

## Artificial Radioactivity Produced by Neutron Bombardment. II

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*Proc. R. Soc. Lond. A* 1935 **149**, 522-558

doi: [10.1098/rspa.1935.0080](https://doi.org/10.1098/rspa.1935.0080)

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We wish to express our gratitude to Professor G. E. Uhlenbeck for his many helpful suggestions and valuable advice so generously given throughout the course of this work.

One of the authors (A. S. R.) also wishes to express his indebtedness to the Commonwealth Fund of New York for a Fellowship.

#### SUMMARY

The dispersion of sound in hydrogen is investigated. The results show that no variation of velocity occurs below frequencies of the order  $10^6$  cycles per second. This is to be expected from the classical theory of Jeans as well as from the quantum treatment of the inelastic collision between two hydrogen molecules as the calculation shows.

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## Artificial Radioactivity produced by Neutron Bombardment—II

By E. AMALDI, O. D'AGOSTINO, E. FERMI, B. PONTECORVO, F. RASETTI, and E. SEGRÈ

(Communicated by Lord Rutherford, O.M., F.R.S.—Received February 15, 1935)

#### INTRODUCTION

We describe in this paper some further results on artificial radioactivity induced by neutron bombardment, which have been obtained in the Physical Laboratory of the University of Rome, after the publication of our first paper on the same subject.\* Preliminary reports containing the main results have been already published by several of us.†

\* Fermi, Amaldi, D'Agostino, Rasetti, Segrè, 'Proc. Roy. Soc.,' A, vol. 146, p. 483 (1934).

† Fermi, Amaldi, Pontecorvo, Rasetti, Segrè, 'Ric. Scient.,' vol. 2, p. 280 (1934); Fermi, Pontecorvo, Rasetti, 'Ric. Scient.,' vol. 2, p. 380 (1934); Amaldi, D'Agostino, Segrè, 'Ric. Scient.,' vol. 2, p. 381 (1934); Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti, Segrè, 'Ric. Scient.,' vol. 2, p. 467 (1934); vol. 1, p. 123 (1935). Some of our experiments have been repeated with analogous results by Bjerge and Westcott 'Proc. Camb. Phil. Soc.,' vol. 31, p. 145 (1935); we thank them for having submitted their manuscript to us.

By far the most interesting new result concerns the effect of hydrogenated substances on the activation of several elements by neutron bombardment (§ 1). We give an interpretation of this phenomenon by assuming that neutrons are slowed down by impacts against hydrogen nuclei (§ 2). Some anomalously large absorption cross-sections for slow neutrons are discussed in § 3. In many cases the anomalous absorption is connected with the emission of  $\gamma$ -rays (§ 4). § 5 describes some attempts to get information about the energy of the slow neutrons. Scattering experiments on slow neutrons are described in § 6, while § 7 deals with the production of slow neutrons through the action of non-hydrogenated substances. In § 8 the above results are discussed from a theoretical point of view. §§ 9 and 10 are dedicated to chemical methods for the separation of radioactive isotopes, and to improvements in the technique of the measurements. Results of a systematic investigation of the different elements are given in § 11. These results are collected in a table at the end of the paper.

#### § 1—EFFECT OF HYDROGENATED SUBSTANCES ON THE ACTIVATION

In our previous work we had noticed some irregularities in the intensity of the activation of silver by neutrons from a radon + beryllium source, which apparently depended upon some not very clear geometrical factors. Further investigation showed that the activation was strongly influenced by objects surrounding the neutron source, and in particular that the activation could be enormously increased by surrounding the source and the activated substance with a large amount of water or paraffin wax. This effect appeared at once to be due to the presence of hydrogen, as other substances not containing hydrogen failed to give comparable effects (see § 7).

To ascertain whether these large activations were due to the neutrons or to the  $\gamma$ -rays emitted very strongly from our source, we repeated the experiment using as a source 100 mg radium, without beryllium, and found no induced radioactivity. It follows that the effect is actually connected with the neutrons. As a check on this point, we observed the same hydrogen effect with a Po + Be neutron source with an intensity in accordance with the number of neutrons emitted.

Not every substance which is activated by neutrons shows an increase in activity when irradiated under water. Among the strongly influenced activities are: Na (15 h); Al (2.3 m); V (3.75 m); Ag (22 s, 2.3 m); Cu (5 m); Rh (44 s, 3.9 m); I (25 m). The activation of other elements, or possibly of single decay periods, is not influenced by water; among

these are: Si (2.3 m); Al (10 m); Mg (40 s); Mn (3.75 m); Zn (5 m). We have observed that in every case where the active element is known to be an isotope of the bombarded one (about 20 cases), the activation is increased by the presence of water.

In order to measure approximately the sensitivity of the different activations to the action of water or paraffin, we express it in a conventional scale. A cylinder (about 2 cm in diameter and 5 cm in height) of the substance under investigation is irradiated by putting the source in its centre. Source and cylinder are sustained by thin metallic supports at some distance from other objects. The activity reached after a convenient time of irradiation is measured. Afterwards the same cylinder is irradiated with the same source in the same relative position for an equal length of time surrounding both with a large cylindrical block of paraffin 27 cm in diameter and 20 cm in height. The ratio of the activities with and without paraffin is taken as a measure of the sensitivity to hydrogenated substances and denoted by  $\alpha$ . In this scale  $\alpha = 1$  means that the substance shows no increase in activity when irradiated in paraffin. Of course this definition of the sensitivity coefficient is only an empirical one, as it depends somewhat on the geometrical conditions.

Sensitivity coefficients different from one have been observed ranging from  $\alpha = 1.6$  for U (13 m, 100 m) to  $\alpha = 40$  for V (3.75 m). However, it must be pointed out that the last figure may not be the largest, because many substances fail to show some activities when irradiated in air, and these activities appear only by irradiation under water, and then rather weakly.

The ratio of increase in activation due to the presence of paraffin or water is much larger if source and irradiated substance are kept some centimetres apart (see § 5).

## § 2—INTERPRETATION IN TERMS OF SLOW NEUTRONS

The experiments described in the preceding section can be explained on the hypothesis that the effect of water, or better of hydrogen, surrounding the source is due to scattering and slowing down of the primary neutrons by elastic collisions with hydrogen nuclei.

It is easily shown that an impact of a neutron against a proton reduces, on the average, the neutron energy by a factor  $1/e$ . From this it follows that 10 impacts reduce the energy to about  $1/20,000$  of its original value. Assuming the initial energy to be  $4 \cdot 10^6$  electron volts, the energy after 10 impacts would be about 200 electron volts; and less than 20 impacts would be necessary to reduce the energy to thermal equilibrium values.

The phenomena that we have described can now be explained on the assumption that slow neutrons are more easily captured by some nuclei than fast ones. In this and in the following sections we shall discuss our experiments in terms of this hypothesis.

The increase in activity through the action of hydrogen may be ascribed both to the scattering which causes an increased neutron flow through the substance to be activated and to the higher efficiency of the impact of a slow neutron as compared with that of a fast one. In order to show that this second factor is largely important, we performed the following experiment.

A silver cylinder was irradiated with a Rn + Be source containing 350 mC; the distance between source and cylinder was 20 cm. If nothing but air surrounded the source and the cylinder, no activity could be detected in this last after irradiation. Keeping the distance between the source and the irradiated cylinder constant, we then put round the source a cylinder (14 cm diameter and 14 cm depth) filled with water. A strong activity is shown by the silver irradiated in these conditions (about 100 impulses per minute in our counters). This experiment shows that the slow neutrons are much more effective than the fast ones. Indeed, the numbers of neutrons impinging on the silver per second is not increased (possibly slightly lowered through absorption) by the water surrounding the source. As the activation is much increased by the water in these conditions, we must conclude that the yield of activation per impinging neutron is very much enhanced by the slowing down of the neutrons.

### § 3—ABSORPTION OF SLOW NEUTRONS

From the result of the preceding section, that the collision cross-section of slow neutrons for the activation of many elements is much larger than that of fast neutrons, one is led to investigate whether slow neutrons are strongly absorbed by the elements which are most strongly activated by them. We therefore made a systematic investigation of the absorption of slow neutrons in the different elements.

The main purpose of this investigation was to find elements with anomalously large absorption coefficients for the slow neutrons, and hence we generally used fairly thin layers of absorbing material. The arrangement for these absorption measures is shown in fig. 1. The source of neutrons, a Rn + Be tube (S) is put inside a paraffin cylinder P (24 cm diameter, 14 cm height) about 2 cm under the upper surface. A second paraffin cylinder P' was put on P; in its lower surface a hole a few centimetres diameter and 2 or 3 cm in depth was excavated. The

detector of the slow neutrons, a rhodium plate (sometimes also a silver plate), was put inside this hole and its activation measured, after irradiation during a standard length of time, once without absorbing layers and once interposed between two layers A of the absorbing material, as shown in the figure. The ratio of the activities without or with absorbing layers gives the absorption. In these as well as in many other experiments, we generally used rhodium as detector of the slow neutrons, because the activity induced in this element is very large and can be measured very exactly with an ionization chamber; moreover, of the two periods of Rh, 44 s and 3.9 m, practically the first only is of importance, which makes the reduction of the measurements very easy.

As a result of these absorption measures, we found that the half-value thickness  $\delta$  for the absorption of slow neutrons for the different elements

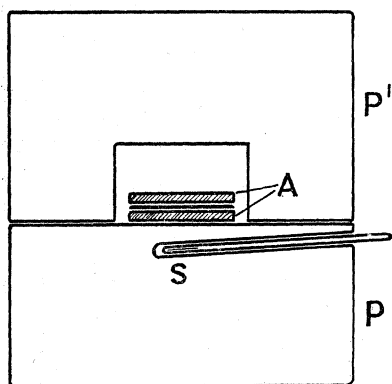


FIG. 1

varies over a very wide range. For several elements it is exceedingly small. As an example we found for boron  $\delta = 0.004 \text{ gm/cm}^2$ ; for yttrium  $\delta = 0.015 \text{ gm/cm}^2$ ; for cadmium  $\delta = 0.014 \text{ gm/cm}^2$ ; for some other elements instead,  $\delta$  is several thousands times larger; for instance, several centimetres of lead absorb less than a few milligrams per  $\text{cm}^2$  of boron.

