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Measurement of Neutron Absorption Cross Sections with a Pile Oscillator*

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The following paper deals with the measurement of thermal neutron cross sections by a technique referred to as the pile oscillator. In this method, a neutron absorber is moved back and forth in a field of thermal neutrons such as that existing in a chain reactor. In the vicinity of the absorber there is a depression in the neutron flux and the motion of this depression past a nearby ion chamber produces an oscillating signal whose amplitude is proportional to the total neutron absorption cross section of the sample. In the instrument considered here, the oscillating signal generated in the ion chamber is amplified, rectified, and fed to an integrating circuit. The absorption cross section of an unknown sample is determined by comparing the charge accumulated in the integrating circuit with that produced by a standard gold absorber. The instrument will detect a total absorption cross sestion of about 10^{-3} cm^2 and hence it is suitable for measuring cross sections of separated isotopes which are available only in small quantities. One of the major difficulties in making this instrument useful for absorption measurements has been the elimination of the effects of scattering by the sample. It was found that the signals caused by absorption and scattering are slightly out of phase with each other, and by properly phasing the rectifier, absorption cross sections which are no greater than 1 percent of the corresponding scattering cross sections can be measured. A number of thermal cross section measurements have been made with this instrument, most of which will be published separately; as illustrations, the following thermal cross sections are given: In, 191.2*b*; Ag^{107}, 29.9*b*; Ag^{109}, 83.7*b*.

INTRODUCTION

WHEN a neutron absorber is placed in a chain-reacting pile it produces a change in reactivity which is proportional to the total absorption cross section of the absorbing sample. This fact has been used in the so-called "danger coefficient" method[1] of measuring absorption cross sections in which the change in reactivity produced by a sample is balanced by the change in position of a calibrated control rod. Because of the inherent fluctuations of neutron intensity in a chain reactor, this static danger coefficient method requires rather large amounts of absorber for accurate cross-section measurements. It is therefore not adequate for the measurement of capture cross sections of rare substances such as separated isotopes. In this paper a high sensitivity "pile oscillator" method which has been developed for the measurement of cross sections of small samples will be described. This method is now being used at the Oak Ridge National Laboratory to make a systematic survey of the absorption cross sections of separated isotopes.

The pile oscillator method,[2] as proposed originally by E. P. Wigner, involves oscillating the unknown absorber in and out of the reactor or back and forth within the pile from a region of low neutron flux to a region of high flux. The reactivity of the chain reactor undergoes oscillations about criticality with the same frequency as the oscillations of the absorber, the reactivity being reduced when the absorber is in the pile. This reactivity oscillation produces an oscillation in the over-all neutron flux which then gives rise to a corresponding signal in a neutron-sensitive ion chamber located in the pile. Since the amplitude of the flux oscillation is proportional to the absorption of the sample, the instrument constitutes a means of determining unknown neutron-absorption cross sections by making comparisons with substances for which accurate direct measurements have been made. The gain in sensitivity of the oscillator method over the static danger coefficient method accrues both because it averages effects over so many cycles that random fluctuations tend to cancel, and because it makes a.c. amplification possible.

The original pile oscillator depended for its working on the change in pile reactivity caused by the motion of the absorber. The recording ion

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[1] H. L. Anderson, E. Fermi, A. Wattenberg, G. Weil, and W. Zinn, Phys. Rev. **72**, 16 (1947).

[2] The theory of the pile oscillator is given in A. M. Weinberg and H. C. Schweinler, Phys. Rev. **74**, 851 (1948).

chamber in this method is best placed at a large distance from the absorber so that local neutron density changes near the absorber are not picked up. However, it was pointed out in a paper on the theory of the pile oscillator by A. Cahn, A. T. Monk, and A. M. Weinberg,[3] that a significant increase in sensitivity could be achieved by placing the ion chamber very close to the oscillating sample. By this means not only the over-all fluctuation in pile intensity, but also *the local depression in neutron flux which exists in the neighborhood of an absorber is measured.* Thus when the local neutron depression is measured, the pile is used primarily as a neutron flux field; the fact that the reactivity of the chain reactor also depends on the position of an absorber within it is only incidental. It is therefore possible to make measurements wherever there is a strong neutron flux, irrespective of how the reactivity of the pile is affected by the absorber. For example, to measure thermal cross sections it is advantageous to oscillate the sample in the graphite reflector of the pile where there are very few epithermal neutrons, even though the reactivity is little affected by motion of an absorber in the reflector.

