Proton Induced Radioactivities

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Results are tabulated of the radioactivities produced by 4 Mev protons in targets of $_{7}N$, $_{8}O$, $_{20}Ca$, $_{24}Cr$, $_{27}Co$, $_{30}Zn$, $_{34}Se$, $_{42}Mo$, $_{46}Pd$, $_{48}Cd$, $_{49}In$. In most cases the reactions are of the p-n type, and lead to isotopes which emit either + or − electrons. A detailed study was made of O, Zn and Se. The reaction $O^{18}(p,n)F^{18}$ (107 min.) shows a threshold at 2.56 Mev and a positron energy of 0.74 Mev in good agreement with the energy relations. The cross section for the reaction at 4 Mev is about 2×10^{-25} cm^2 and there is a resonance maximum at 3.55 Mev. The cross section for the reaction $O^{16}(p,\gamma)F^{17}$ is 4000 times smaller. The isomeric Br^{80} periods (17.4 min. and 4.45 hr.) are observed in the reaction $Se^{80}(p,n)Br^{80}$. At 4 Mev the ratio of the short to long period activities for infinite bombardment is about 15 but the thresholds are at about 3.0 and 3.2 Mev, respectively. The cross section for the reaction is about 0.6×10^{-26} cm^2 at 4 Mev.

I. Introduction

THE present report is an extension and summary of work briefly described in earlier communications[1] on nuclear reactions produced by protons of energies up to 4 Mev. Since previous observations with protons had been confined to energies below about 1 Mev our first experiments were exploratory in nature. These preliminary experiments are summarized herein together with more complete data on certain of the more interesting reactions.

The cyclotron used for this work follows closely, except for size, the design developed at Berkeley.[2] The magnet, of Armco iron, has a pole diameter of 26 inches tapered at 45 degrees to a 20-inch tip, accommodating a 20-inch accelerating chamber. A 26-inch chamber which uses the full pole diameter is now being installed. The diameter of the last ion path in the smaller chamber is 17 inches. The beam emerges through a Pt foil window (0.1 mil) into a target chamber at atmospheric pressure. With an oscillator frequency of 21 megacycles and a resonant magnetic field of about 14,000 gauss the calculated beam energy is 4.2 Mev. After emerging through the Pt window the range of the beam was about 4.8 mils of Al, corresponding to an energy of 3.9 Mev. After allowance is made for the stopping power of the Pt window the beam energy in the chamber is close to the computed value of 4.2 Mev. In some of the later experiments the beam energy was raised, by increasing the frequency, to 4.2 Mev after emergence. The proton currents were usually between 1 and 3 μa. Since the resonance conditions were chosen to accelerate protons (not molecular ions) there is no danger of deuteron or other ion contamination in the beam.

The homogeneity of a cyclotron beam is of some interest. This certainly will depend very greatly on the design of chamber, arrangement of shims and other factors. We have attempted an evaluation for our particular case by measuring the current to a collector (in the target chamber) in front of which Al foils could be inserted. The collector current is plotted against foil thickness (in mils of Al) in Fig. 1. The upper curve is for the magnetic field set at the value to give maximum total beam. For the lower curve the magnetic field was increased a few tenths of a percent above this value. The beam homogeneity is thereby greatly improved and there is an increase in intensity of the long range portion. The dashed curve (S) indicates the theoretical straggling to be expected for an initially homogeneous beam. The observed straggling is about three times greater, which could be accounted for by a 4 percent energy spread (half-width) in the beam. Since, however, observed straggling even for a homogeneous beam usually exceeds the theoretical by a factor of 1.5 to 2 we may conclude that the actual energy spread of the primary proton beam is not more than ±2 percent from the mean, that is, ±80 kv at a mean value of 4 Mev.[2a] Further

[1] S. W. Barnes, L. A. DuBridge, *et al.*, Phys. Rev. **51**, 775 (1937); Phys. Rev. **51**, 995 (1937); Phys. Rev. **51**, 1012 (1937).