Expressing the absorption coefficients in terms of a nuclear cross-section  $\sigma$  for the activating impact of a slow neutron, we find in some cases surprisingly large values, e.g.,  $\sigma = 3000 \cdot 10^{-24}$  for B;  $\sigma = 7000 \cdot 10^{-24}$  for Y;  $10,000 \cdot 10^{-24}$  for Cd. This last is the largest cross-section as yet found. It is remarkable how much larger these cross-sections are than the geometrical cross-sections of nuclei, while, as is well known, fast neutrons have cross-sections comparable to the geometrical cross-sections.\* Indeed, we found directly that the absorption of boron for ordinary fast neutrons is at least 1000 times less than the absorption found for slow neutrons in the same element.

These absorption measurements do not refer to homogeneous neutrons. Indeed, the absorption curves are by no means exponentials; the absorption coefficient decreases with increasing thickness, as shown, for instance, by the absorption curve of cadmium, fig. 2. It must also be noticed that the half-value thickness depends to some extent upon the arrangement of

\* Dunning, 'Phys. Rev.', vol. 45, p. 586 (1934).

the paraffin, as this affects the average velocity of the neutrons. For instance absorption of the neutrons inside a hole in paraffin is larger than outside a paraffin block.

#### § 4—EMISSION OF $\gamma$ -RAYS THROUGH THE CAPTURE OF THE NEUTRON

In connection with the anomalous absorption of slow neutrons, one is led to investigate more closely the absorption process. The simplest assumption is to admit that the neutron is captured by the nucleus with formation of an isotope heavier by one mass unit. If this heavier isotope

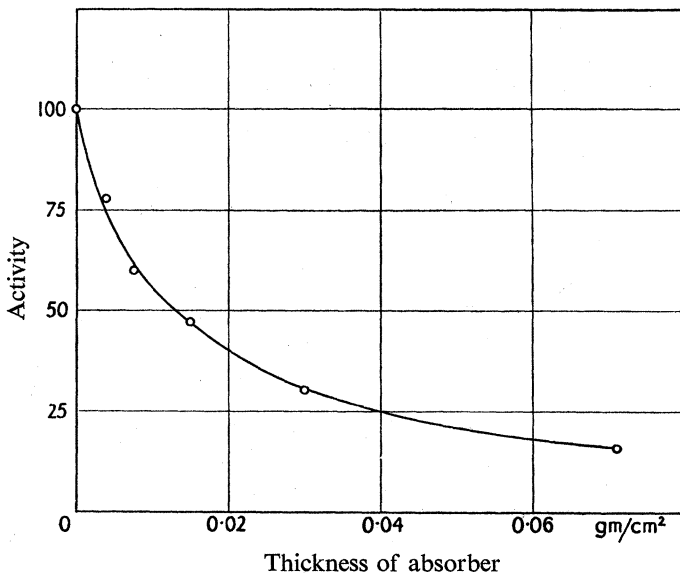


FIG. 2

is unstable, we might expect a strong induced radioactivity for the strongly absorbing elements. This occurs, for example, for indium and iridium, which are known to go over into radioactive isotopes. A rough evaluation of the intensity of the activation of the absorbing substance and of the number of the absorbed neutrons, shows that approximately an activated atom is formed in each absorption process. In other cases it is found that no activation, or at least no strong activation, corresponds to an anomalously large absorption (B, Y, Cd). In these cases we might expect that the capture of the neutron leads to the formation of a stable nucleus. It is obvious that this should happen more easily for elements with many stable isotopes differing in atomic weight by one unit (Cd, Hg).



In both cases, if the slow neutron is simply captured, we might expect that the absorption process should be accompanied by the emission of a  $\gamma$ -radiation, with energy corresponding to the binding energy of the neutron. For fast neutrons a  $\gamma$ -radiation of this type has already been announced by Lea.\*

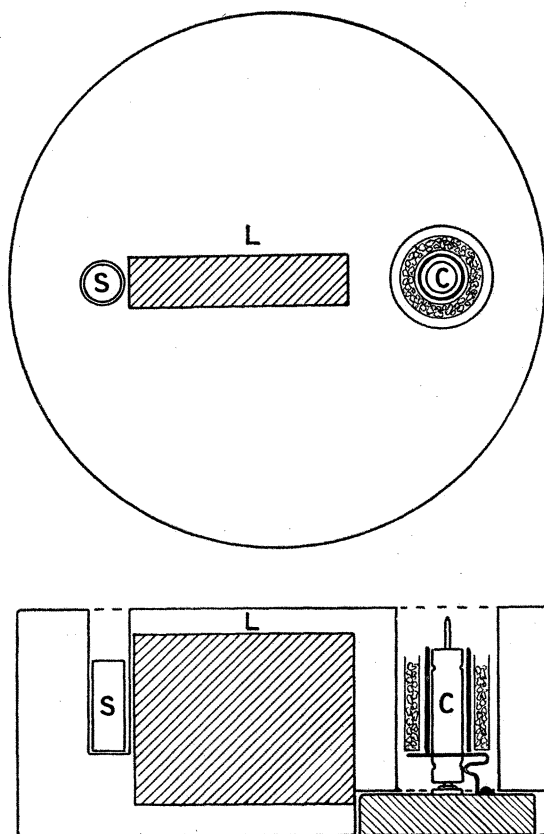


FIG. 3

In the case of the absorption of slow neutrons we were able to show for some elements the emission of a relatively strong  $\gamma$ -radiation of this type by the following experiment.

A Po + Be neutron source S, fig. 3, of about 60 millicuries was immersed in a paraffin block together with one of our standard Geiger Müller counters C. A lead absorber L, 10 cm thick, protected the counter from the direct  $\gamma$ -rays of the source. The counter was surrounded by a lead foil 2 mm thick. The counter registered in these conditions about

\* 'Nature,' vol. 133, p. 24 (1934).



30 counts per minute. Small cylindrical layers of the various substances were placed around the counter, outside the lead foil. We generally observed a very marked increase in the number of counts for the strongly absorbing substances. This was observed, for instance, for Co, Cd, Y, Cl, Ir, Au, Hg, when the number of counts was sometimes more than doubled. An exception to this behaviour is shown by boron and lithium, which, although showing a strong absorption for the slow neutrons, do not emit  $\gamma$ -rays. In these elements the absorption of the slow neutrons has been found to be connected instead to the emission of heavy particles (see § 11). This alternative possibility to the emission of  $\gamma$ -rays appears theoretically likely only for very light elements owing to the low potential barrier.

That the emission of  $\gamma$ -rays, as described before for some substances, was actually due to slow neutrons was shown by the fact that no emission was observed by removing the paraffin. Furthermore, the effect was very much reduced by surrounding substances and counters with a boron screen.

#### § 5—ENERGY OF THE SLOW NEUTRONS

It would be very important to evaluate somehow the mean energy of the slow activating neutrons, and we describe in this section some attempts to do this.

We have already stated that the mean energy of the neutrons is reduced by a factor  $1/e$  for each elastic collision against a proton, provided the values of the energy are large compared with the thermal energy. If the energy of the slow neutrons were really reduced down to this limit, one would expect that the diffusion process might be influenced by temperature. We made an attempt to find such an effect by the following experiment.

A rhodium or a silver detector was irradiated with the Rn + Be neutron source under the same geometrical conditions, once in hot paraffin at 200° C, and once at 20° C in a mixture of benzene and penthane having at this temperature the same density and elementary composition as paraffin at 200° C. No difference in the activation was found in the two tests, within the accuracy of 2%. The mixtures filled a cylinder 26 cm in diameter and 15 cm in height. The detector was on the axis of the cylinder 1 cm under the liquid surface; the source, also on the axis, 2 cm lower. It must be concluded that temperature, at least in these conditions, does not affect the activation, which might be interpreted to mean that the energy of the activating neutrons is higher than the thermal energy. This is, however, not quite conclusive, as the dependency

of both the cross-section for activation and the mean free path upon the velocity for very slow neutrons is unknown.

A direct method of measuring or at least of establishing an upper limit to the energy of the slow neutrons would be to measure the average ionization produced in each collision of a slow neutron against a hydrogen nucleus. This might be done either by measuring the total ionization in a hydrogen-filled chamber or by directly measuring the ionization in a single process by means of a linear amplifier. In both methods the action of the slow neutrons can be separated from that of the fast ones, by screening the chamber with a thin boron layer. Experiments of this kind are being attempted but have not yet led to definite results.

We have investigated whether the increase in activation produced by paraffin is also to be found with different sources of neutrons. We have done this with the neutrons emitted by beryllium irradiated with the  $\gamma$ -rays of radium,\* and found a large effect with this source. This shows that the slow neutrons of paraffin are slower than the neutrons emitted in this nuclear reaction.

#### § 6—SCATTERING AND DIFFUSION OF THE SLOW NEUTRONS

In connection with the absorption measurements described in § 4, we have made a rough survey of the scattering properties of the different substances. The experimental arrangement is shown in fig. 4. The source

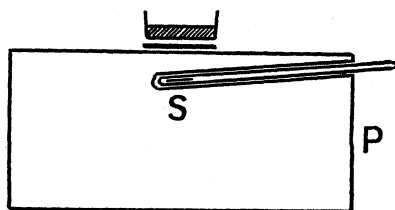


FIG. 4

S is enclosed in the cylindrical paraffin block P as described in § 4. The rhodium plate used as detector was activated by placing it on the top of this paraffin block. The activation was measured once with the substance to be investigated placed in a layer above the rhodium plate, and once without it. In some cases an increased activation was found when a substance was superimposed, indicating an action from slow neutrons scattered back from the substance. For instance, a layer of carbon a few centimetres thick increases the activation about five times. Similar

\* Szilard and Chalmers, 'Nature,' vol. 134, p. 494 (1934).

results are found with several light elements (Be, C, Si). Heavy elements generally produce a much smaller effect. Boron, which has a large absorption coefficient, does not produce any appreciable increase of activity. This shows that the anomalously large absorption of this element is mainly due to real absorption and not to scattering.