While the measurement of the local depression increases the signal to noise ratio and makes it possible to measure thermal neutron cross sections unperturbed by epithermal neutrons, it has a serious disadvantage, namely, that the details of the spatial distribution of the neutron density around an absorber depend on its scattering cross section as well as its absorption cross section. It is therefore necessary to arrange the position of the ion chamber with respect to the absorber so as to reduce the scattering effect as much as possible. The extent to which the scattering signal has been eliminated is discussed below.

The Oak Ridge[4] pile oscillator has undergone a number of changes during its development; it will be described here in its present state in which the absorber is oscillated in close proximity to the ionization chamber and the local depression signal is recorded.

APPARATUS

The pile oscillator consists essentially of a mechanical device to oscillate the sample, a neutron-sensitive ionization chamber and an electrical system to amplify and convert the voltage pulse generated within the chamber to a quantity which can be readily measured.

The mechanical system for oscillating the sample is shown in Fig. 1. It consists of a washing machine gear box, on the drive shaft of which is mounted an aluminum wheel 30 in. in diameter. A braided steel cable passes over this wheel, over two idler pulleys and over another pulley inside the pile. On the lower cable, which passes into the pile through a $\frac{1}{2}$ in. square channel in a graphite stringer, there is fastened a rectangular beryllium boat with inside dimensions $\frac{3}{8}$-in. $\times\frac{3}{8}$-in. $\times 2\frac{1}{2}$-in. This boat carries the samples to be measured. The to and fro motion of the wheel produces an oscillation of the boat along the graphite channel with a total travel of 32 in. The large drive wheel can be disengaged from the drive shaft so that the boat can be reeled out of the pile for loading and then reeled back into position. The frequency of oscillation which has been used is about 1 cycle per second.

Two types and locations of ionization chambers have been used. One which shall be referred to as A was an aluminum chamber, 10 in. long, 1 in. in diameter, coated internally with about

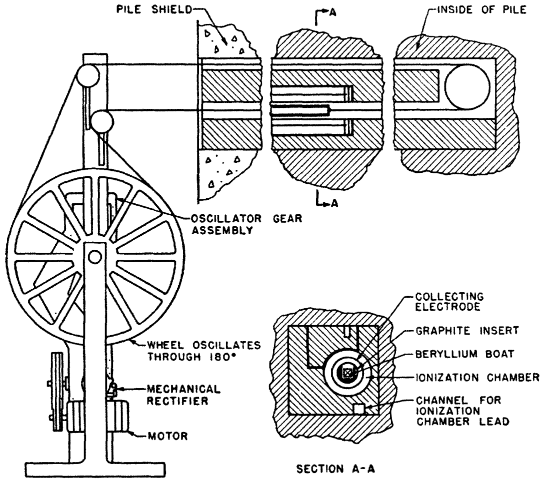

FIG. 1. Pile oscillator mechanical assembly. The portion of the drawing showing the graphite channel and ionization chamber has been enlarged to show details.

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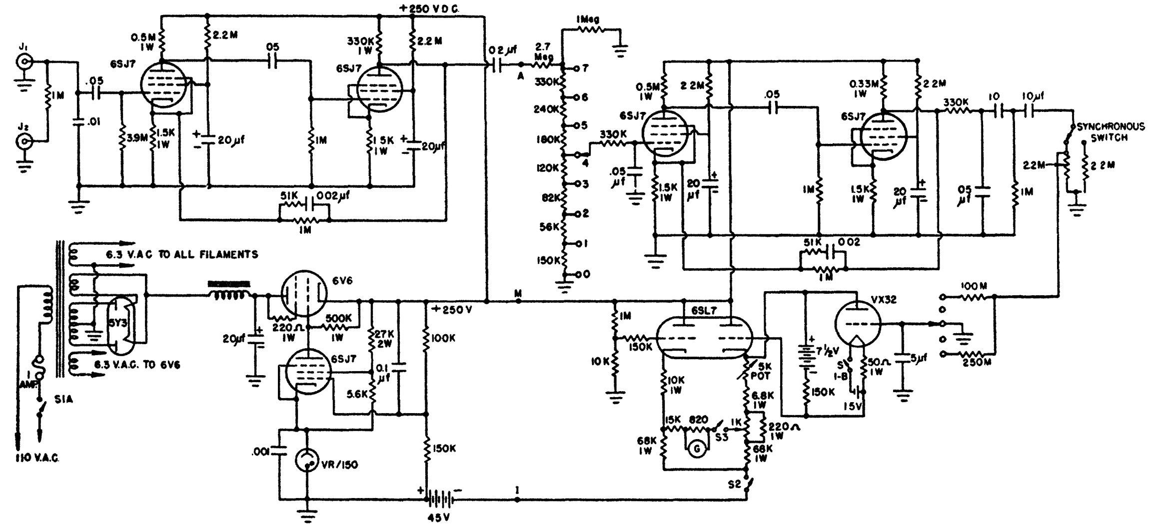
[3] Manhattan Project Report CP-G-2907.