[2] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 608 (1934); E. O. Lawrence and D. Cooksey, Phys. Rev. **50**, 1131 (1936).

[2a] Cf. M. C. Henderson and M. G. White, Rev. Sci. Inst. **9**, 19 (1938).

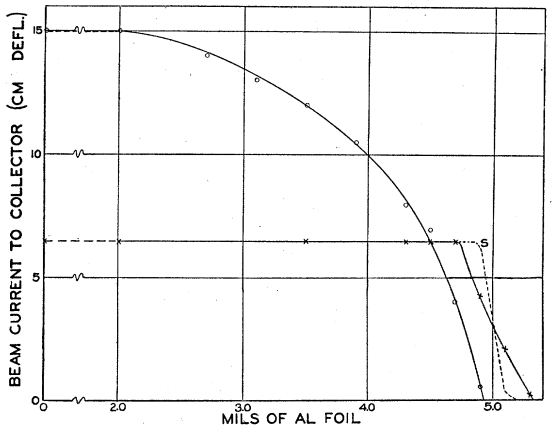

FIG. 1. Curves showing degree of homogeneity of proton beam from cyclotron. Open circle, resonance field; cross, field above resonance; S, theoretical straggling.

evidence for homogeneity will be found in the excitation curve for the O^{18} p-n reaction to be described below which shows a resonance peak whose half-width is about 100 kv.

Most of the activities herein reported were measured with an ionization chamber and amplifier unit of the type described by Hafstad.[3] The chamber was filled with nitrogen at 1 atmosphere. Its sensitivity as compared with a Geiger-Müller counter is such that 100 counts/min. on the counter corresponds to a 1 cm deflection of the amplifier-galvanometer. The sensitivity to β-rays, periodically checked against a constant natural source, is such that a 1 cm steady deflection is produced by the passage through the chamber of about 12 β-rays per sec. The solid angle subtended is such as to collect 1/10 of the β-rays from a given source. For strong samples the sensitivity is quickly changed by the galvanometer shunt. The initial activities of the proton-bombarded samples varied widely from a fraction of a microcurie to about 0.1 millicurie but were generally of the order of 10 microcuries.

II. General Discussion of Proton Reactions

In the energy region below 1 Mev two types of proton reactions are known,[4] namely, p-γ and p-α. (A p-d reaction is observed in the single case Be^9 (p-d) Be^8.) In our earliest work[1] radioactivities were observed in a number of cases where p-γ and p-α reactions would lead only to stable isotopes. These could be explained by assuming a p-n type reaction. This was confirmed by chemical tests, by the identification of certain known periods, and by observation of neutrons produced by the proton beam. A more detailed study of this new type of reaction was then made.

The type reaction is

$$Z^A+H^1\rightarrow(Z+1)^A+n^1.$$

Since neighboring isobaric isotopes are almost unknown in the periodic table the product nucleus $(Z+1)^A$ will almost always be unstable and will either emit positrons or electrons.

Case A. Positron emission

$$(Z+1)^A\rightarrow Z^A+e^+. \quad (1)$$

The mass-energy relation becomes

$$Z^A+H^1+E_p=(Z+1)^A+n^1+E_n$$
$$=Z^A+e^++(e^-)+n^1+E_n+E_+,$$

where E_p=proton energy, E_n=neutron energy, E_+=maximum positron energy.

The minimum value of E_p will be that for which $E_n=0$. Hence

$$E_{p0}=n^1-H^1+2e+E_+=1.80+E_+ \text{ Mev}. \quad (2)$$

Also

$$E_{p0}=(n^1-H^1)+[(Z+1)^A-Z^A]$$
$$=0.78+\Delta_1 \text{ Mev}, \quad (3)$$

where we have taken $(n^1-H^1)=0.00084$ mu[5] $=0.78\pm0.07$ Mev and Δ_1 stands for the mass difference of the two adjacent isobars (expressed in Mev). If E_+ is known the threshold for the reaction, E_{p0}, is determined and the measurement of either E_+ or E_{p0} determines Δ_1. Since positron energies usually lie between 0.2 and 5 Mev the threshold for this reaction will never be less than about 2 Mev and in most cases not greater than about 7 Mev.