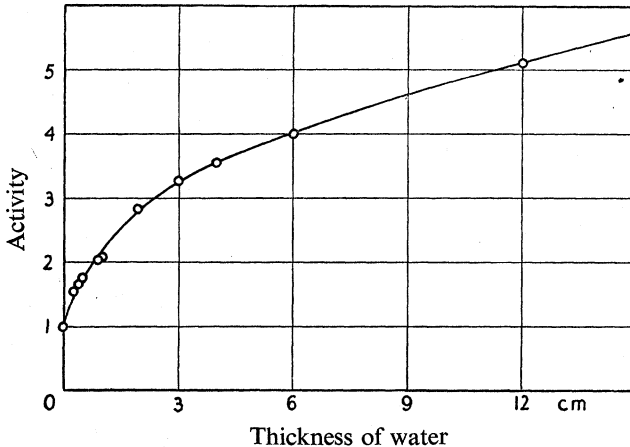


FIG. 5

We have repeated the same experiment with water, obtaining the curve reproduced in fig. 5 for the intensity of activation as a function of the thickness of the scattering layer. A similar result is obtained employing paraffin instead of water.

Other experiments on scattering have been made by the arrangement shown in fig. 6. A paraffin cylinder, 13 cm diameter and 11 cm high, contains in its centre the neutron source S. The detector, a silver or rhodium cylinder R, is kept at a distance of 30 cm from the source. A screen D of the scattering material is interposed between the source and the detector; its dimensions are such as to intercept exactly all the neutrons which might reach the detector from the paraffin block. The difference in the activity of the detector measured when irradiated with or without the screen D gives a measure of the effect of scattering + absorption.

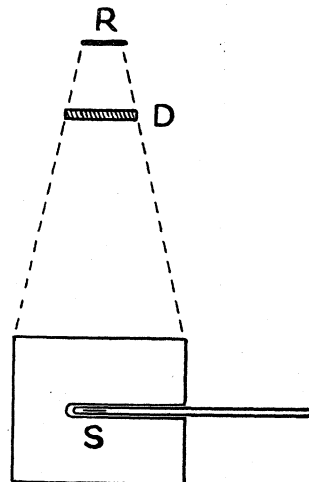


FIG. 6

We found by this apparatus that a paraffin layer  $0.5 \text{ gm/cm}^2$  thick reduces the intensity of the silver activation to about one-half;

here also we found that the absorption is not exponential. A similar result is obtained, using water as a scatterer. From this we might deduce the order of magnitude of the mean free path of slow neutrons in water or paraffin, this being comparable with the observed half-value thickness.

For carbon the half-value thickness is about  $5 \text{ gm/cm}^2$ . Lead of a thickness of about  $4 \text{ gm/cm}^2$  gave a slight decrease in intensity, which is mainly due to scattering.

Another set of experiments was made in order to determine how the intensity of the activation of rhodium in water decreases with the distance from the source. The source was immersed in a water basin of the dimensions  $40 \times 40 \times 100 \text{ cm}$  and the activity induced in rhodium at

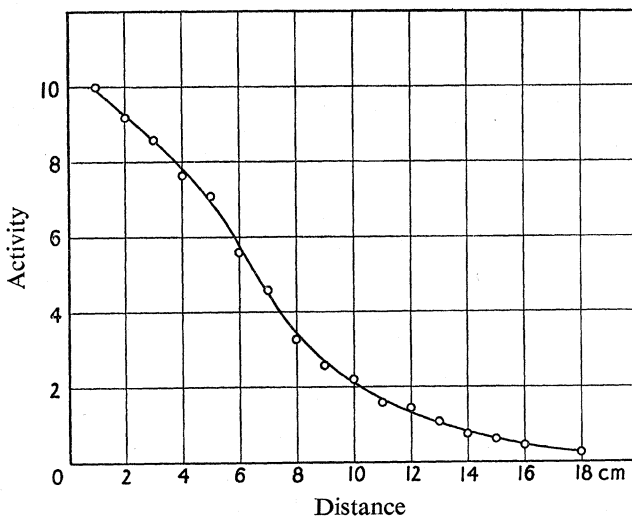


FIG. 7

different distances measured with an ionization chamber. The result is shown in fig. 7, where the activity is plotted as a function of the distance. A similar experiment was made with 2% boric acid dissolved in the water, obtaining a curve similar to the former, only very much reduced in intensity, owing to the absorption of boron.

The general picture of the slowing down process, which one gets from these experiments is the following. The largest part of the diffusion process takes place when the primary neutron still retains much of its original energy, having collided only a few times with hydrogen nuclei; as the velocity is reduced down to the value for which the absorption by boron becomes appreciable, the mean free path in hydrogen appears to

be fairly small, so that the diffusion process after this stage is contained in a small volume. This explains also the fact that the absorption coefficient of boron for the neutrons in water is fairly independent of the distance from the source.

#### § 7—EFFECT OF NON-HYDROGENATED SUBSTANCES ON THE ACTIVATION

We have tried if effects similar to that of water or paraffin could be obtained also with non-hydrogenated substances. Owing to the large amount of materials necessary for these experiments, only a few substances have been tested; Pb, SiO<sub>2</sub>, C, Fe. In all these, with the exception of Fe, a definite increase of the activation of rhodium was observed. Under the geometrical conditions to be described later, the effect was increased by a factor two to five, while in water the factor would have been of some hundreds. That this increase in activity is not simply due to scattering of the neutrons but mainly to their loss in velocity, is proved by the fact that the activation of silicon under the same circumstances is not increased; moreover, very thin cadmium absorbers considerably reduced the activation of rhodium. These facts seem rather surprising chiefly for lead, as it would be difficult to explain the slowing down of neutrons as due to elastic collisions.

The details of these experiments were as follows. We built with lead blocks a cube with a side of about 50 cm. The neutron source was placed in the centre and the rhodium detector 10 cm apart. The activity is about three times larger than in the same geometrical conditions without lead. A similar result is obtained with silver as detector instead of rhodium, while a silicon detector shows no increase in the activity. This fact is to be taken as an indication that the velocity of the neutrons is reduced by the impact against lead nuclei. This view is confirmed by the observation that the activation of rhodium in lead is reduced to half value by an absorber of cadmium of 1 gm/cm<sup>2</sup>. Comparing this absorption of cadmium with that of the same element for the slow neutrons in water (half-value thickness 0·014 gm/cm<sup>2</sup>), one is led to assume that the average velocity of the neutrons in lead is not so low as in water.

A similar experiment was performed, using a graphite cylinder of 3 litres volume. The source was placed in the centre and the rhodium detector at 5 cm distance. The increase in activation was about the same as that produced by lead in the former experiment and the absorption of the neutrons in cadmium was certainly not smaller. Similar results were obtained with silica in the form of "Kieselguhr." The failure to

observe a similar effect with iron is probably due to its relatively large absorption.

We conclude from these experiments that the property of intensifying the activation is shown by most materials. The possible action of surrounding objects, possibly even of air, must always be considered, when experimenting quantitatively on the activation of some substances. For these reasons, the figures given above for the effect of some materials are to be taken only as an indication.

#### § 8—THEORETICAL CONSIDERATIONS ON THE PROPERTIES OF SLOW NEUTRONS

In our first paper we left undecided the question whether whenever the neutron bombardment gives rise to a radio element isotopic with the original one, the neutron is captured or causes the expulsion of a neutron from the struck nucleus, *i.e.*, whether the atomic weight of the radioactive product is  $A + 1$  or  $A - 1$ . We believe that the evidence accumulated since then speaks all in favour of the first hypothesis. The main points of this evidence are the following.

(a) Two new weak activities have been recorded: one, a 15-hour period in sodium, the other a 2.3-minute period in aluminium. Both their identity with known periods, in the first case of aluminium and magnesium, in the second case of silicon and phosphorus, and the chemical evidence in the case of sodium (see § 11), show that the radio elements are respectively isotopes of Na and Al. As both these elements have only one stable isotope, *i.e.*,  $^{23}\text{Na}$  and  $^{27}\text{Al}$ , the choice remains for radio sodium between  $^{24}\text{Na}$  and  $^{22}\text{Na}$ , for radio aluminium between  $^{28}\text{Al}$  and  $^{26}\text{Al}$ . Now both the lighter isotopes  $^{22}\text{Na}$  and  $^{26}\text{Al}$  are known from other nuclear reactions to have different periods and to emit positrons instead of electrons, so that it seems well founded to admit that, at least in these cases, the neutron is captured.

(b) While there is no theoretical difficulty in understanding how a neutron with a negligible kinetic energy can be captured by a nucleus, it seems unlikely that it could knock out a stably bound neutron from the nucleus. Still more difficult it would be to understand that in the latter case energy could be left over to account for the emission of  $\gamma$ -rays.

We shall, then, discuss the experimental facts on the standpoint that the neutrons, and particularly slow ones, are easily captured by many nuclei.

There are, however, some theoretical difficulties in understanding this capture process, or at least to account for the large cross-sections that

have been experimentally observed in some cases. Nevertheless it may be worth while to state some general consequences of the theory, which must be always kept in mind in the discussion of this problem.

Let us admit, as it has been generally assumed, that the forces acting between the neutron and a nucleus extend about as far as the nuclear radius itself. If this is so, the de Broglie wave-length is, for fast neutrons, of the order of the radius of action, and consequently for slow neutrons is much larger. The well-known theory of the impact, in which the nucleus is treated as a potential hole, takes in this case an extremely simple form. Let  $\psi$  be the  $s$ -eigenfunction corresponding to zero energy. In fig. 8 the product  $r\psi$  is plotted as a function of the radius vector  $r$ , and  $\rho$  represents the radius of action. The curve has an irregular shape for  $r < \rho$ , while for  $r > \rho$  it becomes a straight line. We normalize  $\psi$  taking  $\psi(0) = 1$ . Let then the equation of the straight line into which  $r\psi$  goes over for large values of  $r$  be

$$r\psi \rightarrow \eta(a + r),$$

where the geometrical meaning of  $\eta$  and  $a$  is clear from the figure. The values of these two quantities could be easily calculated if one knew the form of the potential hole representing the nucleus. The cross-section for elastic collisions in the limit for low velocities is found to be

$$\sigma_{el} = 4\pi a^2, \quad (1)$$

while the density of probability for the neutron to be found in the centre of the nucleus is

$$P = n/\eta^2, \quad (2)$$

where  $n$  is the density of the neutrons outside of the nucleus.

