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The Radioactive Isotopes of Bromine: Isomeric Forms of Bromine 80

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A study of the radioactive isotopes of bromine has been carried out by cross checking a number of nuclear reactions induced by bombarding the elements arsenic, selenium, bromine, krypton and rubidium with neutrons, deuterons, and electrically accelerated α-particles. The results indicate that the active isotope bromine 80 exists in two isomeric forms, from which it decays with the respective periods of 18 minutes and 4.5 hours. An example of two successive β-transitions is found to occur in the active isotopes selenium 83 and bromine 83, and a new type of transmutation is observed in which the emission of two neutrons apparently follows deuteron capture, the reaction in this case being Se^{82} $(D, 2n)$ Br^{82}. The discovery of a number of new radioactive isotopes and the identification of isotopes which carry some of the already-known activities have led to results which may be summarized in the following table of decay periods:

Arsenic 78:	65±3 min.
Selenium 83:	17±5 min.
Selenium 79 or 81:	57±1 min.
Bromine 78:	6.4±0.1 min. (positron emitter)
Bromine 80:	18.5±0.5 min. and 4.54±0.10 hrs.
Bromine 82:	33.9±0.3 hr.
Bromine 83:	2.54±0.10 hr.
Krypton:	74±2 min., 4.5±0.1 hrs. and 18±2 hrs.
Rubidium 86 and 88:	18±2 min., 18±1 days (not necessarily respectively).

All of these activities emit negative electrons except bromine 78 and possibly the krypton 18-hr. activity, which is very weak. Absorption measurements for the β-rays of the bromine isotopes have given the following values for their maximum energy: Br^{78} (6.4 min.) 2.3 Mev; Br^{80} (18 min.) 2.2 Mev; Br^{80} (4.5 hrs.) 2.0 Mev; Br^{82} (33.9 hrs.) 0.4 Mev; Br^{83} (2.45 hrs.) 1.3 Mev. The emission of γ-rays accompanies the activity of Br^{78}, Br^{80} (18 min.) and Br^{82}, but is not found with Br^{80} (4.5 hrs.) nor Br^{83}.

IN 1935 Fermi and his collaborators[1] bombarded bromine with neutrons, and discovered two induced activities which emitted negative electrons, and decayed with periods of 18 min. and 4.5 hrs., respectively. It was shown chemically that these belonged to isotopes of bromine, and it was accordingly surmised that they were due to bromine 80 and 82, formed by simple neutron capture from the two stable bromine isotopes 79 and 81. Later Kourchatow, Kourchatow, Myssowsky and Roussinow[2] found that a third activity also is induced by the action of neutrons upon bromine; this activity emitted soft β-particles, and had a decay period of 35 hrs. It also was found to be chemically inseparable from bromine. This third activity was found also by Johnson and Hamblin.[3] Meanwhile Blewett[4] made a search with the mass spectrograph for a third stable isotope, the presence of which would account for the existence of one of the bromine radioactivities. His results indicated that no other isotope (excluding bromine 78, 80 and 82, which if stable would be of no assistance in explaining the extra induced activity) could be present to an extent greater than 1/3000 of the equally abundant isotopes 79 and 81. Therefore,

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[1] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti and Segrè, Proc. Roy. Soc. **A149**, 522 (1935).

[2] Kourchatow, Kourchatow, Myssowsky and Roussinow, Comptes rendus **200**, 1201 (1935).

[3] Johnson and Hamblin, Nature **138**, 504 (1936).

[4] Blewett, Phys. Rev. **49**, 900 (1936).

if one of the activities resulted from neutron capture by a third, rare stable isotope, the effective cross section for capture must be very large—a possibility which at the time could not be definitely excluded, in view of the large cross sections known to be possessed by a few elements for slow neutron capture. A different solution was proposed both by Kourchatow, Kourchatow, Myssowski and Roussinow and by Johnson and Hamblin, who suggested that one of the activities might be due to bromine 78, formed from bromine 79 by neutron capture followed by the expulsion of two neutrons according to the reaction $\mathrm{Br}^{79}(n, 2n)\mathrm{Br}^{78}$.

The present investigation was undertaken in an attempt to clarify the situation by bombarding bromine and its adjacent elements with deuterons, neutrons and artificially accelerated α-particles. It was hoped that some light could be thrown on the problem if bromine 78, 80 or 82 could be formed independently by transmutations from arsenic, selenium, krypton, or rubidium. The experiments led to the conclusion that the only satisfactory explanation of the occurrence of the three bromine activities lies in ascribing two activities to bromine 80. These are the 18-min. and the 4.5-hr. activities; the 35-hr. activity appears to belong to bromine 82. During the course of the work evidence has been obtained for two other interesting phenomena; *viz.* the emission of two neutrons following deuteron capture, and the growth of a radioactive bromine from a radioactive selenium. The latter discovery is interesting mainly insofar as it is one of the first examples of successive β-transformations found in the field of induced radioactivity.[5] Two new radio-bromines have been discovered, as well as some new active isotopes of arsenic, selenium, krypton and rubidium. The latter are merely mentioned, in anticipation of their fuller study later. These results were presented at the Washington Meeting of the American Physical Society,[6] and with respect to the isomeric forms of bromine 80 they are in agreement with the photodisintegration results briefly noted by Bothe and Gentner[7] near the conclusion of the present investigation.

EXPERIMENTAL

The Berkeley cyclotron[8] served as a source of the deuterons, α-particles and neutrons used in activating the various samples. Except when otherwise stated, the neutrons were made by the deuteron-beryllium reaction. The deuteron beam had an energy of about 5.5 Mev, and amounted usually to between 10 and 20 microamperes. The α-particle beam was usually easily resolvable (by means of the magnetic field control) from the deuteron beam arising from residual deuterium contamination of the helium in the accelerating chamber, and the freedom of the samples from deuteron-induced activities attested its purity. Usually it amounted to 0.2μA at 11 Mev.

Arsenic and selenium were bombarded in the metallic form, and bromine was used in the form of bromides of lead, potassium and sodium. Ammonium bromide was used for neutron activation, but is not suitable for direct deuteron bombardment; this was found to be true also for lithium bromide, but the latter has the added disadvantage of being hydroscopic. All targets were chemically pure. A quartz-fiber electroscope was used to follow the decay of the samples and also to obtain absorption curves of the β-rays. The activities of the samples are here given in the arbitrary units of "electroscope divisions per second," but their absolute activities can be roughly gauged from the estimate that a fall through one electroscope division corresponds to a total emission of about 10^4 β-particles from the sample. The sign of the emitted particles was measured with a crude but adequate magnetic deflector used in air in conjunction with the electroscope: whenever necessary the results were checked by means of the cloud chamber.

Chemical separations of arsenic, selenium, bromine and krypton were accomplished in more than one way. When a separation was required from an activated bromine sample, the sample was usually dissolved in hot water,

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[5] Cork and Thornton (Phys. Rev. **51**, 608 (1937)) have recently described another case found in cadmium and indium.

[6] April 29, 1937. For abstract see Phys. Rev. **51**, 1011 (1937).

[7] Bothe and Gentner, Naturwiss. **25**, 284 (1937) (April 30), and Zeits. f. Physik **106**, 235 (1937).

[8] Lawrence and Cooksey, Phys. Rev. **50**, 1131 (1936).

to which traces of ammonium selenite and arsenious anhydride had previously been added. The selenium was then precipitated in the metallic form by the addition of hydrazine sulphate. Boiling accelerated this precipitation, and also drove off any active krypton which might have been present. The bromine was next removed as AgBr, and finally the arsenic was taken out by a sulphide separation. When selenium was activated, the procedure was slightly different, but either of the two following methods was found to be adequate: (1) the sample was dissolved in boiling, concentrated nitric acid, to which a little lead bromide and arsenic had been added, and the evolved gases were passed through an ammoniacal solution of silver nitrate. Neutralization of this solution caused the complete precipitation of the bromine in the form of AgBr. The selenium in the acid solution was then precipitated by neutralizing the solution and adding hydrazine sulphate, and the arsenic was brought down with H_2S. (2) The selenium was fused with sodium peroxide and dissolved in water. The solution was neutralized with HNO_3 and the separation proceeded as above. If the bromine fraction was required before the selenium, the solution was acidified, and the bromine was precipitated by adding $AgNO_3$. The acid prevented the simultaneous precipitation of the selenium in the form of silver selenite. In most of this work the major interest lay in the bromine and selenium fractions; when it is desired to study the arsenic fraction more closely methods of separation which are more specific for arsenic undoubtedly will have to be used.