Case B. Electron emission

$$(Z+1)^A\rightarrow(Z+2)^A+e^-. \quad (4)$$

[3] L. R. Hafstad, Phys. Rev. **44**, 201 (1933).
[4] M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 245 (1937). Hereafter referred to as LB.
[5] See LB. p. 373.

In this case the mass relations are

$$E_{p0}=(n^1-H^1)+[(Z+1)^A-Z^A]$$
$$=0.78+\Delta_1 \text{ Mev} \quad (5)$$

and

$$E_{p0}=(n^1-H^1)+E_-+[(Z+2)^A-Z^A]$$
$$=0.78+E_-+\Delta_2 \text{ Mev}, \quad (6)$$

where Δ_2 is the mass difference between the isobars differing by 2 in atomic number. Evidently $\Delta_1-\Delta_2=E_-$. In this case measurements of E_{p0} and E_- will determine the mass differences of three isobaric isotopes. Since stable isobars differing by 2 in atomic number are commonly both stable the mass difference Δ_2 may be expected to be small. Hence the threshold for the reaction will be of the order of $(0.78+E_-)$ Mev, e.g., from 1 to 4 Mev. For the same β-ray energy the threshold may then be 1 Mev lower than for the positron emitting case.[6] Examples of these relations will be considered below.

Results

Of the large number of activities found in the first exploratory work with high energy protons three elements, O, Se, and Zn, showed such strong activity that they were selected for more detailed study. These will be discussed first, followed by a tabular summary of other reactions.

A. Oxygen

Targets containing oxygen in any form when exposed to the proton beam show two strong activities of 1.23 ± 0.1 min. and 107 ± 4 min. The first checks with the known[7] period of F^{17} which is probably formed by the reaction $O^{16}(p,\gamma)F^{17}$. The second is in agreement with the period for F^{18} formed by Snell[8] in the reaction Ne-*d*-α and by Pool, Cork and Thornton[9] in the reaction F-*n*-2*n*. This period appears only in the fluorine precipitate (CaF_2 from bombarded NaOH). The active F^{18} isotope may be produced by proton bombardment of oxygen by either of two reactions: $O^{17}(p,\gamma)F^{18}$ or $O^{18}(p,n)F^{18}$. The former can hardly contribute more than a small fraction of the total F^{18} activity, as will be shown below. As a further test we have bombarded samples of water, kindly supplied to us by Professor H. C. Urey, in which the O^{18} concentration was about 4.5 times the normal. The 107-min. activity produced in this water was between 3 and 4 times as great as in ordinary water, in rough agreement with the concentration ratio. The difficulty of handling liquid targets and of avoiding activity due to contamination of the cell by air and water vapor have precluded more precise comparisons. Since Dr. Urey states that the O^{17} concentration is only about twice normal in this water the results give strong evidence for attributing the 107-min. activity to the reaction involving O^{18}.

Excitation functions.—The thick target (quartz) excitation functions for the F^{17} and F^{18} periods are shown in Fig. 2, each corrected for the finite bombardment time. It is seen that the F^{17} period (1.23 min.) can be detected at energies as low as 1.6 Mev while the F^{18} activity sets in sharply at about 2.6 Mev and then rises so rapidly that at 4 Mev it is four times stronger than the F^{17}. This indicates that two different types of process must be in operation. The $O^{17}(p\text{-}\gamma)F^{18}$ process would be expected to behave in the same way as the $O^{16}(p\text{-}\gamma)F^{17}$, the latter being stronger by a factor of 2500, the ratio of abundance. We attribute the main part of the F^{18} activity therefore to the O^{18}-*p*-*n* reaction, and the energy relations to be given presently confirm this hypothesis.