[4] An oscillator depending on the reactivity change effect has been developed by A. Langsdorf at Argonne National Laboratory, a preliminary report of which was presented at the Washington Meeting April 29, 1948.



ALL RESISTORS ½ WATT UNLESS SPECIFIED

FIG. 2. Amplifier and integrator circuit. The ionization chamber is connected to *J*1 and a 500-volt battery is connected to *J*2.

2 mg/cm^2 of enriched boron. This chamber was buried in the graphite stringer with its axis parallel to and its center line 1 in. below the axis of the boat channel. A second annular chamber, which will be referred to as *B*, has been found more satisfactory, and this is the chamber that is now being used. This chamber, shown in Fig. 1, is placed coaxially to the path of the oscillating sample, and its center coincides with the mid-point of the absorber's path of oscillation. The active volume is contained between two concentric aluminum tubes 11 in. long, the outer tube being 2 in. o.d. and the inner tube $\frac{7}{8}$ in. o.d. The collecting electrode, which is insulated from the rest of the chamber by polystyrene, is 10 in. long and $1\frac{1}{2}$ in. o.d. It is coated on both sides with 0.5 mg/cm^2 of boron enriched in B^{10}. A graphite insert machined to the dimensions of the boat channel extends through the inner tube which protrudes 8 in. on either side of the ionization chamber. Graphite was used for the insert to reduce scattering effects which might arise from the streaming of neutrons through the boat channel. The protruding sections of the tube are coated with about 2 mgs/cm^2 of enriched boron in order to reduce abrupt changes in neutron density at the boundary of the active section of the chamber.

The voltage pulses from the ionization chamber have been measured by both a resonance galvanometer and by an integrating circuit method. The latter method, which is more convenient, is now being used. The signal from the ionization chamber is first amplified by a fairly narrow band feed-back amplifier, then rectified by a synchronous switch and integrated by means of a long time-constant R-C circuit. (See Fig. 2.) The voltage appearing across the integrating condenser is measured by means of a vacuum tube voltmeter. The use of the synchronous rectifier and integrator discriminates strongly against frequencies other than the desired signal so that noise from the chamber, because of statistical fluctuations in the ion current, is greatly reduced.

The rectifying switch is operated by a cam connected to the gear box of the mechanical driver and provision is made for selecting any desired portion of the cycle. This is of importance in connection with reducing the effects of scattering, as discussed in the next paragraph.

THE EFFECT OF SCATTERING BY THE SAMPLE

The voltage pulse from the ionization chamber is primarily caused by the absorption of neutrons in the sample. It is found, however, that by oscillating a piece of graphite which has negligible absorption, a signal may also be generated by the scattering of the sample. To make the oscillator a satisfactory instrument for absorption cross-section measurements, the effects of scattering must be largely eliminated. Toward this end the voltage pulses produced by absorbers and scatterers have been studied for both the *A* and *B* types of chambers. With the *A*-type chamber, which was located below the oscillating sample, the absorption and scattering signals are so similar that their effects cannot be separated. With the *B*-type chamber, which is coaxial with the oscillating sample, the signal from a nearly pure scatterer is considerably different from that resulting from a sample with a large absorption cross section. This can be seen from the patterns shown in Fig. 3 which were traced from an oscilloscope with an approximately linear sweep on the horizontal plates. The trace started at the left each time the oscillating sample reached its outermost position. Thus two traverses through the chamber appear on each sweep, the first as the sample moves into the pile, the other as it moves out. One of the vertical deflection plates of the oscilloscope was connected to the last stage of the amplifier, ahead of the rectifier and integrator.