Bromine 78

As noted above, it has been suggested that one of the three neutron-induced bromine activities might be attributed to bromine 78, formed according to the reaction $Br^{79}(n, 2n)Br^{78}$. It seemed attractive to test this hypothesis by preparing bromine 78 in a different way, and a very direct method which suggested itself was to bombard arsenic with α-particles in the hope of inducing the reaction $As^{75}(\alpha, n)Br^{78}$. (This is reaction (1) as given in the isotope scheme given of Fig. 10.) The chances of obtaining a conclusive answer to the question were unusually good, because arsenic has only one stable isotope, and unless a reaction of a novel kind were to take place, bromine 78 should be the only unstable transmutation product formed.

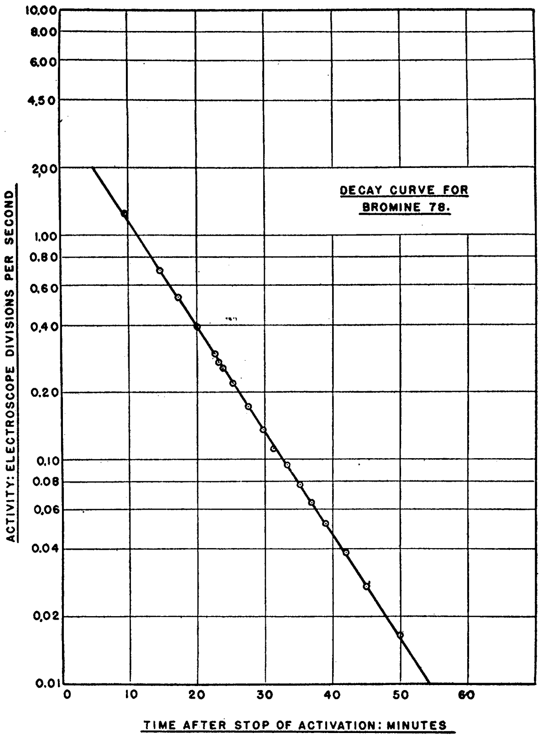

FIG. 1. Decay curve for bromine 78, made by α-particle bombardment of arsenic. Positive electrons are emitted, the decay period being 6.4±0.1 minutes.

Metallic arsenic was bombarded with a beam of $0.2\mu A$ of 11 Mev α-particles. The sample showed an activity which was fairly strong compared to other activities which have been induced in this laboratory with α-particles, and it was possible to follow the decay for six half-lives. The decay curve of the sample (see Fig. 1) showed that a single radioactive isotope was formed, but unlike any of the familiar bromine activities, it had a half-life of 6.4±0.1 minutes. Observation of the particles in the cloud chamber, and by means of a Thibaud magnetic focusing device, indicated that 95 percent or more were positives.[9] Absorption measurements were made, the results of which are given in Fig. 2. The end-point for transmission of the

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[9] I am indebted to J. R. Richardson and E. M. Lyman for help in these experiments.

positrons is seen to occur at an absorber thickness of about 4.1 mm of aluminum. According to Feather's[10] empirical formula "range in grams per $\mathrm{cm}^2 = 0.511$ (energy in Mev) -0.091," this means that they had an upper energy limit of about 2.3 Mev. The γ-ray background indicated in the figure was due, in part at least, to the annihilation radiation from the positive electrons.

A simple chemical separation was carried out to make sure that the activity was due to an isotope of bromine. Arsenic trioxide[11] was activated in the cyclotron, and then dissolved in sodium hydroxide solution, to which a little KBr had previously been added. The solution was made slightly acid with HNO_3, and the bromine was precipitated as AgBr. All of the activity separated with the precipitate. The arsenic was later precipitated with hydrogen sulphide, and was found to be inactive; this indicated that there were a negligible number of deuterons in the beam, for otherwise, the 26-hr. period of arsenic 76 would have been present. The experiments appeared to give a definite identification of bromine 78.

Since it might be expected that bromine 78 could also be made by the transmutation $\mathrm{Se}^{77}(D, n)\mathrm{Br}^{78}$ (reaction 2 of Fig. 10), it was possible to carry out a confirmatory experiment by activating selenium with deuterons for a short time. Fig. 3 gives the decay curve of a bromine separation from a sample of selenium metal which had been bombarded with deuterons for 5 minutes. Subtraction of the longer-lived activity (shown later to be due to bromine 83, simultaneously made from other selenium isotopes) showed that a period of 6.2 ± 0.2 minutes was indeed present, and the emission of positive electrons was confirmed by magnetic deflection. The strengths of the activities, corrected to infinite activation time, were 1.5 div. per sec. for the short period, and 3.2 div. per sec. for the longer period.

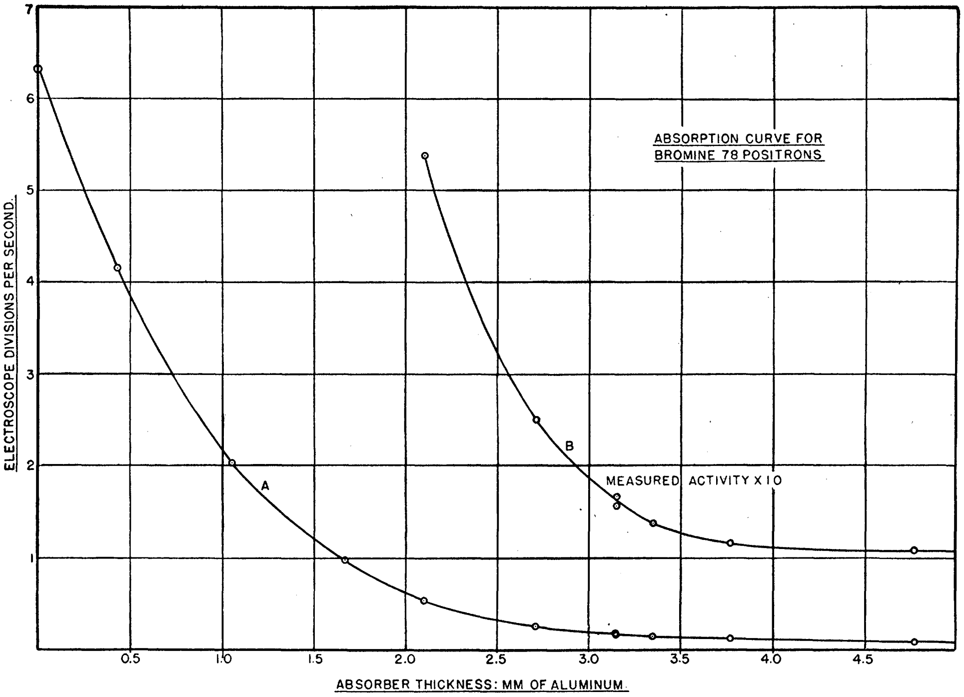

FIG. 2. Absorption curve for the positrons emitted from bromine 78. *A* is the complete curve, but in order to show the end-point clearly the flattening part of the curve has been replotted in *B*, using ten times the vertical scale of *A*. The end-point is seen to occur at an absorber thickness of about 4.1 mm of aluminum which shows that the positrons have a maximum energy of about 2.3 Mev.

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[10] N. Feather, Phys. Rev. **35**, 1559 (1930).

[11] Oxygen has given no detectable radioactive product when bombarded with α-particles in this laboratory.