By the method of stacked Al foils, bombarded in O_2, the thin-film excitation curve for the F^{18} activity was obtained as shown in Fig. 3. The lower curve (triangles) was obtained with a homogeneous initial beam of 3.8 Mev and shows a pronounced peak at 3.55 Mev. This curve was repeated many times with different foils and foil arrangements. Later when the initial energy was raised to 4.1 Mev the upper curve (circles) was repeatedly observed. Since the high energy beam was less homogeneous and since the straggling was also greater in the thicker absorbing layers the peak is flattened out as would be expected. This peak is reflected in the thick target curves where it is found also for the O^{16}-*p*-γ reaction. Its

[6] Cf. LB. p. 315.

[7] The data are tabulated by L. N. Ridenour and W. J. Henderson, Phys. Rev. **52**, 889 (1937). We have not yet accounted for our failure to obtain a closer check on the value 1.07 ± 0.1 min. for this period obtained in the careful experiments of these authors. The limits of error however just overlap.

[8] A. H. Snell, Phys. Rev. **51**, 142 (1937).

[9] M. L. Pool, J. M. Cork and R. L. Thornton, Phys. Rev. **52**, 239 (1937).

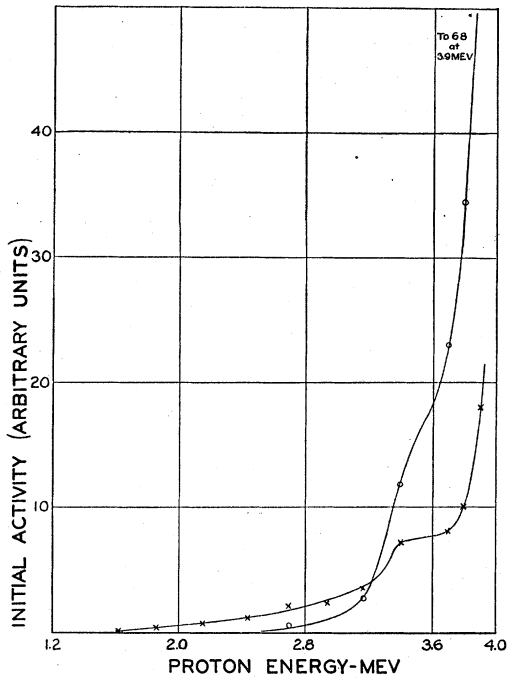

FIG. 2. Thick target excitation curves for the F^{17} and F^{18} periods each corrected for the finite bombardment time. O=F^{18}, ×=F^{17}.

position is considerably above the top of the potential barrier (2.4 Mev according to Bethe's formula[10]). It may simply represent a flattening of the rise in penetration probability or may indicate a resonance level. The rapid rise of the yield above 3.7 Mev suggests further levels above the barrier.

When corrected for bombardment time the activity at the peak of the curve (3.55 Mev) corresponds to the production of about one F^{18} atom per 10^9 protons. The effective range of the F^{18} recoil atoms is calculated (following Newson[11]) to be about 0.075 cm. The cross section for the reaction at 3.55 Mev (taking into account the abundance of O^{18}) is thus roughly 0.75×10^{-25} cm². At 4 Mev it is about three times larger. The average cross section calculated from the solid target data is about 0.6×10^{-25} cm². The latter value is about 4000 times greater than for the $O^{16}(p,\gamma)F^{17}$ reaction, the cross section for which is 0.15×10^{-28} cm². The F^{17} activity is four times weaker than the F^{18} though the abundance ratio favors the former by a factor of 500 and the effective beam range is twice as great.[12]

Energy relations.—The extrapolated threshold for the production of F^{18} is found to be close to 2.56 ± 0.04 Mev. From Eq. (4) we have then

$$E_+=2.56-1.80=0.76\pm0.07 \text{ Mev}.$$

The positron absorption curve in Al is shown in Fig. 4. The end point corresponds to a maximum energy of 0.74 Mev in excellent agreement with the prediction. This agreement incidentally furnishes good evidence for the accuracy of the accepted mass difference $(n^1-H^1)=0.78$ Mev.