Whatever the mechanism of capture may be, it is natural to assume that, at least to a certain approximation, the probability of capture of the neutron by a nucleus per unit time will be proportional to  $P$ , *i.e.*, that this probability will be given by  $kn/\eta^2$ , where  $k$  is a constant for each nucleus.

This probability can be expressed in terms of a cross-section  $\sigma_{ca}$  for the capture process through the relation

$$\sigma_{ca} = k/\eta^2 v, \quad (3)$$

where  $v$  is the velocity of the neutron. A limit to the validity of (3) is set by the fact that  $\sigma_{ca}$  can evidently be at most of the order of magnitude of the square of de Broglie wave-length. This allows one, considering the largest cross-sections found experimentally (Cd,  $\sigma_{ca} = 10^{-20}$ ), to set



an upper limit for the energy of the slow neutrons. This limit turns out to be a few hundred volts. However, it must be borne in mind that this deduction is valid only under the explicitly stated assumptions.

Formula (3), valid only for low velocities, gives a cross-section for capture for a given nucleus inversely proportional to the velocity of the neutron. This explains why the cross-section for capture is generally larger for slow neutrons than for fast ones. This result may also be expressed by saying that the mean life of a slow neutron in a substance is independent of its velocity. While the capture cross-section is inversely proportional to the velocity, the cross-section for elastic impact expressed

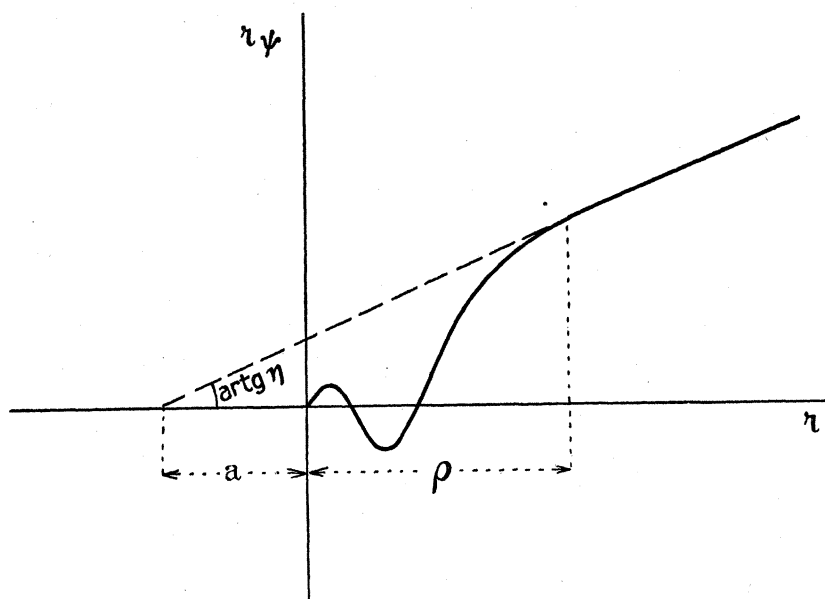


FIG. 8

by (1) is independent of the velocity, which means that the mean free path for this type of collisions is also independent of the velocity.

One more feature of (3) is that  $\sigma_{ca}$  is inversely proportional to  $\eta^2$ . The straight line of fig. 8 may occasionally be nearly parallel to the abscissæ axis. In this case  $\eta$  is very small and consequently the capture section becomes very large. This behaviour of the eigenfunction is probably responsible for the anomalously large cross-sections observed for a few nuclei.

In order to deduce from (3) the absolute values of the cross-sections, it would be necessary to know also the values of  $k$ , which quantity depends on the physical mechanism of the capture. We know from the experi-

ments that there are two different processes. In some light elements (Li, B) the capture of the neutron is followed by the emission of heavy particles; while in the heavier elements the normal process seems to be capture with emission of a  $\gamma$ -ray. The binding of an additional neutron to a nucleus sets free an amount of energy which, on the average, has a value about 7 million electron volts. This surplus energy might in some cases produce the emission of an  $\alpha$ -particle, provided the potential barrier surrounding the nucleus is low enough to allow for a quick escape of the particle. Therefore this process is expected to occur only for the lightest elements, whereas in the activation by fast neutrons, owing to the kinetic energy of the impinging neutron which is to be added to the binding energy, the emission of charged particles is possible also in elements of higher atomic weight.\*

In the case of capture with emission of a  $\gamma$ -ray, which is observed for elements of any atomic weight up to the highest, the ordinarily accepted mechanism of irradiation gives apparently rather too small a value for  $k$ .  $k$  depends upon two factors which are rather difficult to evaluate: matrix elements and energy of the emitted  $\gamma$ -quanta. As the probability of emission of a  $\gamma$ -quantum is *ceteris paribus* proportional to  $\nu^3$ , it is to be assumed that processes by which the binding energy of the neutron is very large ought to be preferred; this binding energy might in several cases considerably exceed 10 million volts. Nevertheless one ought probably to assume rather too high a value for the matrix elements in order to get a plausible interpretation of the relative frequency of the anomalously large cross-sections. From the theory one would also expect that generally anomalous cross-sections for the capture process were connected to anomalous elastic cross-sections. No experimental evidence has yet been found for this fact.

In this section we shall also discuss briefly the velocity distribution law for the slow neutrons in hydrogen. From the above theoretical considerations (formulae (1) and (3)), it seems plausible to assume that neutrons having velocities smaller than a certain limit have a constant mean free path  $\lambda$  for elastic collisions, and a constant mean life-time  $\tau$  before they are captured by the nuclei present in the medium. One can then easily show that the number of neutrons in a hydrogenated substance, having velocities between  $v$  and  $v + dv$ , is proportional, for  $v$  less than the above-stated limit to

$$\frac{v dv}{(v + \lambda/\tau)^3} \quad (4)$$

\* Meitner, 'Naturwiss.', vol. 45, p. 789 (1934).

This distribution law of the slow neutrons might be used for interpreting the fact that the absorption curves of the slow neutrons are not exponentials.

### § 9—SEPARATION OF RADIOACTIVE ISOTOPES

Szilard and Chalmers\* were the first to separate radioactive iodine from usual iodine by chemical methods. We have extended their procedure to some other cases, using also inorganic instead of organic compounds. The principle of the method is the following: let us suppose that the element before irradiation is bound in a molecule or radical which, once decomposed, has a practically negligible probability of being rebuilt. As the neutron strikes the atom, this is generally knocked out from the molecule, and has a tendency to remain in an atomic or ionic state. It follows that, after irradiation, the modified atoms are in a different chemical state from the main bulk of the unmodified ones, and can thus be separated by convenient reactions. The energy of the chemical bond is a few volts, and, even admitting that the impinging neutron might have a negligible energy, the recoil of the  $\gamma$ -quantum emitted in the capture of the neutron would be sufficient to overcome the binding chemical forces.

Szilard and Chalmers separated radioactive iodine from irradiated ethyl iodide adding a trace of free iodine, reducing and precipitating  $I^-$  with  $AgNO_3$ . The same method was applied by us to bromoform, chloroform, carbon tetrachloride and some other halogenic organic compounds. We were always able to separate almost completely the radioactive halogen from the bulk of the inactive substance.

We have also separated radioactive chlorine (35 m) starting from sodium chlorate. The chlorine atom is knocked out of the  $ClO_3^-$  ion by the neutron impact; adding a small trace of  $Cl^-$  to carry the radioactive isotope and precipitating carefully with  $AgNO_3$  with addition of  $HNO_3$  to prevent precipitation of  $AgClO_3$ , one finds the activity concentrated in the  $AgCl$  precipitate. Similar results are obtained also with bromates and iodates. In these last compounds the nitric acid was substituted with ammonia. From 70 to 90% of the total activity is concentrated in the precipitate.

Cacodylic acid  $(CH_3)_2AsOOH$  is a good starting substance for concentrating radioactive arsenic. By precipitating arsenic sulphide from irradiated cacodylic acid, it is possible to concentrate the activity.

Potassium permanganate irradiated and then filtered through an ordinary paper filter left much of the activity on the filter with manganese

\* 'Nature,' vol. 134, p. 462 (1934).

dioxide formed in the oxidation of the paper. About 80% of the total activity was separated, adding a trace of a manganous salt and precipitating the manganous carbonate. The  $\text{MnO}_4^-$  ion is dissociated by the neutron bombardment and the manganese atom is left in lower states of oxidation; these are collected with manganese peroxide on the filter, or precipitated by carbonates. We tried this separation starting either from solid salts or from solutions; there is no very remarkable difference between the two cases. Also no marked influence of the acidity or alkalinity of the irradiated solution was observed.

A physical method for separating radioactive isotopes is analogous to the well-known method of collecting the active deposit from the emanation. We tried this method with a gaseous iodine compound. A glass cylinder of about 1 litre volume with a large cylindrical aluminium electrode close to the wall and a nickel electrode along the axis was filled with methyl or ethyl iodide saturated vapour. The temperature of the cylinder was regulated so that the vapour pressure was about an atmosphere. The whole apparatus was immersed in hot water and a potential difference of 3000 volts applied to the electrodes. A  $\text{Rn} + \text{Be}$  source was placed outside the wall. After irradiation the nickel electrode was removed and showed an activity decaying with the iodine period. Nickel was used as a support because it does not become radioactive under neutron bombardment. The yield was rather a poor one. Inversion of the polarity of the electrodes gave no consistent results.

#### § 10—METHODS OF MEASUREMENTS

The observed fact of the intense activating effect of the slow neutrons opened the possibility of obtaining stronger sources of the artificial radio elements than were available last year. This allows more accurate measurements of the constants of these substances by using an ionization chamber instead of Geiger Müller counters.