Figures 3c and 3d were taken with the amplifier normally used with the equipment, Figs. 3a and 3b with a specially constructed amplifier having extended low and high frequency responses. Traces *b* and *d* are reproductions of the signal shape with graphite in the boat; traces *a* and *c* were obtained by putting cadmium in the boat and reducing the amplifier gain by a factor of 4.

One observes that the scattering signal is opposite in sign, has a steeper rise, and reaches its maximum at an earlier time than the absorbing signal. This can be explained if one assumes that the scattering signal arises chiefly from neutrons which are scattered out of a beam of neutrons streaming down the channel. Neutrons that otherwise would have passed through the central hole of the chamber without being detected are deflected into the chamber by the sample which blocks the channel. This is substantiated by the results obtained when graphite rods are attached to either side of the boat and oscillate with it. In this instance one still obtains a scattering signal caused by the hole in the boat, but, since the channeling of the neutrons is largely eliminated, the amplitude of the signal is much reduced.

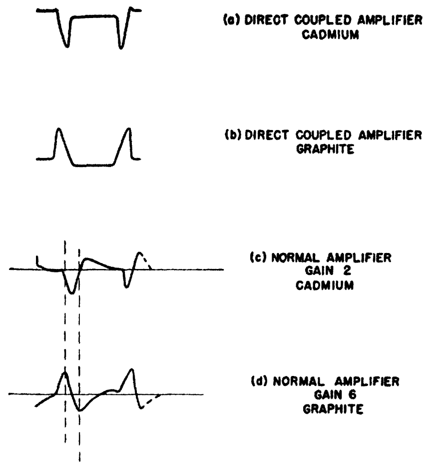


FIG. 3. Wave forms of voltage from amplifier for one complete stroke of sample past the ionization chamber. a and c correspond to pure absorption (cadmium) and b and d to pure scattering (graphite).

The low frequency response of the amplifier normally used with the pile oscillator has a considerably higher low frequency cut-off than the special amplifier previously referred to, so that transients and low frequency disturbances are very much reduced in the actual measurements. This causes considerable distortion of the signal as shown in (c) and (d). By phasing the rectifying switch so that signal integration occurs only during the interval indicated between the two vertical lines, the galvanometer deflection because of a scatterer can be made very small. With an

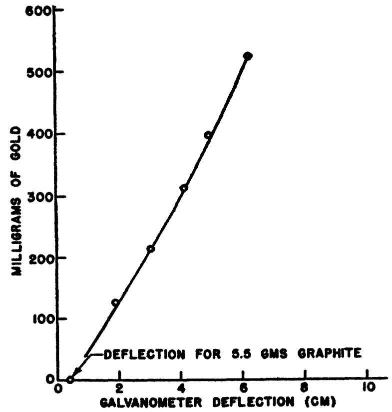


FIG. 4. Galvanometer deflection *versus* amount of gold.

TABLE I. Thermal neutron absorption cross sections of indium and silver.

Sample	σ (barns) Run 1	Run 2	Run 2	Av.	Absorption cross section of sample in mm^2
In (153 mg)	189.5	192.7	191.5	191.2	15.3
In (5.0 mg)	186	197	224	202	0.5
Ag^{107} (444 mg. 90%)	29.0	30.8		29.9	9.7
Ag^{109} (373 mg. 92%)	83.9	83.5		83.7	16.6
$0.52\sigma^{107}+0.48\sigma^{109}$	55.6	56.2		55.9	
Natural Ag (357 mg)	59.2	60.5		59.8	11.6

absorbing sample, integration over this same time interval produces a large deflection. It is by this means that we have been able to measure small absorption cross sections without correcting for the scattering by the sample. Evidence for this can be seen from Fig. 4, where the boat was loaded with 5.5 g of carbon corresponding to 132 mm^2 of scattering cross section and 0.13 mm^2 of absorption cross section. With the *A* type chamber, 20 mm^2 of scattering cross section were required to give a signal equal to that produced by 1 mm^2 of absorption cross section. Although a scattering signal of this size is of minor significance when measurements are being made on materials with large capture cross sections, it would introduce large errors with materials having small capture cross sections. With the *B*-type chamber it was found that by carefully adjusting the rectifier switching points, the scattering effects were reduced to the point where equal scattering and absorption cross section in the sample gave signals in the ratio of 1 to 200. Under these conditions the effects of scattering need only be considered when the samples to be measured have absorption cross sections of a fraction of a barn. When the scattering cross section of a sample is known, corrections can be made for it by subtracting from the over-all response the signal yielded by a graphite sample having the same scattering cross section as the unknown absorber.