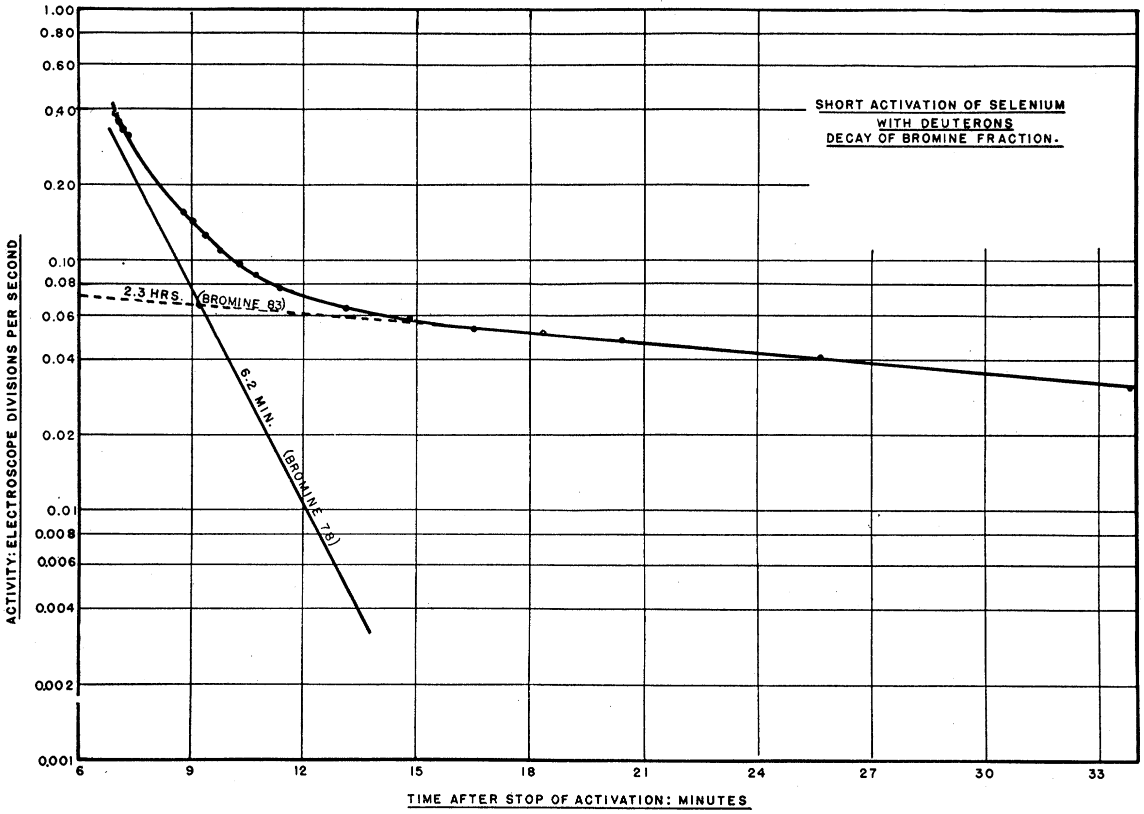

FIG. 3. Decay curve of a bromine separation from a sample of selenium which had been activated for 5 minutes with deuterons. The short periods only are shown. The 6.2-minute period belongs to bromine 78, and the 2.4-hour period to bromine 83.

During the course of this work notes from other laboratories[12, 13] have appeared, reporting the formation of bromine 78 by bombardment of bromine with energetic γ-rays (reaction $\mathrm{Br}^{79}(\gamma, n)\mathrm{Br}^{78}$), and with very fast neutrons (reaction $\mathrm{Br}^{79}(n, 2n)\mathrm{Br}^{78}$). In all of these reports the value of decay period is quoted as 5 minutes, and the sign of the emitted particles is not given. It seemed worth while to repeat the second of these experiments, to see if closer agreement could be obtained with the results of the arsenic and selenium activations just described. Accordingly, a sample of sodium bromide was activated for $3\frac{1}{2}$ minutes with neutrons from the (Li+D) reaction. The presence of a short-period activity was immediately apparent. The decay curve was followed into the 18-minute and 4.5-hour periods before the sample became too weak to measure; subtraction of these two activities then gave for the shortest period a value of 6.4 minutes. The presence of positive electrons with the short-lived activity was confirmed. The agreement of the results of all of these experiments upon bromine 78 is therefore very satisfactory. As indicated in Fig. 10, the substance has now been made in a total of four different ways, its identification is definite, and it has radioactive properties different from any of the three activities first formed by the action of neutrons upon bromine.

BROMINE 83

The decay curve of Fig. 3 has already been referred to; it was taken from a bromine separation from selenium activated for a short time with deuterons. In addition to the short period attributed to bromine 78, a period of about $2\frac{1}{2}$ hrs. is seen to be present. This bromine activity has been studied more closely, and

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[12] Heyn, Nature **139**, 842 (1937).

[13] Chang, Goldhaber and Sagane, Nature **139**, 962 (1937).

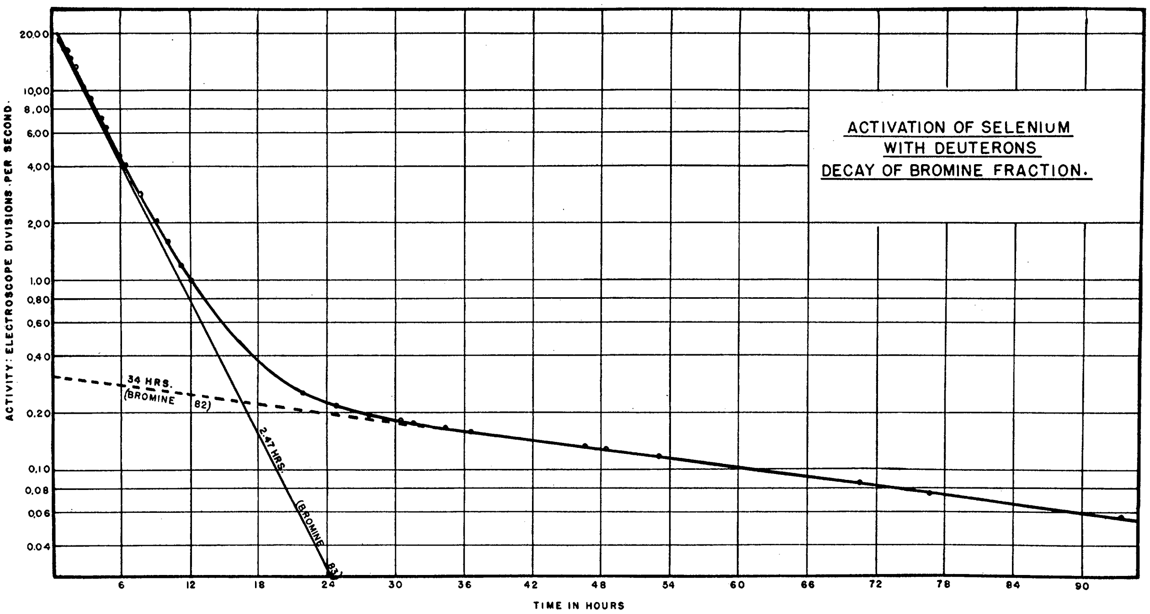

FIG. 4. Decay curve of bromine separation from selenium activated with deuterons; long periods. The 2.4-hr. period of bromine 83 shown in Fig. 3 reappears here, and is accompanied by the 34-hr. period of bromine 82.

Fig. 4 gives the decay curve of a bromine separation from selenium which had been given a much longer bombardment with deuterons (beam, 4μA; time of exposure, 1 hr.). The short bromine 78 activity had disappeared by the time this sample reached the electroscope, and therefore does not show in this decay curve. However, two other activities are present; their decay periods are 2.45±0.10 hrs. and 34 hrs., and their saturation strengths are, respectively, 92 and 15 divisions per sec. The 34-hr. period is one of the three original neutron-induced activities; its presence here has special significance which will be referred to later. The 2.45-hr. activity is new. It is accompanied by the emission of negative electrons, with no γ-rays.

Figure 5 gives absorption curves taken from one of these samples. In this figure, curve *A* includes the effects due to both the 2.45-hr. and the 34-hr. activities; the latter is partly responsible for the very rapid initial fall of the curve, because its β-particles are low in energy, and it also contributes the relatively weak γ-ray background. Curve *B* shows the absorption of the high energy β-particles in greater detail. To obtain this curve, the 34-hr. γ-ray effect was measured a day later, corrected for decay, and subtracted from the electroscope readings. The differences were then corrected for the decay of the sample while the absorption measurements were being taken, on the assumption of a half-life of 2.45 hrs. These differences were very small, and were multiplied by 100 for the purpose of plotting. As the 34-hr. β-rays are absorbed by less than a millimeter of aluminum (see Fig. 8), curve *B* applies to the β-rays of the 2.45-hr. activity alone. The limit of absorption occurs at about 2.1 mm of aluminum, and this corresponds to an upper energy limit of 1.3 Mev, according to Feather's rule.

The existing isotopes of selenium and bromine leave bromine 83 as the only possible carrier of this new activity. Since it is unlike any of the bromine activities so far discovered, it is not due to bromine 78, 80 or 82, and since it emits negative electrons it probably is not due to bromine 77. It is apparent that if this identification is correct, then there is a possibility that the substance was being made in two ways:

(a) By direct transmutation, according to the reaction Se^{82} (D, n) Br^{83};

(b) By growth from a hitherto unknown selenium 83; i.e., the bromine 83 would be the second member in a pair of successive β-emitters, the reactions being Se^{82} (D, p) Se^{83}, $\mathrm{Se}^{83} \rightarrow \mathrm{Br}^{83} + \epsilon^-$, $\mathrm{Br}^{83} \rightarrow \mathrm{Kr}^{83} + \epsilon^-$.