Applying Eq. (3) we obtain the mass of F^{18}

$$F^{18}=O^{18}+1.76 \text{ Mev}$$
$$=18.00557\pm0.00025 \text{ mass units},$$

if the accepted value[5] of 18.00369 for O^{18} is used. The uncertainty in the mass of F^{18} is due mainly to the uncertainty in this value.

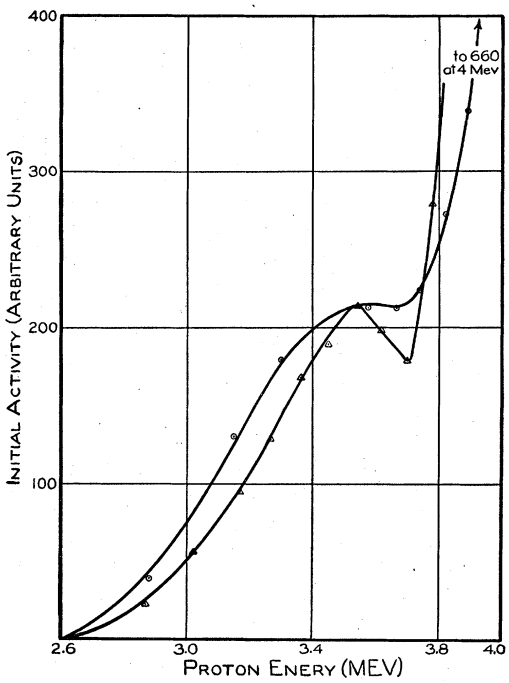

FIG. 3. Curve for thin-film excitation for the F^{18} activity. Lower curve (triangles) for 3.8 Mev protons; upper curve (circles) 4.1 Mev protons.

[10] H. A. Bethe, Rev. Mod. Phys. **9**, 172 (1937).
[11] H. W. Newson, Phys. Rev. **51**, 620 (1937).

[12] In view of this large ratio one might assume the F^{17} to be produced by the reaction $O^{17}(p,n)F^{17}$. But since the positron energy for F^{17} is 2.1 Mev this reaction is energetically excluded for protons below 3.9 Mev, though it actually occurs down to 1.6 Mev.

B. Zinc

Proton bombardment of Zn results in a strong radioactivity which is resolved into three periods: 18.2 min. (electron), 68 min. (positron) and 82 hour (electron). The two short periods appear in the Ga precipitate and are at once identified with the known periods of Ga^{70} and Ga^{68}, respectively. The reactions are then $Zn^{70}(p\text{-}n)Ga^{70}$ (18.2 min.) and $Zn^{68}(p\text{-}n)Ga^{68}$ (68 min.). Ga^{68} might also be formed by the reaction $Zn^{67}(p\text{-}\gamma)Ga^{68}$ but this would appear to be less probable.

The thick target excitation function for the electron-emitting Ga^{70} (18.2 min.) activity shows a threshold at about 1.6 Mev. In contrast, the positron emitting isotope Ga^{68} is not produced at proton energies under about 3.6 Mev. The positron energy as recently measured by Ridenour and Henderson[7] is 1.85 Mev leading to an expected threshold of 3.63 Mev.

The 82-hour period which is always present in activated Zn does not come down with Cu, Zn, Ga or Ge precipitates. It is probably due to an impurity though we find this period in none of the most probable impurity elements so far investigated.