The ionization chamber was built of steel; the inner electrodes were a wire net and a brass rod. The  $\beta$ -rays entered the chamber through a circular window of 6 cm diameter on the top, closed by an aluminium foil 0.01 cm thick. The chamber was filled with  $\text{CO}_2$  at 3 atmospheres pressure. The chamber was connected to a Perucca electrometer, the total capacity of the system being about 20 cm, the electrometer sensitivity 0.02 volts per scale division. The system was perfectly constant both in its zero effect and in its sensitivity (as controlled by a uranium standard preparation). We reproduce in figs. 9 and 10 some decay curves obtained with this apparatus.

The same apparatus was used for measurements of the absorption of the  $\beta$ -rays. The window was protected with aluminium foils and the activity plotted as a function of the aluminium thickness. The absorption

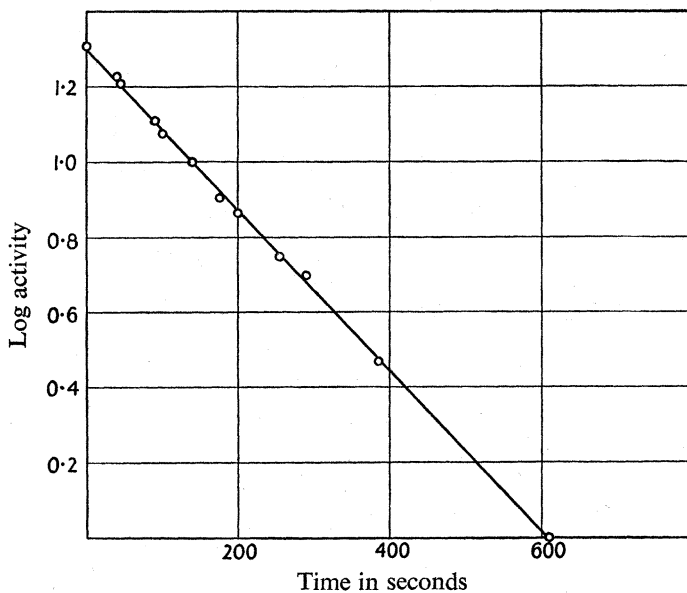


FIG. 9

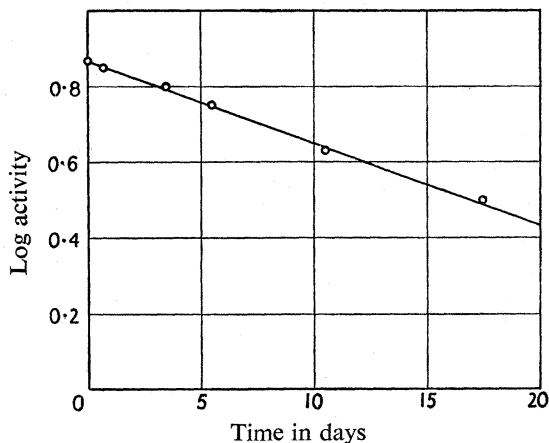


FIG. 10

curve was roughly exponential. Sometimes substances show besides the  $\beta$ -activity a strong  $\gamma$ -radiation. We conventionally attributed to  $\gamma$ -rays the residual ionization when the chamber was screened with 2 mm of lead. In the evaluation of the absorption coefficients for  $\beta$ -rays allow-

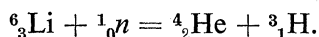
ance has been made for the  $\gamma$ -radiation. To check the measurements, the absorption coefficients of the  $\beta$ -rays of RaE and UX<sub>2</sub> have been measured and results obtained in accordance with the values commonly admitted.

### § 11—SYSTEMATIC INVESTIGATION OF ELEMENTS

In this section we shall report all the new data that we have found about each element, both as regards the induced activities and the properties with respect to slow neutrons. Some data differ slightly from our previous ones, owing to the increased precision of our measurements.

1—*Hydrogen*—No activity could be detected either in water or in paraffin irradiated in a large can of water with 500 millicuries Rn + Be for several days.

3—*Lithium*—Lithium hydroxide was found to be inactive after irradiation with slow neutrons (14 hours, 400 millicuries). Although lithium remains inactive, it strongly absorbs the slow neutrons; half-value thickness  $\delta = 0.05$  gm/cm<sup>2</sup>. This absorption is not accompanied by a  $\gamma$ -radiation. It was shown independently by Chadwick and Goldhaber\* and by us that when the slow neutrons are absorbed, heavy charged particles are emitted. According to Chadwick and Goldhaber, the nuclear process is represented by the following reaction,

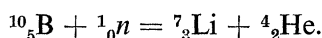


4—*Beryllium*—Metallic beryllium (purity 99%), strongly irradiated with slow neutrons, showed only an extremely weak activity possibly due to impurities. Owing to the very strong activation of several elements when irradiated under water, impurities might easily be misleading.

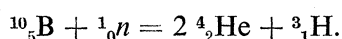
5—*Boron*—Metallic boron irradiated 14 hours under water with 500 millicuries was found inactive. Boron has the highest absorption coefficient as yet found for slow neutrons,  $\delta = 0.004$  gm/cm<sup>2</sup>, corresponding to a cross-section of about  $3 \cdot 10^{-21}$  cm<sup>2</sup>. No  $\gamma$ -rays have been found to accompany this absorption: instead of a  $\gamma$ -radiation in this case as well as for lithium,  $\alpha$ -particles are emitted, as was shown by Chadwick and Goldhaber\* and by us. This effect can be easily detected by the strong discharge in an ionization chamber filled with boron trifluoride surrounded by paraffin and irradiated with a Po + Be neutron source. Screening the ionization chamber with a thin cadmium foil

\* 'Nature,' vol. 135, p. 65 (1935).

in order to absorb the slow neutrons, reduces considerably the ionization current. The same effect was observed with the ionization chamber filled with air, some boron being spread on its floor. The emission of  $\alpha$ -particles was also detected with a small ionization chamber connected to a linear amplifier, either spreading some boron on its walls or filling it with boron trifluoride. In order to explain these phenomena we have proposed the nuclear reaction,



Chadwick and Goldhaber have proposed instead the reaction,



We do not think that there is at present sufficient evidence to decide between these two possibilities, and we are now experimenting to try to get a more exact measurement of the number of ions formed in each process in an ionization chamber containing boron either in a gaseous form (total process) or spread on its walls (effect of only one or two particles). We are also trying to observe the disintegration in a Wilson chamber containing a gaseous compound of boron.\*

6—*Carbon*—No activity; see hydrogen. For the scattering properties see § 6.

7—*Nitrogen*—Ammonium nitrate irradiated 12 hours with 600 millicuries under water showed no activity.

8—*Oxygen*—No activity, see hydrogen.

9—*Fluorine*—Both activities of this element (periods 9 seconds and 40 seconds)\* are not sensitive to hydrogenated substances.

11—*Sodium*—This element has two activities: one of these (period 40 seconds) is not sensitive to hydrogenated substances. A very weak activity with a long period was reported by Bjerger and Westcott.† As this activity is strongly enhanced by water, we were able to measure its period with reasonable accuracy and found it to be 15 hours. Owing to the theoretical importance of this activity (see § 8), we compared very carefully its decay curve with that of the long period of aluminium in order to check their identity. For a chemical investigation of the active substance we irradiated pure sodium carbonate (Kahlbaum). We dis-

\* [Note added in proof—Taylor and Goldhaber, 'Nature,' vol. 135, p. 341 (1935), have shown that the reaction takes place according to the first scheme.]

† Bjerger and Westcott, 'Nature,' vol. 134, p. 286 (1934).



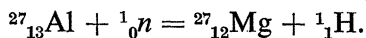
solved the irradiated substance in hydrochloric acid and added aluminium and magnesium chlorides. A precipitate of the hydroxides of the latter elements obtained by adding ammonia, was found inactive. Afterwards we added some sodium fluoride to the solution, and precipitated the fluorine as barium fluoride; this precipitate was also found inactive. The solution containing the original sodium was then evaporated and ignited gently, in order to eliminate neon, where an active isotope of this element would have been formed. The activity was found in the dried sodium salt. We conclude that the active product is an isotope of sodium,  $^{24}\text{Na}$ . The same isotope was produced by us last year by bombarding magnesium or aluminium with neutrons.  $^{24}\text{Na}$  has also been produced recently in considerable amounts and studied very completely by Lawrence\* bombarding several elements with artificially accelerated particles.

12—*Magnesium*—Pure magnesium oxide (Kahlbaum), especially tested by us in order to ensure that it was aluminium free, was irradiated under water. The substance was kept at some distance from the source in order to prevent the activation of the periods not sensitive to water. A new very weak activity with a period of about 10 minutes was found. As this period coincides with the 10-minute period of aluminium, which is known to be due to  $^{27}\text{Mg}$  (see aluminium), it is very likely that it is due to the same isotope formed by the capture of a neutron by  $^{26}\text{Mg}$  present in an amount of 11% in ordinary magnesium.

The 40-second period is insensitive to water.

13—*Aluminium*—Aluminium irradiated in water shows a fairly strong new activity decaying with a period of 2.3 minutes (measured with an ionization chamber). Irradiated outside of water, this activity is extremely weak. As the period of the new activity coincides with the 2.3-minute period of silicon, which is due to  $^{28}\text{Al}$ , we assume that this activity is also due to the same isotope formed by capture of a neutron from  $^{27}\text{Al}$ .

The second period of aluminium has been measured with the ionization chamber and found to be 10 minutes instead of 12. This activity is insensitive to water. A chemical separation of the carrier of this activity has been performed. Irradiated metallic aluminium was dissolved in a caustic soda solution and magnesium chloride was added. The precipitate of magnesium hydroxide carried the 10-minute activity. We assume that the active isotope is  $^{27}\text{Mg}$  formed according to the reaction,



\* 'Phys. Rev.,' vol. 47, p. 17 (1935).

14—*Silicon*—We have determined with the ionization chamber the short period of this element, finding it to be 2·3 minutes. This activity is insensitive to water.

