^{78}$ | $6.3 \pm 0.2$ min.  | +                | 2.3             | 4.1                  | 1.4                                    |                                                           |
| $\text{Br}^{80}$ | $17.4 \pm 0.5$ min. | —                | 2.2             | 2.9                  | 5.5                                    | 0.82                                                      |
| $\text{Br}^{80}$ | $4.4 \pm 0.3$ hr.   | —                | 2.0             | 3.1                  | 0.82                                   | 0.23                                                      |
| $\text{Br}^{82}$ | $33 \pm 1$ hr.      | —                | 0.7             | <3.5                 | 2.6                                    |                                                           |

\* At 5.3 Mev for a thick target of the pure isotope.

for nuclear isomers that the two states of the nucleus have a large spin difference which forbids gamma-ray transitions. If one assumes the long period isomer to have a spin of, say, six units, then neutrons of high angular momentum will have to be emitted in the ( $p$ - $n$ ) reaction forming this isomer. This would make the reaction improbable near the threshold where the neutron energies are small and their wave-lengths large compared with nuclear dimensions. The formation of the long lived isomer will therefore not be probable until comparatively fast neutrons can be emitted. On the other hand, if the spin of the short lived isomer is zero, it will start being formed as soon as its threshold energy is reached. Thus the threshold of the 4.4-hr. activity may actually be lower than the 17.4-min. activity, but owing to the very low probability of formation no activity is detectable until the proton energy has been raised considerably above the threshold.

#### CROSS SECTION FOR $\text{Br}^{80}$ REACTIONS

To obtain data on the thin film excitation and the cross section of these reactions thin targets

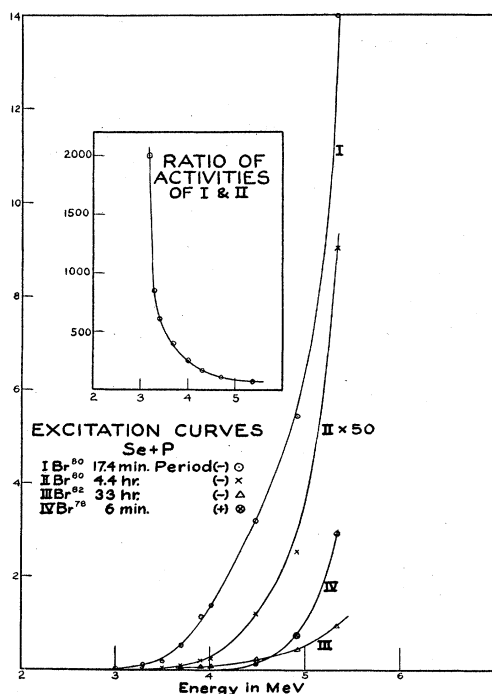


FIG. 4. Thick target excitation curves for Se (10-min. bombardment).

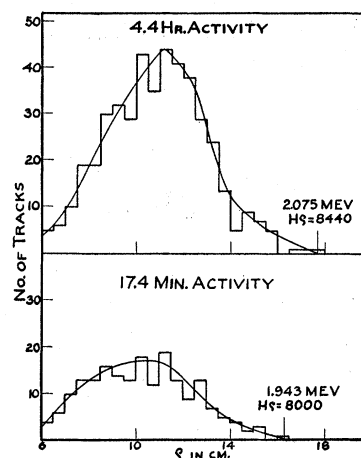


FIG. 5. Distribution of cloud chamber tracks for electrons from  $\text{Br}^{80}$ .

of Se were made by evaporating a film of selenium on 0.1-mil Al foil. From the center of the evaporated sheet ten small targets were cut and mounted. The thickness of the Se film was found by placing the ten targets in the proton beam and comparing the resulting reduction in range with that produced by Al foils alone. By this method the thickness of the Se film was computed to be 0.04 mil or 0.46 mg/cm<sup>2</sup>.

During bombardment the ten foils were "stacked," and separated by sufficient Al foil to reduce the energy of the proton beam in steps of approximately 0.25 Mev.

A 3-minute bombardment at 0.12  $\mu\text{a}$  was sufficient to bring out the 17.4-min. period at energies down to 3.5 Mev and the 4.4-hr. period down to 4.5 Mev. Longer bombardments were prevented by heating of the front foils and evaporation of the selenium.