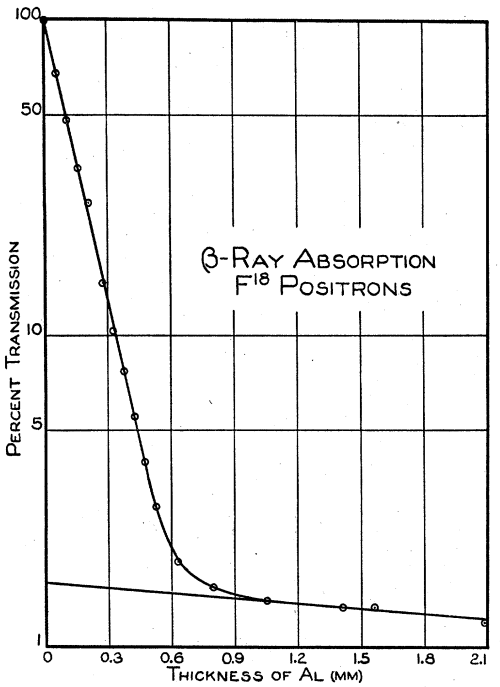

FIG. 4. Positron absorption curve in aluminum.

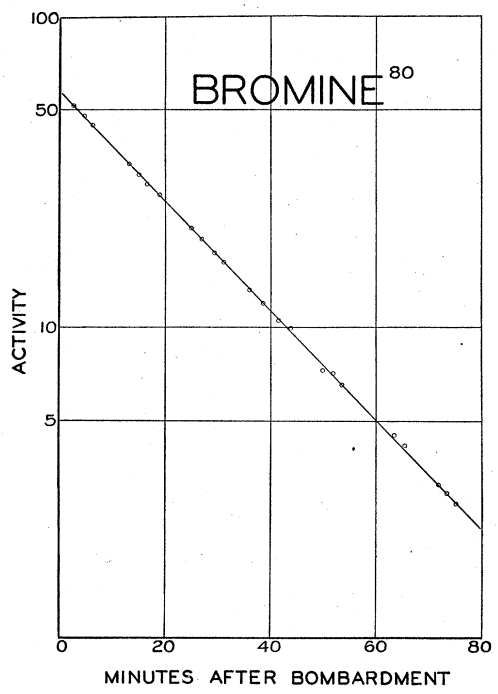

FIG. 5. Decay curve for the activity of Br^{80}. (17.4 min.)

C. Selenium

The strongest radioactivity so far found in proton bombardment appears in Se. A ten-minute bombardment with 2 μa of 4 Mev protons gives an initial activity of the order of 0.1 millicurie. The decay curves show three periods, all electron-emitting: 17.4 ± 0.25 min., 4.45 ± 0.10 hr. and 33.0 ± 1.0 hr. The two short periods are at once identified with the isomeric Br^{80} and the long period with Br^{82}. The values are in good agreement with Snell's[13] except for the 17.4-min. period which he reports as 18.5 ± 0.5. We have taken repeated runs on this period using a 5 sec. bombardment which is sufficient to bring out this short period with no detectable presence of the longer ones. One such decay curve is shown in Fig. 5 and all runs give an average value of 17.4 min. with a computed probable deviation of 0.25 min.

Since the 79 and 81 isotopes of Se are not stable the above periods must be attributed to the reactions $Se^{80}(p\text{-}n)Br^{80}$ and $Se^{82}(p\text{-}n)Br^{82}$. One might expect to observe the 6.4-min. positron activity of Br^{78} formed from the abundant Se^{78}.

[13] A. H. Snell, Phys. Rev. **52**, 1007 (1937).

However, Snell finds the positron energy to be 2.3 Mev giving a predicted threshold of 4.1 Mev which is just over the limit of our beam.