There was evidence for the second process in the decay curves of the selenium fractions obtained in the experiments, and it appeared that the selenium 83 had a half-life of 17±5 minutes. The two following experiments were then carried out, which established without doubt the presence of a pair of successive β-transformations.

1. Selenium was activated with deuterons, and selenium, bromine and arsenic were separated chemically. Decay readings were started on the bromine and arsenic fractions, and part of the selenium. The remainder of the selenium was thoroughly washed, and 3 hours later it was redissolved, and bromine was again extracted from it. This second extraction showed a weak activity, which decayed with the expected period of 2.4 hrs.

2. Selenium was activated for one hour with *slow neutrons*. There was therefore no possibility of a direct transmutation to bromine, and selenium 83 was made (along with some other active selenium isotopes) initially unaccompanied by bromine 83. The selenium was dissolved, and three fractions were taken from the solution, in the following order: (1) part of the selenium, immediately after activation; (2) the remainder of the selenium, half an hour later; (3) bromine, immediately after (2).

The decay curves of the three fractions are given in Fig. 6. The bromine was active, and showed the expected decay period. The selenium curves showed a mixture of periods, due mostly to other selenium isotopes. (Other experiments have shown that none of these has a period as long as 2.45 hrs. The strongest and longest-lived selenium activity induced by neutrons or deuterons has a half-life of 57 minutes.) The fact that the two curves are not parallel shows that the selenium samples were not identical in radioactive composition. The second sample had so much less of the long-lived constituent that its decay curve fell quickly and crossed that for the first sample. This is in agreement with the supposition that the 2.45-hr. activity was growing in the selenium samples after their separation, and was much stronger in the first sample

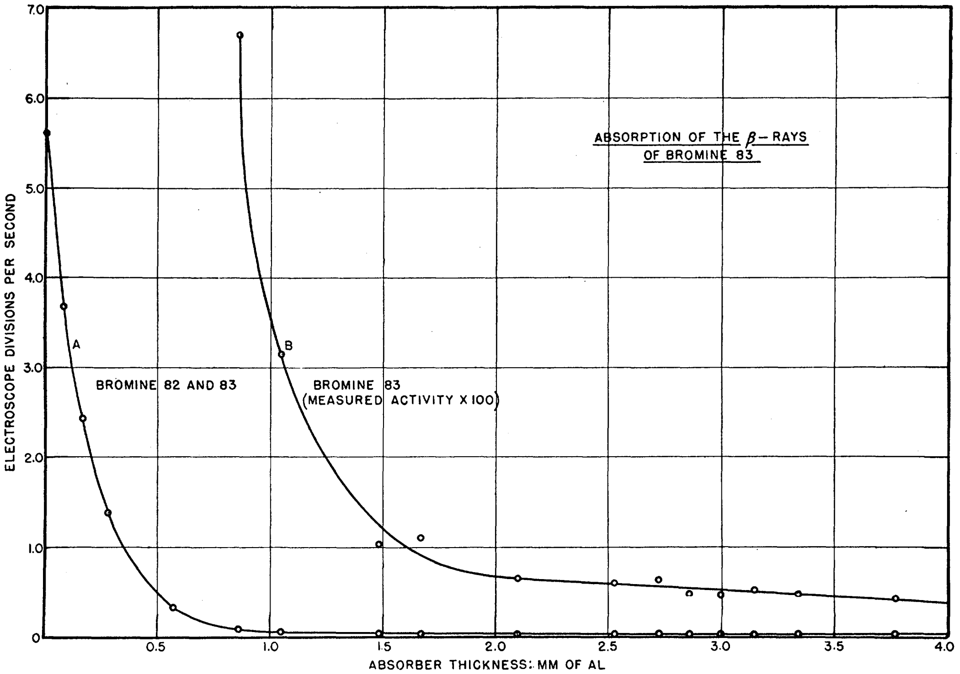

FIG. 5. Absorption curves for the β-rays of bromine 83. Curve A shows the absorption of the radiations from a mixture of bromine 83 (2.4 hrs.) and bromine 82 (34 hrs.); the γ-ray background is due to the latter activity. Curve B gives the absorption of the high-energy particles of bromine 83 alone, for the γ-ray effect has been subtracted, and the bromine 82 β-rays do not penetrate this far into the aluminum. Note that the true ordinates for curve B are 1/100 of the figures given at the left.

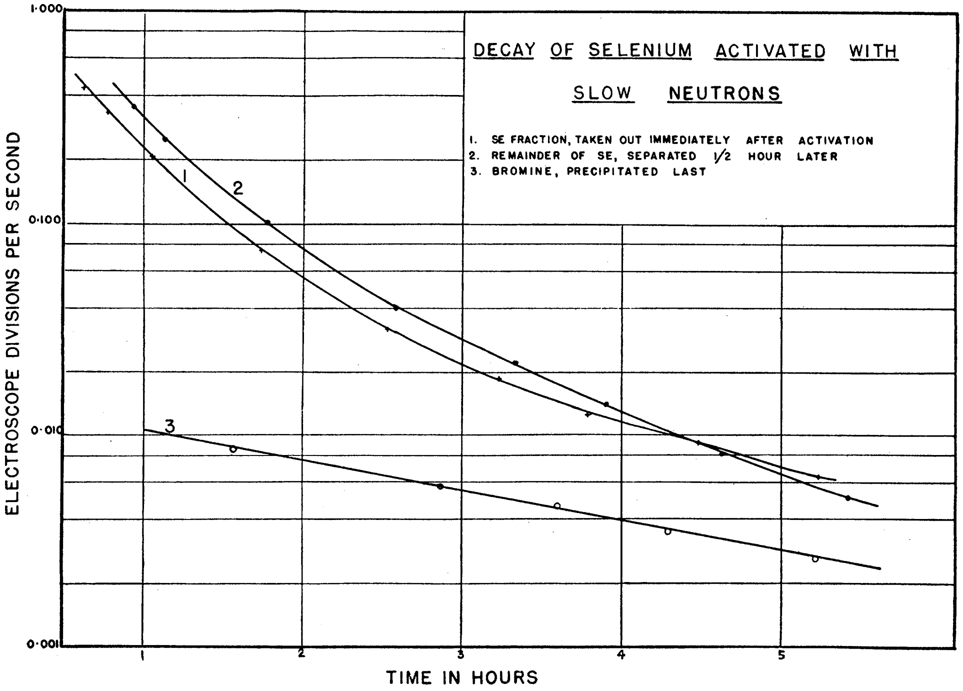

FIG. 6. These decay curves show that active bromine 83 (2.4 hrs.) results from the decay of active selenium 83 (17 min.). The selenium was activated with slow neutrons, and therefore the fact that the bromine fraction was active (curve 3) shows that a double β-transition must take place. The rate of decay shown by curve 3 corresponds to a half-life of 2.4 hrs. The selenium decay curves 1 and 2 slope off toward the 2.4-hr. bromine 83 period, because this substance has grown in the samples, and it lasts longer than any of the selenium active isotopes. The long period (bromine 83) is stronger in curve 1 than in curve 2, because the former sample, being removed first, had in it much more selenium 83 (17 min.) from which the bromine 83 subsequently grew.

because there was more selenium 83 present in the mother liquor when the first sample was taken out. The absolute activity of all of these samples was much less than that obtained by deuteron activation.

The establishment of these two successive β-transformations leaves no doubt as to the correctness of attributing the 2.45-hr. activity to bromine 83. Evidently this isotope is not connected in any way with the three original bromine activities, and if a third stable isotope is responsible for one of them, one can now say that it must be lighter than bromine 77 or heavier than bromine 83.

BROMINE 80 AND 82

A substantial contribution to the study of the bromine activities resulted from the activation of bromine with deuterons. The element was bombarded in the form of various bromides, and three activities resulted which decayed with the same half-lives as result from slow neutron activation. All three activities again followed the chemistry of bromine, and the absorption curves of the β-rays for each activity were the same as those obtained from neutron-activated samples. Therefore it seems quite definite that the same three activities are induced by deuterons as are induced by neutrons. The values of the decay periods, taken from a selection of the best decay curves obtained from both neutron- and deuteron-activated samples, are respectively 18.5 ±0.5 minutes, 4.54 ± 0.10 hours, and 33.9 ± 0.3 hours. Since the only known (or easily conceivable) reaction whereby bombardment of bromine with 5 Mev deuterons would give active bromine isotopes is $\mathrm{Br}^{79,\,81}(D,\,p)\mathrm{Br}^{80,\,82}$ (both of which would probably take place according to the process suggested by Oppenheimer and Phillips[14]) these results joined with the results

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[14] J. R. Oppenheimer and M. Phillips, Phys. Rev. **48**, 500 (1935).