Besides this activity, we found a new longer period of some hours in fused silica irradiated in water. This activity is very weak and very sensitive to water. We think probably that its carrier is  $^{31}\text{Si}$  which is obtained by irradiated phosphorus and has a period of 2·4 hours.  $^{31}\text{Si}$  could be formed by capture of a neutron from  $^{30}\text{Si}$  present in an amount of 3%.

15—*Phosphorus*—The short-period activity of this element (2·3 minutes) is not enhanced by water. Curie, Joliot and Preiswerk\* ascribe this period to  $^{28}\text{Al}$ . A chemical test in favour of this hypothesis is the following: we irradiated phosphoric acid, neutralized the solution with sodium carbonate, and added aluminium chloride; the activity was found to be concentrated in the precipitated aluminium.

We have observed, with the aid of the ionization chamber, the decay-curve of the longer period of phosphorus. Its period is 2·4 hours instead of 3 as given before. We have also measured, with the ionization chamber, the half-value thickness of the corresponding  $\beta$ -rays and found it to be 0·15 gm/cm<sup>2</sup> Al.

16—*Sulphur*—We have determined in the ionization chamber the period of phosphorus extracted from irradiated sulphur. We found: period 14 days, half-value thickness of the  $\beta$ -rays 0·10 gm/cm<sup>2</sup> Al.

17—*Chlorine*—Chlorine irradiated under water showed a new period of 35 minutes measured electrometrically. For the chemical tests on the carrier of this activity see § 9.

Chlorine absorbs fairly strongly the slow neutrons (half-value thickness  $\delta = 0\cdot3$  gm/cm<sup>2</sup>). The process of absorption is accompanied by emission of  $\gamma$ -rays.

19—*Potassium*—We have found in irradiated potassium an induced activity strongly sensitive to water, decaying with a period of 16 hours. A chemical investigation of the carrier of the activity, performed by the same method described for sodium, excluded the elements Cl, A, Ca. We conclude therefrom that the activity is probably carried by an isotope of potassium. According to v. Hevesy†, this isotope is to be identified with a  $^{42}\text{K}$ , that was obtained by him by neutron bombardment of scandium, and has the same decay period.

\* 'C.R. Acad. Sci. Paris,' vol. 198, p. 2089 (1934).

† 'Nature,' vol. 135, p. 96 (1935).

20—*Calcium*—No activity was found in calcium fluoride irradiated 14 hours in water with a 600 millicuries source.

23—*Vanadium*—The decay of the activity induced in vanadium has been measured in the ionization chamber with the following results: half-value period 3.75 minutes; half-value thickness of the  $\beta$ -rays 0.17 gm/cm<sup>2</sup> Al. The  $\beta$ -rays are accompanied by a  $\gamma$ -radiation. The activation of vanadium is strongly sensitive to hydrogenated substances; with the definition of § 1,  $\alpha = 40$ .

24—*Chromium*—The activity of chromium is insensitive to water.

25—*Manganese*—The activity with short period (3.75 minutes) is insensitive to water ( $\alpha = 1$ ). On the other hand, the activity with longer period (2.5 hours measured in the ionization chamber) is strongly enhanced by water ( $\alpha = 23$ ). Half-value thickness of  $\beta$ -rays measured electrometrically is 0.14 gm/cm<sup>2</sup> Al; the disintegration is accompanied by  $\gamma$ -rays. The 2.5-hour product is known to be an isotope of manganese; in § 9 a method for concentrating the activity has been described. In order to get new evidence in favour of the fact that the active product is really an isotope of manganese, we first concentrated the activity obtained in irradiated manganese permanganate by a precipitation of manganese carbonate. The carbonate containing the activity was then dissolved in hydrochloric acid, and large amounts of chromium, vanadium and iron salts were added to the solution. Afterwards the manganese was separated once again as dioxide, with nitric acid and sodium chlorate. The manganese precipitate carried the activity, while the fractions containing chromium, vanadium and iron were found to be inactive.

26—*Iron*—The activity of this element (period 2.5 hours) is insensitive to water. Half-value thickness for the absorption of slow neutrons 8 gm/cm<sup>2</sup>.

27—*Cobalt*—This element absorbs fairly strongly the slow neutrons; half-value thickness 0.7 gm/cm<sup>2</sup>. The absorption is accompanied by the emission of a  $\gamma$ -radiation.

28—*Nickel*—Strongly irradiated nickel showed only a dubious trace of activity.

29—*Copper*—Both induced activities of this element (periods 5 minutes, measured electrometrically, and 10 hours) are strongly enhanced by water. For the first  $\alpha = 15$ . Copper absorbs the slow neutrons with a half-value thickness of about 3 gm/cm<sup>2</sup>; this absorption is accompanied by a weak  $\gamma$ -radiation.

Irradiated metallic copper was dissolved in hydrochloric acid, and small quantities of cobalt, nickel and zinc salts were added. Copper sulphide was precipitated from the acid solution and found to be active. The precipitates of the zinc, cobalt and nickel sulphides, obtained by neutralizing the solution and adding ammonium sulphide, were inactive. As the time employed for this test was rather long, the test refers only to the longer period. The carrier of this activity can then be assumed to be an isotope of copper, as suggested by Bjerge and Westcott (*loc. cit.*).

30—*Zinc*—The activity of the short period of zinc is not enhanced by water. The longer period was measured electrometrically and found to be 10 hours. The carrier of this activity has been investigated by means of the following test: irradiated metallic zinc was dissolved in hydrochloric acid, and a small quantity of copper, nickel and cobalt salts added. Copper was precipitated partially by reduction on small traces of undissolved metallic zinc and partially as sulphide in the acid solution. The collected copper was strongly active. Neutralizing the solution and adding ammonium sulphide, the other elements were precipitated and found to be inactive. This confirms the results of Bjerge and Westcott that the long period of zinc is due to copper, probably to the same isotope of copper which is responsible for the longer period of this element. There is only a certain difference between these authors and us with respect to the period (6 hours according to Bjerge and Westcott).

31—*Gallium*—The 20-minute period (measured electrometrically) is not very sensitive to water ( $\alpha = 3$ ). Half-value thickness of the corresponding  $\beta$ -rays is 0.17 gm/cm<sup>2</sup> Al. The carrier of this activity is probably an isotope of gallium. In order to test this point, we irradiated gallium nitrate and afterwards added to the solution traces of copper and zinc. Copper was separated as a metallic deposit on zinc powder and zinc as zinc mercury sulphocyanate after adding mercury sulphocyanate. Both elements were found to be inactive.

Besides this 20-minute activity, we have also found, irradiating under water, a new activity which is accompanied by a rather strong  $\gamma$ -radiation; it decays with a period of 23 hours (measured electrometrically).

33—*Arsenic*—The activity of this element is strongly sensitive to water ( $\alpha = 6$ ). We have measured electrometrically its period (26 hours) and its half-value thickness of the  $\beta$ -rays (0.16 gm/cm<sup>2</sup> Al). For a concentration of the activity, see § 9.

34—*Selenium*—The activity of this element (period 35 minutes) is sensitive to water ( $\alpha = 4$ ). Irradiated selenious anhydride was dissolved in 30% hydrochloric acid and some arsenious anhydride added to the

solution. We precipitated metallic selenium by reduction with gaseous sulphurous anhydride, and found it strongly active. We precipitated from the solution arsenic sulphide and found it inactive. This test seems to rule out also germanium, and we conclude that the activity is due to an isotope of selenium.

35—*Bromine*—Both activities of this element are sensitive to water. The short-period activity has  $\alpha = 10$ . The periods have been measured electrometrically; they are 18 minutes and 4.2 hours. The half-value thickness of the  $\gamma$ -rays is for both activities  $0.12 \text{ gm/cm}^2 \text{ Al}$ , and both are accompanied by  $\gamma$ -rays. For the concentration of the activity see § 9.

38—*Strontium*—No activity was found after a long and strong irradiation under water.

39—*Yttrium*—Strongly irradiated yttrium oxide showed only a very weak activity possibly due to impurities. Yttrium absorbs very intensively the slow neutrons (half-value thickness  $\delta = 0.015 \text{ gm/cm}^2$ ). This absorption is accompanied by  $\gamma$ -rays.

40—*Zirconium*—Strongly irradiated zirconium nitrate showed only a very weak activity probably due to impurities.

41—*Niobium*—The same as zirconium.

43—*Rhodium*—The short-period activity is sensitive to water ( $\alpha = 15$ ). Period and half-value thickness of the  $\beta$ -rays have been determined electrometrically (44 seconds;  $0.15 \text{ gm/cm}^2 \text{ Al}$ ). We also made a more accurate measurement in the ionization chamber of the longer period and found it to be 3.9 minutes. The activity is accompanied by a weak  $\gamma$ -radiation. Rhodium absorbs fairly strongly the slow neutrons (half-value thickness  $0.3 \text{ gm/cm}^2$ ): the absorption probably corresponds to the formation of the active isotopes.

46—*Palladium*—Also the activities of this element are sensitive to water. We find at least two periods: a short one of about a quarter of an hour and one of about 12 hours. McLennan, Grimmett, and Read record a period of 14 hours, which is consistent with our precision.

47—*Silver*—The two periods have been redetermined with the ionization chamber. They are 22 seconds and 2.3 minutes. They are both very sensitive to water, having  $\alpha = 30$  and 15 respectively. To the strong activation of this element corresponds a considerable absorption for slow neutrons (half-value thickness  $\delta = 1.2 \text{ gm/cm}^2$ ).

We added palladium nitrate and rhodium chloride to a solution of irradiated silver nitrate. Adding hydrochloric acid, we precipitated silver which was found active. From the filtered solution we precipitated

palladium with dimethylglyoxime and rhodium by reduction. Both were inactive. This test is valid only for the longer period, owing to the time employed, and shows that the carrier of the activity is probably an isotope of silver.

48—*Cadmium*—Cadmium irradiation under different conditions showed several weak activities with various periods not yet identified. Cadmium absorbs with great intensity the slow neutrons. (Half-value thickness  $0.013 \text{ gm/cm}^2$ .) The corresponding cross-section is the largest as yet found for slow neutrons ( $\sigma = 10^{-20} \text{ cm}^2$ ). The absorption is accompanied by an intensive  $\gamma$ -radiation and probably corresponds to the transformation of a stable isotope of cadmium into another stable isotope of the same element.