The excitation functions (thick target) of the two Br^{80} periods are shown in Fig. 6. The cross section for the 17.4-min. period is about 0.6 $\times 10^{-26}$ cm^2 for a primary energy of 4 Mev. It is of interest that the thresholds for the two periods of Br^{80} differ by about 0.2 Mev. The inset curve showing the ratio of the two activities give evidence that this threshold difference is real. This "branching ratio" increases with decreasing energy, going to infinity at 3.2 Mev which is taken as the threshold for the 4.5-hr. period. At 4 Mev the branching ratio of short to long period, corrected for bombardment time, is about 15.

From Eq. (6) it is evident that since the initial and final isotopes (Se^{80} and Kr^{80}) are the same for the two periods of Br^{80}, there should exist a difference in β-ray (or γ-ray) energy of

TABLE I. *Summary of proton induced radioactivities.*

Target	Obs. Periods	Emitted Particles	Active Isotope	Reaction	Chem. Sep.	Previously Obs. Periods	Produced by	Ref.
7 N	125 sec.	(+)	O^{15}	N^{14}-p-γ (N^{15}-p-n)		126 sec.	N-d-n O-γ-n O-n-$2n$	LB
8 O	74 sec.	+	F^{17}	O^{16}-p-γ		64–78 sec.	N^{14}-α-n O^{16}-d-n	7
	107 min.	+	F^{18}	O^{18}-p-n	Yes	112 min. 108 min.	Ne^{20}-d-α F^{19}-n-$2n$	8 9
20 Ca	~70 min. ~3.3 hr. >32 hr.	(+) (+) (+)	Sc^{41} $Sc^{42, 43}$ Sc^{44}?	Ca^{40}-p-γ Ca-p-n(?) Ca^{44}-p-n		53 min. 4.0 hr. 52 hr.		16 16 16
24 Cr	42 min.	(+)	Mn^{53}	Cr^{53}-p-n (Cr^{52}-p-γ)		46 min.	Cr^{52}-d-n	LB
27 Co	2.5 hr.		Ni(?)	Co-p-n		2 hr. (Ni^{57}?) 3 hr. (Ni^{59})	Ni^{58}-n-$2n$(?) Ni^{58}-n-γ	9 LB
30 Zn	18.2 min.	−	Ga^{70}	Zn^{70}-p-n	Yes	20 min.	Ga-n-γ Ga-γ-n	LB
	68 min.	+	Ga^{68}	Zn^{68}-p-n (Zn^{67}-p-γ)	Yes	68 min.	Cu-α-n	7
	82 hr.	−	?	?	Not Cu Zn, Ga			
34 Se	17.4 min.	−	Br^{80}	Se^{80}-p-n	Yes	18.5 min.	Br-d-p	11
	4.45 hr.	−	Br^{80}	Se^{80}-p-n	Yes	4.54 hr.	Br-d-p	11
	32.8 hr.	−	Br^{82}	Se^{82}-p-n	Yes	33.9 hr.	Br-d-p	11
42 Mo	30 sec.		?					
	31 min.		?					
46 Pd	2.41 min.	(−)	Ag^{108}	Pd^{108}-p-n		2.3 min.	Ag-n-γ	LB
	24.4 min.	(+)	Ag^{106}	Pd^{106}-p-n (Pd^{105}-p-γ)		24.5 min.	Ag-n-$2n$	14
48 Cd	70 sec. 6 min.? 37 min.? 128 min.		In^{112}? In^{117}?	(Cd^{112}-p-n) (Cd^{116}-p-γ)		72 sec. 138 min.	In-n-$2n$ Cd-d-p	15 15
49 In	14 min.		Sn^{113}?	In^{113}-p-n		18 min.?	Sn-n-γ Sn-d-p	LB

[14] M. L. Pool, Phys. Rev. **52**, 380 (1937).
[15] J. M. Cork and R. L. Thornton, Phys. Rev. **51**, 608 (1937); **52**, 531 (1937).
[16] H. Walke, Phys. Rev. **51**, 439 (1937).