Although no extensive theory of the scattering effect has been developed, there are certain qualitative observations concerning scattering which are relevant. The scattering signal should vanish identically if the neutron flux were every where the same, since as many neutrons will be scattered into a given solid angle as are scattered out of it. Under ideal circumstances the oscillator should therefore be placed at some point in the pile where the flux is constant over a distance of a few mean free paths.

Since such a position of uniform flux was not available, it was necessary to place the ion chamber in a region where the flux had gradients both in directions parallel and perpendicular to the direction of oscillation. In a spatially varying neutron flux a pure scatterer will perturb the neutron density in its neighborhood because it blocks neutrons originating in a region of high flux from reaching the region of low flux. A chamber placed on the low neutron intensity ("downstream") side of a scatterer will record a lower signal than when the scatterer is absent, while if the chamber is placed on the high-neutron-intensity side the opposite will be true. Thus, if a chamber is arranged to surround the scatterer, it is plausible to suppose that, to a first approximation, the scatterer will give no signal.

In the actual arrangement in which the chamber is coaxial with the oscillating sample, the effect of the transverse gradient is fairly well eliminated. There is still a large longitudinal gradient and there is a beam of neutrons flowing through the channel. As stated previously, we believe the deflection into the chamber of neutrons streaming down the channel to be the chief cause of the observed scattering signal.

There is one other consequence of using a coaxial chamber and that is a hardening of the neutrons at the center of the chamber as a result of absorption of slow neutrons by the surrounding boron. This can be reduced at some sacrifice in sensitivity by keeping the boron coating thin.

PERFORMANCE

There are certain obvious factors which must be considered in order that accurate measurements of absorption cross sections can be made with the pile oscillator. The scattering effect has been considered separately because it has a fundamental bearing on the design and properties of the instrument. The interpretation of cross-section measurements which involve a comparison of the effects produced by two absorbers placed alternately in a thermal neutron flux must be considered. A standard with which to compare other materials must be chosen for which the cross section is accurately known. We have used gold as a standard since it fulfills the above requirement and in addition its cross section has a $1/v$ dependence in a thermal region. If the unknown absorber sample also has a $1/v$ dependence, a comparison with gold will give an unambiguous value for the cross section of the unknown. If the sample in question has a cross section with an unknown energy dependence, then any comparison method will give some average cross section for the existing neutron energy spectrum. The energy spectrum of the neutrons at the center of the chamber has been investigated by measuring the activity of indium foils with and without cadmium covers. A cadmium ratio of 35 was obtained, showing that the neutrons are well thermalized. Further evidence for this is seen in Table I where the cross section of indium as measured with the pile oscillator agrees very closely with the value of $\sigma_{\mathrm{In}}(0.025\text{ ev})=190$ barns obtained by McDaniel[5] and Borst.[6]

As with all cross-section measurements the preparation of the samples with respect to their purity, their physical shape, and their location in the sample carrier must be considered. The samples should be made thin enough so that the corrections for self-absorption are small. Since the measurement involves a comparison, best results can be achieved if the unknown samples and the standard are made as nearly alike as possible with respect to shape, location in the boat, and amount of self-protection.

The amplifier and integrator are subject to the characteristic limitations of electronic equipment. Although the sensitivity changes slightly from day to day, drifts in the instrument are negligible over the period of time necessary to make a measurement. A device has been installed to check the instabilities throughout the electronic circuit and switching mechanism. It consists of an artificial signal which can be introduced to the first stage of the amplifier. The repetition rate of the signal is controlled by the reciprocator wheel, as is the absorber signal, and its amplitude can be adjusted to approximate the absorber signal from the ion chamber. Measurements with these artificial signals from time to time indicate that the major part of the variations between successive sample determinations are not ascribable to the electrical circuits but are due largely to fluctuations in the current in the ionization chamber and in the neutron density. The average neutron density during a series of measurements is held constant to within less than 0.5 percent. The signals which arise from fluctuations in neutron density and in ionization chamber current should be random and should give a small residual signal which causes a variation in successive absorber signal readings. The maximum over-all variation in the integrated signal in terms of the average gold value is about 0.2 mm^2 and the average deviation from the mean is usually less than 0.1 mm^2.