49—*Indium*—The activity induced in indium shows three periods: the shortest period (13 seconds) corresponds to an activity sensitive to water ( $\alpha = 12$ ). Also the second period (54 minutes, measured electrometrically) is very sensitive to water. Magnetic deflection experiments show that the corresponding electrons are negative; their half-value thickness is  $0.045 \text{ gm/cm}^2 \text{ Al}$ . A still longer period of some hours is recorded by Szilard and Chalmers;\* this last activity is either insensitive to water or is only moderately sensitive.

Chemical tests have been made in order to identify the carriers of the last two activities. To a solution of irradiated indium nitrate, silver was added and precipitated as silver chloride; the precipitate was inactive. Afterwards we added to the solution tin, antimony and cadmium and precipitated them as sulphides with sulphuretted hydrogen. The acidity of the solution must be adjusted in such a way as to leave the indium in solution while precipitating the other metals. This precipitate also was inactive; neutralizing the solution, we precipitated the indium sulphide which carried the activity.

Corresponding to the strong activation of indium, it is found that this element has a considerable absorption power for the slow neutrons: half-value thickness  $\delta = 0.3 \text{ gm/cm}^2$ .

50—*Tin*—Tin strongly irradiated under water showed no activity.

51—*Antimony*—We have found an induced activity in this element, decaying with a period of 2.5 days; the activation is sensitive to hydrogenated substances. The half-value thickness for the emitted  $\beta$ -rays is  $0.09 \text{ gm/cm}^2 \text{ Al}$ . The following chemical test indicates that the carrier of this activity is probably an isotope of antimony. We dissolved metallic

\* 'Nature,' vol. 135, p. 493 (1935).



irradiated antimony in aqua regia and added some tin to the solution; after separation of tin as a sulphide according to Clarke, we found the activity in a precipitate of sulphide of antimony. The antimony sulphide was then dissolved again; indium was added to the solution and antimony separated as a sulphide in a moderately acid solution; the solution was neutralized and indium precipitated and found to be inactive. To a new solution of the antimony we added tellurium and iodine and separated the first by reduction and the second as a silver iodide. Both were inactive.

52—*Tellurium*—Shows a weak activity sensitive to water; the period resulted 45 minutes instead of 30 as given in our former paper.

53—*Iodine*—Period and half-value thickness of the  $\beta$ -rays were determined electrometrically: period 25 minutes; half-value thickness  $0.11 \text{ gm/cm}^2 \text{ Al}$ . The activation is moderately sensitive to water ( $\alpha = 5$ ). For the concentration of the activity see § 9.

56—*Barium*—A new activity sensitive to water ( $\alpha = 8$ ) with a period of 80 minutes has been found. The following chemical test is in favour of the assumption that the carrier of this activity is an isotope of barium. We dissolved irradiated barium hydroxide in hydrochloric acid, and added a small quantity of sodium chloride and precipitated barium sulphate. The activity was carried by the precipitate; we evaporated the solution and found the residual sodium to be inactive.

57—*Lanthanum*—No activity was found after strong irradiation under water.

58—*Cerium*—Same as lanthanum.

59—*Praseodymium*—The short-period activity (5 minutes) is insensitive to water. Irradiating under water we have found a new water-sensitive activity decaying with a period of 19 hours; half-value thickness of the corresponding  $\beta$ -rays  $0.12 \text{ gm/cm}^2 \text{ Al}$  (both measured electrometrically).

64—*Gadolinium*—We irradiated under water a very pure sample of gadolinium oxide kindly given to us, together with the other rare earths, by Professor L. Rolla. We found an activity, decaying with a period of 8 hours.

73—*Tantalum*—Only a dubious activity was found after 12 hours' irradiation under water with 500 millicuries.

74—*Tungsten*—Metallic tungsten was irradiated under water and showed an activity ( $\alpha = 15$ ) decaying with a period of about 1 day.\*

\* Cf. McLennan, Grimmett, and Read, 'Nature,' vol. 135, p. 147 (1935).



We irradiated tungstic anhydride, dissolved it in caustic soda and then added and separated tantalum pentoxide which was found to be inactive. To the tungstic solution we added a nitric rhenium solution and precipitated the tungstic anhydride adding hydrochloric acid. The precipitate carried the activity, while the rhenium, precipitated from the filtrate as sulphide, was inactive. As we have no hafnium, we have made the following experiment in order to exclude an isotope of this element as carrier of the activity. From a solution of irradiated tungstic anhydride in ammonia, we precipitated zirconium hydroxide. The precipitate was inactive. We conclude that the activity of tungsten is probably due to an isotope of this element.

75—*Rhenium*—We irradiated pure metallic rhenium under water; its activity is enhanced by water and decays with a period of about 20 hours. The half-value thickness of the electrons is  $0.12 \text{ gm/cm}^2 \text{ Al}$ . The activity is probably carried by an isotope of rhenium. Irradiated rhenium was dissolved in nitric acid; we added tantalum and tungsten and separated them as tantalum pentoxide and tungstic anhydride. Both were inactive, while rhenium conserved the activity.

77—*Iridium*—The activity induced in this element is strongly sensitive to water. Period and half-value thickness of the  $\beta$ -rays have been measured in the ionization chamber; period 19 hours, half-value thickness  $0.12 \text{ gm/cm}^2 \text{ Al}$ . To the strong activation of iridium corresponds a strong absorption of the slow neutrons; half-value thickness  $0.3 \text{ gm/cm}^2$ ; the absorption is accompanied by the emission of  $\gamma$ -rays.

78—*Platinum*—Very pure metallic platinum (Heraeus 4th purity standard) irradiated under water showed an activity decaying with a period of about 50 minutes. McLennan, Grimmer, and Read (*loc. cit.*) record a period of 36 minutes.

79—*Gold*—The activity of this element is sensitive to water; its period has been measured electrometrically and is 2.7 days. The  $\beta$ -rays were magnetically deflected and found to be negative. They have a very small penetrating power: half-value thickness  $0.04 \text{ gm/cm}^2 \text{ Al}$ .\*

80—*Mercury*—No activity was found after strong irradiation. This element absorbs intensely the slow neutrons, half-value thickness  $0.2 \text{ gm/cm}^2$ .  $\gamma$ -rays are emitted during the absorption.

81—*Thallium*—No activity was found after strong irradiation.

82—*Lead*—The same as thallium.

\* Gold bombarded with slow neutrons emits a strong  $\gamma$ -radiation.

83—*Bismuth*—The same as thallium.

90—*Thorium*—The 1-minute and 24-minute (measured electrometrically) periods are scarcely sensitive to water.

92—*Uranium*—We have also studied the influence of hydrogenated substances on the induced activities of this element. (Periods 15 seconds, 40 seconds, 13 minutes, 100 minutes.) The result was that while the activities corresponding to the first, third and fourth period are slightly increased by water, no increase was found for the activity corresponding to the 40-second period. We have measured the increase in activity for the 15-second, 13-minute, and 100-minute activities. We have found for all these  $\alpha =$  about 1.6. For the 15-second activity the measurement was made with the counters and, owing to the shortness of the period, is not very exact. The  $\alpha$  values for the other two activities have been measured in the ionization chamber, special care being taken in order to verify that the  $\alpha$  values for these two periods are equal. For this we compared three decay curves of the activity obtained by irradiating the same amount of uranium oxide for 14 hours, once in air placing the uranium around the source inside a test tube; once in the same geometrical disposition surrounding the test tube with paraffin, and once inside a large mass of paraffin placing the source at about 5 cm from the test tube containing the uranium. In all these tests we found that the decay curves were proportional, *i.e.*, showed the same ratio of the two activities. We think, therefore, that the test on the identity of the sensitivities to water for these two periods is rather more accurate than the absolute value of the sensitivity coefficient and also than the identity in sensitivity coefficient with the 15-second activity. It is now evident that all the active products arising from the same primary process must have the same sensitivity coefficient. We conclude, therefore, that the 40-second activity is due to an independent primary process while the other three activities are probably due to the same primary process. This conclusion is limited by the possibility of a chance coincidence of the sensitivity coefficients within the rather wide limits of our precision. In this assumption the three activities could either be chain products (the short-period activities being parents of the long-period activities) or their relations could be complicated by branching phenomena. A certain amount of evidence in favour of the former assumption, at least as regards the 13-minute and the 100-minute periods, is given by the following test. We have measured on the electrometer the decay curves of a thick layer of irradiated uranium; these curves analysed in exponentials with the periods 13 minutes and 100 minutes show that the initial activities are in the ratio of about

100:45. The half-value thickness for the  $\beta$ -rays of the 13-minute activity is  $0.14 \text{ gm/cm}^2 \text{ Al}$ ; for the longer period it could not be measured with any accuracy, but is definitely less than the former value and probably about a half. These results are consistent with the assumption that the number of disintegrations for the 13-minute and the 100-minute periods is equal.

In our former paper we gave some chemical evidence which seemed to indicate that the carriers of the 13- and the 100-minute activities were not isotopes of any of the known heaviest elements, and that they were probably due to transuranic elements. Our point of view has in the meantime been criticized by von Grosse and Agruss,\* who, although never having experimented on activated uranium, deduced from our chemical tests the opposite conclusion, that these activities were due to isotopes of protactinium. We have therefore performed some new chemical experiments on the behaviour of these activities.

The precipitation of the activity with a sulphide was repeated, precipitating several metals (silver, copper, lead, mercury); the acidity of the solution (hydrochloric acid) was about 20%; sometimes slightly varied in order to facilitate the precipitation of the sulphide of the metal used. The yield in activity of the precipitate was generally good—about 50%—and varied according to the conditions of the precipitation. Nitric acid lowers the yield of the reaction very much. The usual high yield of the sulphide reaction is also obtained in presence of a hydrofluoric solution of tantalum. We also made a test in order to see whether the induced activities presented a reaction which is given by von Grosse as the most characteristic of protoactinium.