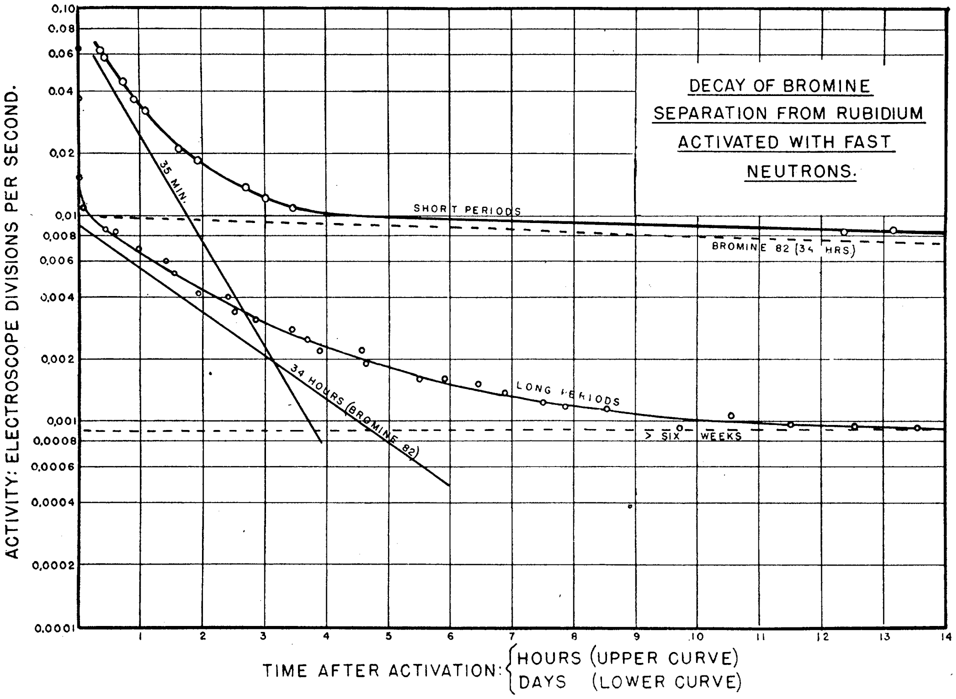

FIG. 7. Decay curve for bromine separation from rubidium nitrate activated for 4 hours with (Li+D) neutrons. The sample was weak, but three decay periods were present, *viz.* 35±3 min., 34±3 hrs. and a long period of more than 6 weeks. The second of these is one of the periods found in neutron and deuteron activated bromine, and its presence here identifies it as belonging to bromine 82 (reaction 12, Fig. 10). Whether or not one of the others belongs to bromine 84 is not yet certain, because of the possible presence of impurities.

of neutron activation in giving a very strong indication that all three activities must arise from the two isotopes bromine 80 and 82. The assumption of the existence of a very rare third stable isotope must now be finally abandoned, because the activities were all strong in the deuteron-activated samples; furthermore, tests with activation by neutrons of various energies have shown that none of the activities is a specifically slow neutron effect.

There remained the problem of finding how the three activities are distributed between the two active isotopes. In the last section it was mentioned that the 34-hour period was present in the bromine separation from selenium activated with deuterons, and the fact that it was unaccompanied by either the 18-minute or the 4.5-hour period might be taken as evidence that it alone belongs to one active isotope. More direct evidence was obtained, however, in two different ways, *viz.* by bombarding rubidium with fast neutrons, and by a study of the relative strengths of the three bromine activities when induced under varying conditions.

Since rubidium has two isotopes, with mass-numbers 85 and 87, it was expected that bombardment with fast neutrons might lead to the expulsion of an α-particle, giving bromine 82 and 84 according to the reactions $Rb^{85,\,87}(n,\,\alpha)Br^{82,\,84}$. (The first of these is reaction 12 in Fig. 10.) In order to test this possibility, a sample of about 25 grams of rubidium nitrate was given an intense bombardment with fast neutrons. After activation, the sample was dissolved in warm water, a drop of bromine water was added, and AgBr was precipitated from the solution by adding silver nitrate. Only an exceedingly weak activity resulted from bombardments of this kind when (Be+D) neutrons were used, but when the experiment was repeated using (Li+D) neutrons, the activity in the bromine fraction was strengthened sufficiently to enable the decay to be followed with some certainty. The decay curve (Fig. 7) shows a short period of 35±3

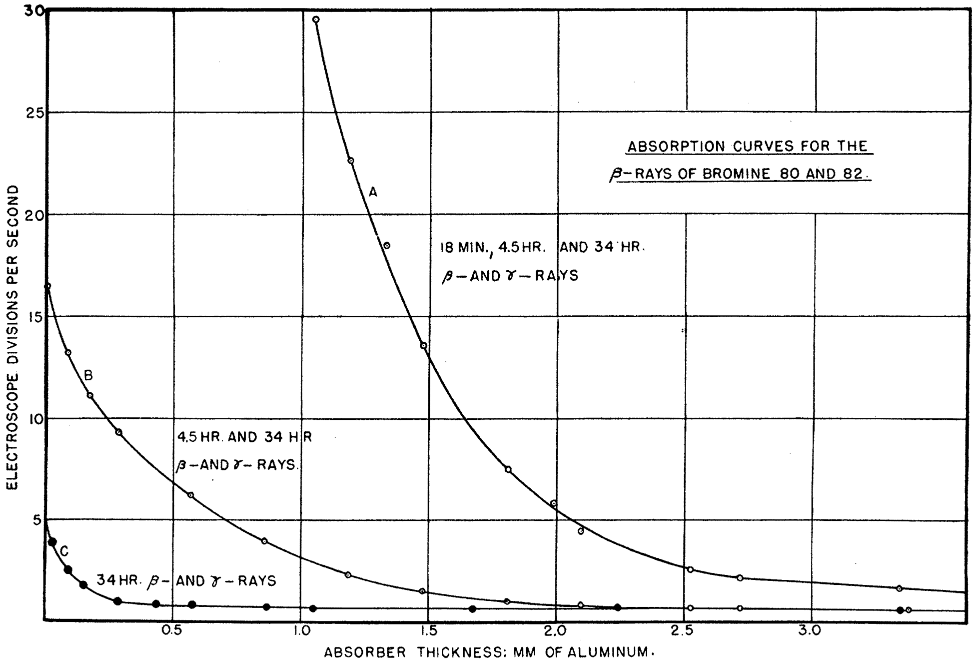

FIG. 8. Absorption curves showing the changes in the character of the radiations emitted by an aging deuteron activated bromine sample. Curve *A* was taken immediately after activation, when the 18-minute period accounted for most of the activity. Curve *B* was taken four hours later, when only the 4.5-hour and the 34-hour activities were present. Curve *C* was taken when only the 34-hour activity was left, and it has been corrected back to the time of curve *B*. It will be noticed that the 34-hour activity accounts for all of the γ-ray ionization present after the 18-minute activity had decayed.

minutes, an intermediate period of 34±3 hours, and a long-lived, very weak activity with a decay period of at least six weeks. For this discussion the presence of the 34±3 hour activity is very important; the other two will be considered later. Here the 34-hour bromine activity has apparently been again produced independently of the 18-minute and 4.5-hour activities, and the manner in which it was made shows that it belongs to bromine 82, and not to bromine 80. Although the sample was weak, a few points on the absorption curve of its β-rays were obtained while it was decaying with the 34-hour half-life, and the agreement with the absorption curve (Fig. 8, curve *C*) for the long period particles from activated bromine was excellent.