RESULTS

The absorption cross sections of a number of elements and isotopes have been measured with this apparatus and these will be reported in a separate paper. The values obtained for indium and silver are reported here in Table I to give some indication of the capabilities of the apparatus. The atomic absorption cross sections are expressed in barns (10^{-24} cm^2) on the basis of $\sigma_{\mathrm{Au}}(0.025\text{ ev})=95$ barns.

The second row in the table is included to show that the apparatus is capable of measuring small samples of material. The results quoted in the third and fourth rows were derived from measurements on two samples of enriched silver isotopes obtained from the electromagnetic separation plant. One sample was enriched to 90.2 percent of Ag^{107} and the other to 92.2 percent of Ag^{109}. The fifth row gives the values to be expected for natural silver on the basis of the above measurements of the isotopic mixtures, and the last row gives the measured cross section of natural silver. The small discrepancy between the values may be due to contamination of the natural silver sample. Havens and Rainwater[7] have quoted a value of $\sigma_{\mathrm{Ag}}(0.025\text{ ev})=57.3$ barns for natural silver.

ACKNOWLEDGMENTS

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[5] B. D. McDaniel, Phys. Rev. **70**, 832 (1946).

[6] Borst, Ulrich, Osborne, and Hasbrouck Phys. Rev. **70**, 557 (1946).

[7] W. W. Havens, Jr., and L. J. Rainwater, Phys. Rev. **70**, 154 (1946).

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

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Radioactive Br Isotopes

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A radioactive isotope of 2.4-day half-life has been produced in bromine by deuteron bombardment of electromagnetically enriched Se^{76}, and by alpha-particle bombardment of electromagnetically enriched Se^{74}. Assignment of the isotope is made to Br^{77}. A positron end point of 0.4 Mev is determined. In addition to annihilation radiation, gamma-rays and *K*-capture are observed. The ratio of *K*-capture to positron emission is determined to be 20. The cross-section ratio of the (d, n) to the $(d, 2n)$ reaction producing the isotope is 0.3. The ratio of the sum of the cross sections for formation of Br^{77} by $Se^{76}(d, n)$ and $Se^{77}(d, 2n)$ to the cross section for Br^{82} by $Se^{82}(d, 2n)$ is 0.4. A radioactive isotope of 1.7-hour half-life has been produced in bromine by deuteron bombardment and by proton bombardment of enriched Se^{74}. Assignment of the isotope is made to Br^{75}. A positron end point of 1.6 Mev is determined. *K*-capture is observed in the activity. No gamma-ray activity other than that due to annihilation is found. The ratio of *K*-capture to positron emission from the $Se^{74}(d, n)$ reaction is determined to be 4.4. The ratio of the cross section for formation of Br^{75} by $Se^{74}(d, n)$ to that of Br^{82} by $Se^{82}(d, 2n)$ is 2.1. The 4.4-hour Br^{80} isotope has been produced by a $Se(\alpha, p)$ reaction and found to emit positrons with an end point of 0.8 Mev. In producing Br^{77} by proton bombardment of Se the (p, γ) reaction is observed to be two-thirds as probable as the (p, n) reaction.

CYCLOTRON bombardments have been made with alpha-particles, deuterons, and protons on electromagnetically enriched selenium.*** Samples in which the stable isotope Se^{74} was enriched from 0.9 percent to 14.1 percent and samples in which the stable isotope Se^{76} was enriched from 9.5 percent to 41.5 percent were used. For comparison purposes, bombardments were also made with Hilger selenium.

As a result of these bombardments, two previously unreported radioactive isotopes in bromine have been found. The location and characteristic radiations of these isotopes, and also results of investigations on the 4.4-hour Br^{80} isotope, will be presented in this paper.

THE 2.4-DAY Br^{77} ISOTOPE

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

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

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

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