0.2 Mev. Such a difference has been found by Snell,[13] who gives 2.0 and 2.2 Mev for the β-ray energies for the long and short periods, respectively. However, *this is in the wrong direction* to account for the threshold difference, since it would predict a higher threshold for the short period. The situation is further complicated by Snell's observation that γ-rays accompany the 2.2 Mev electrons of the short period but there are no detectable long period γ-rays. These results are incompatible with the lower threshold for the short life activity unless an unusual isomerism in the excited states of the intermediate nucleus Br^{81} is assumed. Discussion of this possibility is best postponed until the experimental data are made more precise.

From Eqs. (5) and (6) one finds for the mass differences

$$Br^{80}-Se^{80}=2.2\pm0.2 \text{ Mev}$$

and $$Kr^{80}-Se^{80}=0.0\pm0.2 \text{ Mev}.$$

D. Other reactions

All the radioactive periods so far found with proton bombardment are listed in Table I. Periods obviously due to impurities have been eliminated but in many cases the activity is so weak that chemical tests have not been made and the reactions in some cases are doubtful (indicated by (?)). In other cases identification is possible by comparison with known periods. The results listed in the table supersede the preliminary values sent to Livingston and Bethe for inclusion in their article.[17] The weak 90–110-min. activity found in Mn and attributed in our first reports[1] to Fe^{55} has been traced to oxygen impurity (F^{18}), and we can now find no activity assignable to Fe^{55}. The 109-min. period in As listed in the table might also be due to F^{18} but the β-ray spectrum for the activated As seems to differ appreciably from the F^{18} spectrum and we assign this period tentatively to Se^{75}. The other listings are self-explanatory. Further work is now in progress.

Acknowledgments

The construction of the cyclotron and related equipment and its continued operation have been made possible by the generous cooperation of many individuals and corporations, to all of whom grateful acknowledgment is made. We wish particularly to express our indebtedness to the Rochester Gas and Electric Company, the Rochester Telephone Company, the Stromberg-Carlson Telephone Manufacturing Company, the Consolidated Machine Tool Corporation, the General Electric Company, the Bell Telephone Laboratories, the American Rolling Mill Company, and the Eastman Kodak Company for supplying equipment, materials, and services at reduced or no cost; to the Research Corporation for financial support; to Dr. A. G. Hill, Mr. George Valley, Mr. Tom Perry and Mr. Arthur Gibson, mechanician, for assistance in construction and operation. We are indebted to Drs. E. O. Lawrence, M. S. Livingston and Donald Cooksey for much valuable advice and assistance in planning and building the cyclotron.

Finally we wish to acknowledge the valuable assistance generously given by Professor E. O. Wiig of the chemistry department in planning and carrying out all chemical analyses.

[17] LB. p. 317.

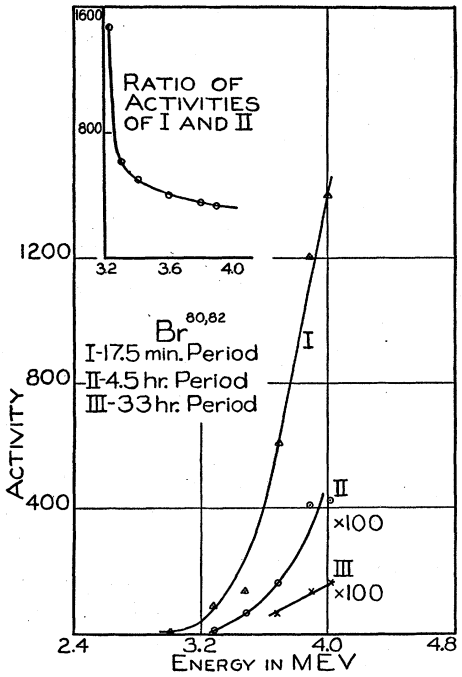

FIG. 6. Excitation functions of the two decay periods of Br^{80}.