The study of the relative strengths of the three activities when induced in bromine by different methods lent support to the ascription of the 34-hour activity to bromine 82, and it also gave good evidence for attributing both the 18 minute and the 4.5-hour activities to bromine 80. In these experiments the active samples were dissolved after activation, and the bromine was precipitated as AgBr. The precipitate was then spread in a thin layer upon a piece of metal, and this type of source was used for the electroscope measurements. It was important to make the samples as uniform as possible; otherwise wide variations in the self-absorption of the β-rays in the source might occur, because of the great difference between the energy of the 34 hour β-particles and those emitted with the shorter periods. The conditions of activation of the samples and the relative strengths of the

TABLE I. *Conditions of activation and relative strengths of the three activities.*

Details of Activation			Relative Intensities		
Sample	Agent	Substance surrounding sample	18 min.	4.5 hr	34 hr.
1	(Be+D) neutrons	Paraffin	2.3	1	0.6
2	(Be+D) neutrons	Cadmium	2.7	1	0.8
3	(Li+D) neutrons	Boron	1.7	1	0.3
4	(Li+D) neutrons	KBr and cadmium	0.8	1	0.2
5	Deuterons	—	2.1	1	1.6

three activities are given in Table I. The intensities have been corrected to infinite activation time, and to facilitate comparisons the strength of the 4.5-hour period in each sample has been taken as unity.

The relative strengths of the 18 minute and 4.5-hour activities do not show wide variations except in the case of sample 4, which was surrounded by KBr. Here the 18-minute activity was weaker, probably because of the absorption in the KBr of the resonance neutrons known to exist for this activity,[15] some of which undoubtedly were present about the cyclotron. (Nothing is known about the existence of resonance levels for the other two periods; the results of sample 4 must therefore be treated with caution.) The variations in the intensities of the 34-hour activity are greater, and extend over a factor of eight. Deuterons (sample 5) emphasize this activity to a great extent, but no reason for this is as yet known. On the other hand the very fast neutrons from the (Li+D) reaction reduce its relative intensity by a factor of about 3 (samples 3 and 4) as compared to the (Be+D) neutrons (samples 1 and 2). This is significant, because neutrons having these high energies are known to be effective in expelling two neutrons from many nuclei (for bromine the reactions would be $\mathrm{Br}^{79,\,81}(n,\,2n)\mathrm{Br}^{78,\,80}$), and bromine 81 nuclei struck by such neutrons would increase the numbers of active bromine 80 nuclei at the expense of bromine 82. Even though unknown resonance effects may have influenced the results of sample 4, the relative intensities of sample 3 therefore point to the conclusion that both of the shorter-lived activities belong to bromine 80. In this respect the evidence of Bothe and Gentner[7] is conclusive in showing that neither of the shorter periods belongs to bromine 82, because they both appear in the photodisintegration of bromine according to the reactions $\mathrm{Br}^{79,\,81}(\gamma,\,n)\mathrm{Br}^{78\ \ 80}$; moreover, the remote possibility that one of them belongs to bromine 78 must be excluded for many reasons given in this paper.

An experiment was performed to test the effectiveness of group *C* (thermal-energy) neutrons in inducing the three activities in bromine. Two equal and similarly-shaped samples of lead bromide were exposed simultaneously to the action of neutrons by placing them side by side between two large paraffin blocks. One of the samples was wrapped in sheet cadmium, and the other was simply wrapped in paper. An exposure lasting half an hour induced activities with the following measured intensities, corrected to the time of stop of activation:

Period	*Cadmium-wrapped Sample*	*Sample without Cadmium*
18 min.	14.5 div./sec.	51 div./sec.
4.5 hrs.	0.62	2.4
34 hrs.	0.04	0.12

It is seen that the cadmium reduced the intensity of all three activities by a factor of about 3.5. It can be concluded that all three bromine activities can be induced by the action of neutrons having thermal energies.

The assumption of the existence of the two isomeric forms of bromine 80 is seen to explain all of these results and there seems to be no contradictory experimental evidence whatever. Until recently, the only definitely established example of nuclear isomerism has been the case of the natural radioactive substances UX_2 and UZ, both of which, according to Hahn,[16] must belong to the isotope of atomic number 91 and mass 234. With the current progress in the study of the induced radioactivities, however, there are already indications[17] that the phenomenon may be quite common. Weizsäcker[18] has recently given some theoretical considerations which indicate that it is possible to understand the occurrence of lifetimes of minutes or hours for low lying excited states in nuclei, without having to assume unreasonably large angular momentum differences from the ground level. Considerations of this kind are of course intimately connected with the energies of the β- and γ-rays emitted by the pair of nuclear isomers.

The β- and γ-rays of bromine 80 and 82

Although an accurate study of the radiations emitted by bromine 80 has not yet been made, absorption measurements made with the electroscope have given some information about their

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[15] von Halban and Preiswerk, J. de phys. **8**, 29 (1937); Goldsmith and Rasetti, Phys. Rev. **50**, 328 (1936).

[16] Hahn Zeits. f. physik. Chemie **103**, 461 (1922).

[17] Meitner, Hahn and Strassmann, Zeits. f. Physik **106**, 249 (1937), and McMillan, Kamen and Ruben, Phys. Rev. **52**, 375 (1937).

[18] Weizäcker, Naturwiss. **24**, 813 (1936).

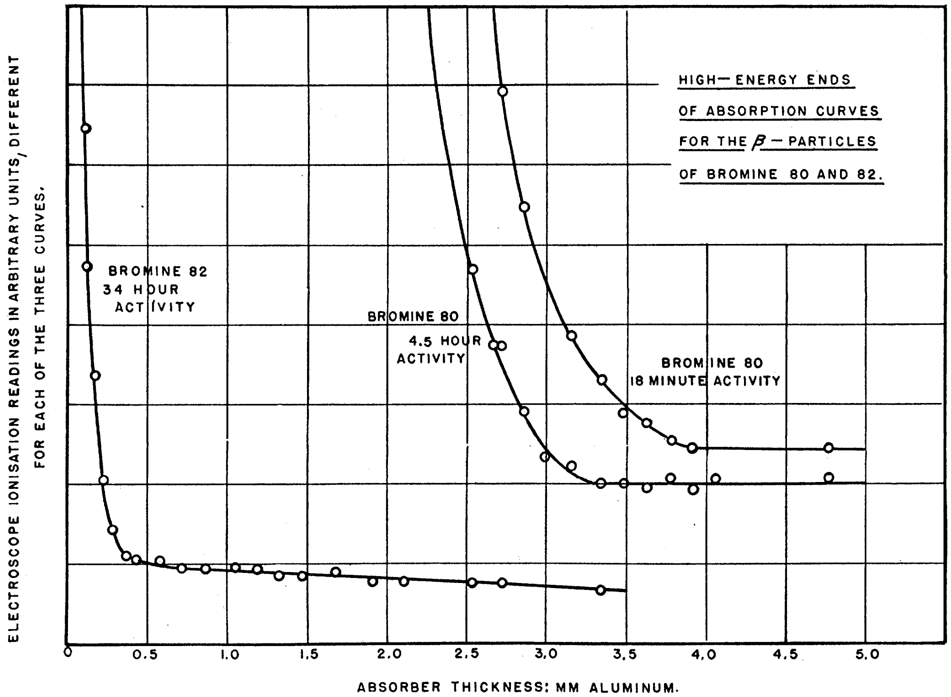

FIG. 9. The high energy ends of β-ray absorption curves taken from three different bromine samples, activated in such a way as to separate the 18-min., 4.5-hr. and 34-hr. activities. The curves are not to be compared with one another with respect to the scale of ordinates, and are intended only to show the positions of the end-points for the penetration of the β-rays. A slight difference is apparent in the energies of the β-rays emitted by the two isomeric forms of bromine 80.

general character which is worth reporting here. The 34-hour activity of bromine 82 was always present in the activated bromine samples, and it therefore came quite naturally under consideration at the same time.

The γ-ray decay curves of the neutron and deuteron activated samples were obtained with the electroscope by interposing sufficient absorber to stop all the β-rays. These decay curves showed a direct change from the 18-minute period to the 34-hour period, indicating that no appreciable γ-rays are emitted by the 4.5-hour activity. As measured in the electroscope, the γ-ray ionization arising from the 18-minute activity is weak, and that of the 34-hour activity is strong. Absorption measurements of the 18-minute γ-rays have shown that they are low in energy (undoubtedly less than 0.5 Mev) while the 34-hour γ-rays are more energetic. Kourchatow, Kourchatow, Myssousky and Roussinow[2] have given the value 0.65 Mev for the energy of the 34-hour γ-rays.