The activity of each Se film was followed for several hours and the initial activity due to each period determined from the decay curves. These activities, corrected to infinite bombardment, are plotted against the proton energy in Fig. 7. From the calibration of the ionization chamber and the measured thickness of the foils the cross section for the production of the 17.4-min. isomer for a 6.3-Mev proton beam was found to be  $0.82 \times 10^{-25}$  cm<sup>2</sup> and for the 4.4-hr. isomer  $0.22 \times 10^{-25}$  cm<sup>2</sup>. The ratio of the two cross sections is 3.60. The ratio rises slowly with decreasing energy to 4.31 at 4.8 Mev. The above values might be in error by a factor of 2.

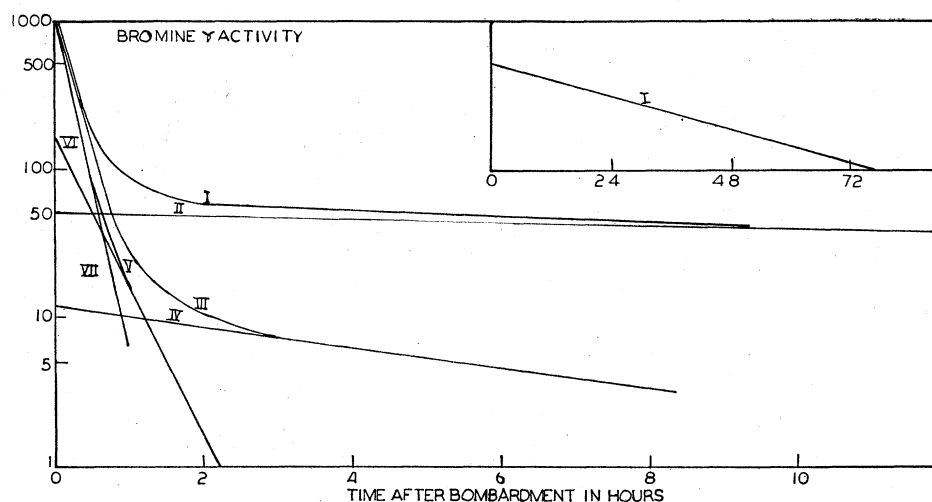


FIG. 6. Gamma-activity from  $\text{Se}+p$ . I. Composite curve; II. 33-hr. activity; IV. 4.4-hr. activity; V. 17.4-min. activity; VII. 6.3-min. activity.

This larger cross section for the short lived isomer also presents a problem if the above remarks on the spin values of two isomers are

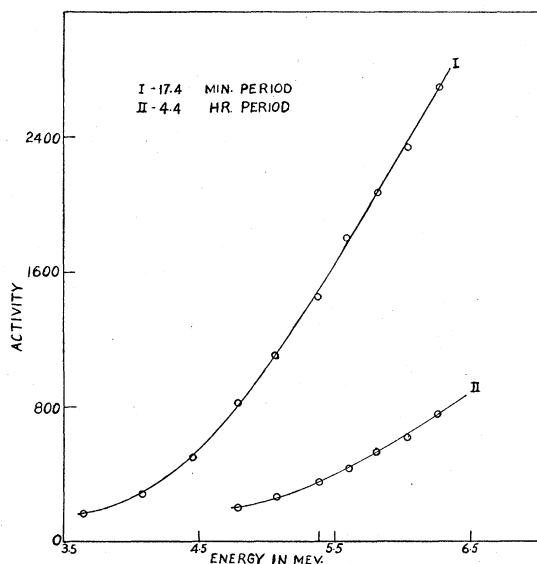


FIG. 7. Thin film excitation curves for  $\text{Br}^{80}$  periods (infinite bombardment).

accepted. If the more stable isomer has a spin value six and the less stable isomer a spin zero, then the statistical weight of the former would exceed the latter in the ratio  $(2I+1)$  or 13. The cross section for formation of the long period at high energies should then be greater than for the short period in this ratio. The evidence is, however, that as higher energies are reached the cross section ratio approaches the value of about 3 in favor of the *short* period. Obviously a simple statistical argument based on current nuclear models fails to account for these results.

In conclusion I wish to express appreciation to Professor L. A. DuBridge and Dr. S. W. Barnes for their continued interest and advice during the course of the work. I am also much indebted to Professor E. O. Wiig of the Chemistry Department who performed the chemical analyses necessary, to Dr. C. V. Strain and to Mr. George Valley for their assistance in taking the data. The work has been supported in part by a grant from the Research Corporation.