We dissolved in a 25% hydrochloric acid solution uranium oxide which had been purified and irradiated; we added to the solution zirconium nitrate and phosphoric acid; the precipitate of zirconium phosphate was inactive. After the separation of zirconium we precipitated a sulphide from the filtered solution, and collected the activity in the sulphide with the usual yield. According to von Grosse and Agruss, this reaction must be considered a proof of the non-identity of the carrier of the activity with a protactinium isotope.

By different chemical experiments, Hahn and Meitner† conclude also that the 13- and 100-minute activities are very probably due to transuranic elements; we have repeated some of their experiments finding the same results.

\* 'Phys. Rev.,' vol. 46, p. 241 (1934).

† 'Naturwiss.,' vol. 23, p. 37 (1935).

The two activities have beyond doubt a similar chemical behaviour. Some slight evidence of a possible separation was obtained only in the following experiment. Carefully purified uranium oxide was irradiated and dissolved in hydrochloric acid. The solution was poured into an ammonium carbonate solution until the uranium precipitate was completely dissolved again. Adding lead or manganese nitrate, we collected the precipitate of carbonates and found it to carry a fraction of the 13- and 100-minute activities. In the filtrate we precipitated copper sulphide, and this also carried a fraction of the activities. It seems that the ratio of the two activities was somewhat different in the two precipitates, the 13-minute activity being more abundant in the sulphide precipitate.

Through these experiments our hypothesis that the 13-minute and 100-minute induced activities of uranium are due to transuranic elements seems to receive further support. The simplest interpretation consistent with the known facts is to assume that the 15-second, 13-minute and 100-minute activities are chain products, probably with atomic number 92, 93 and 94 respectively and atomic weight 239.

We express our warmest thanks to Professor G. C. Trabacchi who supplied us with the radon sources. Our thanks are due also to Professor L. Rolla who placed at our disposal some rare earths, to Dr. G. Fea for help in the experiments and to the Consiglio Nazionale delle Ricerche for making a grant.

#### § 12—TABULAR SUMMARY

The main results of radioactivity induced by neutron bombardment are summarized in the table. Column 1 contains the atomic numbers and symbols of the elements investigated. Column 2 gives the isotopic constitution; numbers in bold type refer to isotopes which represent more than 20% of the element. Column 3 gives the observed half-value periods in order of increasing magnitude. Column 4 gives the half-value thickness of the  $\beta$ -rays in gm/cm<sup>2</sup> of aluminium; the mean energy of the  $\beta$ -rays in millions volts might be very roughly obtained by multiplying the figure of column 4 by 8. Column 5 indicates whether  $\gamma$ -rays have been observed to accompany the  $\beta$ -disintegration. Column 6 indicates the carrier of the activity. Column 7 gives the sensitivity of the activation to hydrogenated substances; this is given either by the numerical value of the sensitivity coefficient (for definition see § 1; sensitivity coefficient 1 means that the activation is not enhanced by hydrogenated substances) or by the letter *a*, which means that the activation is increased by hydrogenated substances, but the sensitivity

Element	Isotopes	Half-periods	Half-value thickness gm/cm <sup>2</sup> Al	γ-rays	Sensitivity to hydrogen	Carrier of the activity	δ gm/cm <sup>2</sup>
1 H	1, 2, 3	—					
2 He	3, 4	—					
3 Li	6, 7	—					0.05
4 Be	9	—					>3
5 B	10, 11	—					0.004
6 C	12, 13	—					>3
7 N	14, 15	—					>0.5
8 O	16, 17, 18	—					>3
9 F	19	9 s; 40 s	0.24; —	yes	1; 1	<sup>16</sup> N (?)	>3
10 Ne	20, 21, 22	—					
11 Na	23	40 s; 15 h	—; 0.12	yes	1; <i>a</i>	<sup>22</sup> Ne (?) <sup>24</sup> Na	>4
12 Mg	24, 25, 26	40 s; 10 m; 15 h	—; 0.07; 0.12	yes	1; <i>a</i>	<sup>23</sup> Ne (?) <sup>27</sup> Mg; <sup>24</sup> Na	>0.5
13 Al	27	2.3 m; 10 m; 15 h	0.16; 0.07; 0.12	yes	<i>a</i> ; 1	<sup>28</sup> Al; <sup>27</sup> Mg; <sup>24</sup> Na	>7
14 Si	28, 29, 30	2.3 m; 2.4 h	0.16; —	yes	1; <i>a</i>	<sup>28</sup> Al; <sup>31</sup> Si	>5
15 P	31	2.3 m; 2.4 h	0.16; 0.15	yes	1; —	<sup>28</sup> Al; <sup>31</sup> Si	>3
16 S	32, 33, 34	14 d	0.10	—	—	<sup>32</sup> P	>2
17 Cl	35, 37	35 m; 14 d	—; 0.10	—	<i>a</i> ; —	Cl; <sup>32</sup> P	0.3
18 A	36, 38, 40	—					
19 K	39, 41	16 h			<i>a</i>	<sup>42</sup> K	>1
20 Ca	40, 42, 43, 44	—					>3
21 Sc	45	16 h					
22 Ti	46, 47, 48, 49, 50	3 m					
23 V	51	3.75 m	0.17	yes	40	<sup>52</sup> V	>2
24 Cr	50, 52, 53, 54	3.75 m	0.17	yes	1	<sup>52</sup> V	>1
25 Mn	55	3.75 m; 2.5 h	0.17; 0.14	yes	1; 23	<sup>52</sup> V; <sup>56</sup> Mn	>2
26 Fe	54, 56	2.5 h	0.14	yes	1	<sup>56</sup> Mn	8
27 Co	59	2.5 h	0.14	yes	—	<sup>56</sup> Mn	0.7



Element	Isotopes	Half-periods	Half-value thickness gm/cm <sup>2</sup> Al	$\gamma$ -rays	Sensitivity to hydrogen	Carrier of the activity	$\delta$ gm/cm <sup>2</sup>
54 Xe	124, 126, 128, <b>129</b> , 130, <b>131</b> , <b>132</b> , 134, 136 .....						
55 Cs	<b>133</b> .....	1.5 h (?) (?)					
56 Ba	135, 136, 137, <b>138</b> .....	3 m; 80 m			1; 8	—; Ba	>3
57 La	<b>139</b> .....	—					
58 Ce	<b>140</b> , 142 .....	—					
59 Pr	<b>141</b> .....	5 m; 19 h			1; a		1.5
60 Nd	<b>142</b> , 143, <b>144</b> , 145, 146. ...	1 h					
61							
62 Sm	144, 147, 148, 149, 150, <b>152</b> , <b>154</b> .....	40 m					
63 Eu	<b>151</b> , <b>153</b> .....						
64 Gd	<b>155</b> , <b>156</b> , 157, <b>158</b> , 160. ...	8 h			a		
65 Tb	<b>159</b> .....						
66 Dy	<b>161</b> , <b>162</b> , <b>163</b> , <b>164</b> .....						
67 Ho	<b>165</b> .....						
68 Er	<b>166</b> , <b>167</b> , <b>168</b> , 170 .....						
69 Tu	<b>169</b> .....						
70 Yb	171, <b>172</b> , 173, <b>174</b> , 176 ..						
71 Lu	<b>175</b> .....						
72 Hf	176, 177, 178, 179, 180. ...						
73 Ta	<b>181</b> .....	?					
74 W	<b>182</b> , 183, <b>184</b> , <b>186</b> .....	1 d			15	W	
75 Re	<b>185</b> , <b>187</b> .....	20 h			a	Re	
76 Os	<b>186</b> , 187, 188, 189, <b>190</b> , <b>192</b>	—					
77 Ir		19 h			a	Ir	0.3



## Artificial Radioactivity

78 Pt		50 m				
79 Au		2·7 d			Au	2
80 Hg	196, 197, 198, 199, <b>200</b> , 201, <b>202</b> , 203, 204 .....	— — — —	0·04	yes	<i>a</i> <i>a</i>	0·2 > 6
81 Tl	<b>203</b> , <b>205</b> .....	—				
82 Pb	203, 204, 205, <b>206</b> , 207, <b>208</b> , 209, 210 .....	— —				> 10
83 Bi	<b>209</b> .....	—			~1; ~1	
90 Th	<b>232</b> .....	1 m; 24 m				
92 U	<b>238</b> .....	15s; 40s; 13 m 100 m	—; —; 0·14 0·07	yes	1·6; ~1; 1·6 1·6	See § 11

coefficient has not been measured. Column 8 gives the half-value thickness for the absorption of the slow neutrons.

In a few cases the periods of products, known from chemical evidence to be equal, but obtained by bombardment of different atoms are given as equal, even when measured very accurately only in one case. The same holds for some half-value thicknesses of  $\beta$ -rays.

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## The Theoretical Determination of the Lift Coefficient for a Thin Elliptic Cylinder

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(Communicated by G. I. Taylor, F.R.S.—Received March 6, 1935)

### 1—INTRODUCTION

This paper is concerned with the calculation of the lift coefficient of an infinitely long, thin elliptic cylinder in a stream of viscous fluid moving with constant velocity at infinity, in a direction inclined at some angle to the major axis. The method used is a general one and may be applied to any cylinder with a stream-line section provided the potential flow past the cylinder, for a given value of the circulation, is calculable.

It is clear from experiment that two cases arise according as the flow in the boundary layer is fully laminar, or partly laminar and partly turbulent. It appears that experimental conditions have not yet been obtained capable of producing a boundary layer turbulent right from the forward stagnation point. Mathematically, however, the discussion of this latter problem is a necessary preliminary to the one in which both laminar and turbulent portions are present.

We shall, therefore, discuss the three cases in which the boundary layer (i) is entirely laminar; (ii) is entirely turbulent; (iii) contains both laminar and turbulent portions.

Joukowski has, of course, given an approximate solution of the problem indicated for the case when the section of the cylinder considered has a sharp trailing edge. The methods used in the present discussion are not dependent on such a salient point existing in the section of the cylinder, and indeed will give much more accurate results than Joukowski's method where that is applicable.