Negative electrons are emitted by the three activities of bromine 80 and 82. No positives have been observed. Absorption measurements of the β-rays, taken at different times during the decay of the sample, are reproduced in Fig. 8. These curves were taken from a sample of lead bromide which had been activated for 70 minutes with deuterons. The readings of curve *A* were obtained immediately after the stop of activation, and the curve shows the absorption of the β-rays (due predominantly to the 18-minute activity) together with a background which originates from a combination of the 18-minute and the 34-hour γ-rays. While the readings were being taken, the sample was decaying with a half-life of very nearly 18 minutes, and the corrections for decay were made on this basis. The activity for zero absorber was 110 divisions per second. Curve *B* was taken four hours later, after the 18-minute β- and γ-rays had disappeared, and accordingly shows a combination of the effects due to the 4.5-hour and the 34-hour periods. Curve *C* was taken 36 hours later still, and gives the absorption of the 34-hour radiations alone. The experimental points of curves *B* and *C* were both corrected to a time four hours after the stop of activation, assuming decay periods of 4.5 hours and 34 hours, respectively; the two curves can therefore be compared directly without further allowance for decay.

It is seen that nearly all of the β-ray ionization of curve *B* is due to the 4.5-hour activity, but the γ-ray background arises entirely from the 34-hour period. In curve *C* the softness of the 34-hour β-rays is quite apparent, and this accounts partly for the fact that the β-ray ionization is unusually weak in comparison with that due to γ-rays, for the sample used in these measurements was quite thick. To obtain a better comparison of the relative strengths of the β- and γ-ray ionization, an active silver bromide precipitate was on one occasion spread thinly upon a piece of metal, and an absorption curve for the 34-hour activity was taken from this source. The ratio of the $(\beta+\gamma)$-ray ionization (i.e., the ionization measured with zero absorber) to that arising from the γ-rays alone was found with this sample to be 10. This is still small compared to the values obtained for many other radioactive isotopes, (values of about 50 for this ratio are more usual) and it may indicate that bromine 82 emits more than one γ-quantum per β-particle.

The high energy ends of the β-ray absorption curves for the three activities were examined more closely, in order to learn more about the maximum energies of the respective groups of β-rays. For this purpose it was necessary to isolate each of the three activities fairly completely from the others. This was accomplished by studying the 18-minute activity from samples which had been given a short slow neutron activation (lasting about 3 minutes), the 4.5-hour activity from longer slow neutron activations, and the 34-hour activity from either neutron or deuteron activated samples. Deuteron activation was not used for studying the shorter lived activities, because it increases the relative strength of the long period activity to an inconvenient extent. The high energy ends of three such absorption curves are reproduced in Fig. 9. They have been plotted to an enlarged vertical scale for the purpose of showing the end-points clearly. Since these curves were taken from three different samples, they are not to be compared with one another except with regard to the positions of the end-points of transmission of the β-rays. These end-points are given in Table II, together with the corresponding maximum energy of the β-rays as obtained from Feather's formula.

For purposes of comparison the values obtained by Alichanow, Alichanian and Dzelepow[19] with the magnetic spectrograph have been added, and also the value obtained by Kourchatow, Kourchatow, Myssowsky and Roussinow[2] for the maximum energy of the bromine 82 β-rays. The agreement in the figures is moderately good, but fails in what is perhaps the most interesting detail; *viz.* the maximum energies of the two groups of bromine 80 β-rays. The absorption measurements indicate that the 18-minute β-rays are slightly more energetic than the 4.5-hour β-rays and the clearness of this result may be judged from the curves of Fig. 9. However, Alichanow, Alichanian and Dzelepow failed to find this difference. Further measurements of both the β-rays and the γ-rays of the bromine activities are needed, and it is hoped that they soon will be obtained. As far as the present information goes, however, it is in agreement with the picture of a low lying metastable state in bromine 80, from which the nuclei decay with a half-life of 18 minutes by the emission either of a β-particle or a low energy γ-quantum, together with a ground level from which decay

TABLE II. *End-points of β-ray transmission from radioactive bromine together with the corresponding maximum energy as obtained from Feather's formula.*

ACTIVE ISOTOPE	DECAY PERIOD	END-POINT OF TRANSMISSION OF β-RAYS	MAXIMUM ENERGY OF β-RAYS	ALICHANOW, ETC.	KOURCHATOW, ETC.
Bromine 80	18 min.	3.9 mm of Al	2.2 Mev	2.00±0.10 Mev	—
"	4.5 hrs.	3.4 " " "	2.0	2.05±0.10	—
Bromine 82	34 hrs.	0.5 " " "	0.4	0.85±0.20	0.6 Mev

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[19] Alichanow, Alichanian and Dzelepow, Physik. Zeits. Sowjetunion **10**, 78 (1936).

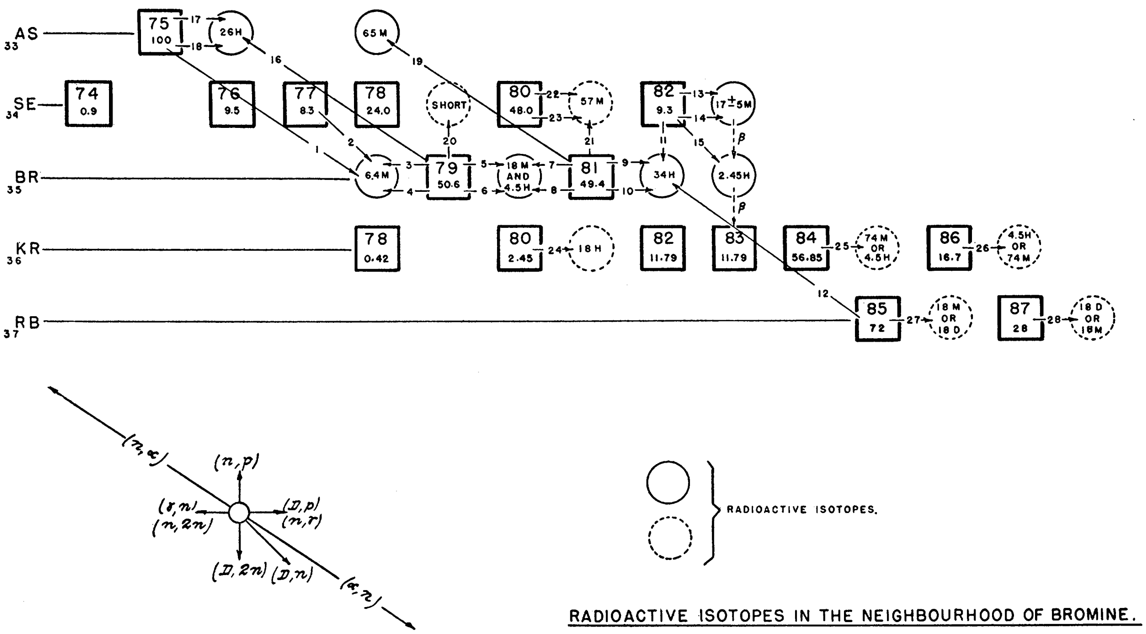

FIG. 10. This diagram gives a summary of all of the transmutations described in this paper. For convenience in reading, a square array has been chosen in which atomic number is plotted against atomic mass. The stable isotopes (squares) are given with their percentage abundance in small figures. Radioactive isotopes are given in circles, which are dotted when there is still some ambiguity as to their exact identification. The numbered arrows indicate transmutations which have been observed, and the reactions describing them are as follows:

1. As^{75}(α, n) Br^{78}
2. Se^{77}(D, n) Br^{78}
3. Br^{79}(n, $2n$) Br^{78}
4. Br^{79}(γ, n) Br^{78} (Bothe and Gentner)
5. Br^{79}(n, γ) Br^{80}
6. Br^{79}(D, p) Br^{80}
7. Br^{81}(n, $2n$) Br^{80}
8. Br^{81}(γ, n) Br^{80} (Bothe and Gentner)
9. Br^{81}(n, γ) Br^{82}
10. Br^{81}(D, p) Br^{82}
11. Se^{82}(D, $2n$) Br^{82}
12. Rb^{85}(n, α) Br^{82}
13. Se^{82}(n, γ) Se^{83}
14. Se^{82}(D, p) Se^{83}
15. Se^{82}(D, n) Br^{83}
16. Br^{79}(n, α) As^{76}
17. As^{75}(n, γ) As^{76}
18. As^{75}(D, p) As^{76} (Thornton)
19. Br^{81}(n, α)As^{78}
20. Br^{79}(n, p) Se^{79}
21. Br^{81}(n, p) Se^{81}
22. Se$^{78, 80}$(n, γ) Se$^{79, 81}$
23. Se$^{78, 80}$(D, p) Se$^{79, 81}$
24. Kr^{80}(D, p) Kr^{81}
25. Kr^{84}(D, p) Kr^{85}
26. Kr^{86}(D, p) Kr^{87}
27. Rb^{85}(n, γ) Rb^{86}
28. Rb^{87}(n, γ) Rb^{88}

by β-emission takes place with a half-life of 4.5 hours. The product nucleus in each case is krypton 80, and presumably it is formed in its ground state.[20]

BROMINE 75, 77 AND 84

It might be expected that deuteron bombardment of selenium would lead to the formation of bromine 75 and 77 from selenium 74 and 76, by the (D, n) type of reaction. However, neither of these isotopes has been found. That bromine 75 was not observed probably was due to the rareness of selenium 74. The absence from the decay curves of any activity which might be attributed to bromine 77 indicates that either (1) its half-life is less than a minute or greater than roughly two months (a very long lived activity has not been looked for); or (2) selenium 76 is not so susceptible to the (D, n) transmutation as are some of the other selenium isotopes.

The bombardment of rubidium with fast neutrons should yield bromine 84 in addition to bromine 82, according to the reactions Rb$^{85, 87}$ (n, α)Br$^{82, 84}$. As a matter of fact, when this experiment was tried the decay curve (Fig. 7) of the bromine separation did show activities other than that of bromine 82, but it is not yet certain whether either of them belongs to bromine 84. A 35±2-minute activity is present, but it might be the 37-minute activity due to

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[20] Alternatively, one may account for the experimental results by supposing that the 18-min. β-rays originate in a transition from a metastable state in bromine 80 to an excited state in krypton 80, which then emits the 18-min. γ-ray in going to the ground level. The 4.5-hr. β-rays would correspond to a transition from normal bromine 80 to the ground level of krypton 80, and the energy of excitation of the metastable state of bromine 80 would be given by the sum of the energy of the 18 min. γ-ray and the 0.2 Mev difference in energy of the two β-rays.

chlorine 36 or 38, made from possible potassium impurity in the rubidium by the reactions $K^{39,41}(n, \alpha)Cl^{36,38}$—the chlorine then separating chemically with the bromine. A long period also is present, but there is a possibility here of confusion with the 18-day rubidium activity, since it is conceivable that the chemical separation was not complete.* Larger samples of rubidium and stronger neutron activations will greatly facilitate a closer study of these possibilities.

FORMATION OF BROMINE 82 FROM SELENIUM

As has already been mentioned, a 34±1-hr. activity was present in the bromine separation from selenium activated with deuterons. It occurred in addition to the shorter lived activities of bromine 78 and 82, and it could be made sufficiently strong to follow for more than a week. Its β-ray absorption curve was obtained and found to be identical with that of the bromine 82 34-hr. period as given in Fig. 8. The γ-rays also were present. Apparently, therefore, bromine 82 can be made by deuteron bombardment of selenium.

Reference to the scheme of stable isotopes (Fig. 10) shows, however, that none of the reactions heretofore encountered in nuclear transmutations can account for a transmutation yielding this product. A reaction of the common type "deuteron in, neutron out" will indeed lead from selenium to bromine (as probably occurs in the case of bromine 78 and 83) but such a reaction is of no assistance here because a stable selenium isotope of mass 81 would be required, and this isotope does not exist in sufficient abundance to be detected by the mass-spectrograph. Radiative capture of the deuteron according to the reaction $Se^{80}(D, \gamma)Br^{82}$ would appear to be a possible solution according to a naïve inspection of the isotope scheme, but for theoretical reasons such a process must be regarded as very improbable. The bromine 82 nucleus would be formed with an excitation energy about twice as great as the individual binding energies of the constituent protons and neutrons, and this is regarded as an excessive amount of energy to be thrown off as γ-radiation. A more satisfying explanation, first suggested to the writer by Professor Bohr, lies in the supposition that selenium 82 captures the deuteron and emits two neutrons (reaction $Se^{82}(D, 2n)Br^{82}$, shown as number 11 in Fig. 10). No other reaction of this type has so far been established, but the comparative ease with which neutrons can be expelled from the nucleus has been demonstrated by current experiments upon disintegration with γ-rays and neutrons of high energy, and it is in good accordance with the Bohr picture of nuclear transmutation. The fact that the apparent reaction $Se^{82}(D, 2n)Br^{82}$ is observed to take place unaccompanied by the parallel reaction $Se^{80}(D, 2n)Br^{80}$ (which presumably would yield the 18-min. and 4.5-hr. bromine activities) may be understood from the lower binding energy of the neutrons in the heavier isotopes, and the consequent greater facility with which the extra neutron could escape.

OTHER TRANSMUTATIONS

Although primarily intended to be a study of the radioactive isotopes of bromine, this work has inevitably spread to the neighboring elements arsenic, selenium, krypton and rubidium. Some new activities have been discovered in these elements, and they are graphically summarized in Fig. 10 together with the transmutations by which they have been produced. Only a few of these new isotopes have yet been studied closely, but it might be well to describe briefly how they were found. Except for bromine 78 (and possibly one krypton isotope) all of these activities are accompanied by the emission of negative electrons.

Reactions 1 to 15 of Fig. 10 have already been described in the paper.

Reactions 17 and 18 are taken from the original work of Fermi[1] and Thornton.[21]

Reactions 16, 19, 20 and 21 resulted from an activation of a large sample of ammonium bromide with (Be+D) neutrons. After a chemical separation, the arsenic fraction showed activities having decay periods of 26±1 hours and 65±3 minutes, with intensities of 0.53 and

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* *Noted added October 9:* Further aging of the samples has shown that the decay period is greater than six weeks, so that this possibility must be ruled out.

[21] R. L. Thornton, Phys. Rev. **49**, 207 (1936).

0.05 divisions per sec. respectively. (All intensities of activities given here have been corrected to infinite time of activation.) The first of these is the familiar period of arsenic 76, and it probably originates from bromine 79, being made by α-particle expulsion according to reaction 16. The second activity is new, and it has been attributed to arsenic 78, presumably made by a parallel reaction (number 19) from the other bromine isotope. It has not been cross checked by an independent transmutation. The selenium fraction showed a clear-cut 57 ± 1 minute activity (intensity 1.3 divisions per sec.) with no evidence of a longer period; there was, however, a slight indication of a shorter period of less than 5 minutes having an intensity of 0.1 divisions per sec. or less. These two active isotopes probably are made from the two stable bromine isotopes by the (n, p) reactions 20 and 21. The 57-minute period has been observed by Heyn,[12] and has been found by the writer in samples which have been activated with deuterons and slow neutrons (reactions 22 and 23). It is not yet certain whether it belongs to selenium 79 or to selenium 81.

Reactions 24, 25 and 26 resulted from activations of krypton gas with deuterons. Activities with decay periods of 74 ± 2 minutes (intensity 3.6 divisions per sec.) 4.5 ± 0.1 hours (2.1 divisions per sec.) and 18 ± 2 hours (0.018 divisions per sec.) were observed, and they can be considered to be identified as belonging to krypton isotopes, because any atomic rubidium or bromine formed in the bombardment would have stuck to the copper walls of the container or to the inside of the slender copper tubing used in handling the gas. The weakness of the 18-hour activity has so far prevented an observation of the charge of the emitted particles, and it may indicate that the activity belongs to krypton 79 or 81, which would be made from one of the rarer isotopes 78 or 80. This supposition is supported by the observation that the strong 74-minute and 4.5-hour activities emit negatives, which shows that they probably belong to krypton 85 and 87. The appearance of a decay period of 4.5 hours in krypton as well as in bromine appears to be fortuitous, because an absorption curve of the krypton 4.5-hour β-rays has shown that they are much lower in energy than the bromine 4.5-hour β-rays.

Reactions 27 and 28 refer to the activation of rubidium nitrate by neutrons. The two activities with 18 ± 2 minute and 18 ± 1 day half-lives appeared in the chemically separated rubidium fraction; they probably belong to rubidium 86 and 88, but lack of cross checks has as yet prevented any closer identification. The strengths of the activities in the small portion of the sample which was used for the electroscope measurements were: for the 18-minute activity, 0.77 divisions per sec., and for the 18-day activity, 4.2 divisions per sec. A weak rubidium activity with a period of "about 100 minutes" has been mentioned in the literature;[22] probably it is the 18-minute period found here with stronger samples. Experimentally, there is no possibility of confusion of this 18-minute activity with the 18-minute activity of bromine 80.

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[22] Pontecorvo, Nuovo Cimento **12**, 223 (1935).