Artificial Radioactivity produced by Neutron Bombardment

By E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti, and E. Segrè

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I—Introduction

This paper aims at giving a fuller account of experiments made in the Physical Laboratory of the University of Rome, on new radio-elements produced by neutron bombardment. Preliminary results have already been announced in short communications.*

Curie and Joliot† first discovered that the product atom of an artificial disintegration need not always correspond to a stable isotope, but could also disintegrate with a relatively long mean life with emission of light particles. As bombarding particles they used α-particles from polonium, and found that the light particle emitted was generally a positron. Similar results were obtained on several elements by other experimenters using α-particles, and artificially accelerated protons and deutons.‡

The use of charged particles for the bombardment, limits the possibility of an activation only to light elements. Indeed, only about ten elements up to the atomic number 15 could be activated by these methods.

It seemed therefore convenient to try the effect of a neutron bombardment, as these particles can reach the nucleus even of the heaviest elements. Available neutron sources are, of course, much less intense than α-particles or proton or deuton sources. But it was reasonable to assume that this factor would be partly compensated by the higher efficiency of neutrons in producing disintegrations. Indeed, experiment showed that more than forty elements out of about sixty investigated could be activated by this method.

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* Fermi, 'Ric. Scient.,' vol. 1, pp. 283, 330 (1934); Amaldi, D'Agostino, Fermi, Rasetti and Segrè, 'Ric. Scient.,' vol. 1, pp. 452, 652, 21 (1934); Fermi, Rasetti and D'Agostino, 'Ric. Scient.,' vol. 1, pp. 533 (1934); Fermi, 'Nature,' vol. 133, pp. 757, 898 (1934). See also Fermi, 'Nuovo Cim.,' vol. 11, p. 429 (1934); Amaldi, Fermi, Rasetti and Segrè, 'Nuovo Cim.,' vol. 11, p. 442 (1934); Amaldi and Segrè, 'Nuovo Cim.,' vol. 11, p. 452 (1934); D'Agostino 'Gazz. Chim. Ital.,' in press (1934).

† 'C.R. Acad. Sci. Paris,' vol. 198, pp. 254, 561 (1934).

‡ Frisch, 'Nature,' vol. 133, p. 721 (1934); Wertenstein, 'Nature,' vol. 133, p. 564 (1934); Cockcroft, Gilbert and Walton, 'Nature,' vol. 133, p. 328 (1934); Neddermeyer and Anderson, 'Phys. Rev.,' vol. 45, p. 498 (1934); Lauritsen, Crane and Harper, 'Science,' vol. 79, p. 234 (1934).

2—The Experimental Method

The neutron source consisted of a sealed glass tube about 6 mm in diameter and 15 mm in length, containing beryllium powder and radon in amounts up to 800 millicuries. According to the ordinarily assumed yield of neutrons from beryllium, the number of neutrons emitted by this source ought to be of the order of 1000 neutrons per second per millicurie. These neutrons are distributed over a very wide range of energies from zero up to 7 or 8 million volts, besides a very small percentage having energies about twice as high as this limit.

The neutrons are mixed with a very intensive γ-radiation. This does not, however, produce any inconvenience, as the induced activity is tested after irradiation, and it was shown that radon without beryllium produced no effect. The neutrons from beryllium are accompanied by a γ-radiation harder than any emitted by the radon products (5 to 6 million volts—about one γ-quantum per neutron). It seems, however, most unlikely that the observed effects are in any way connected with this γ-radiation, as a γ-radiation of enormously greater intensity and only slightly lower energy produces no effect.

The emission of electrons from the activated substances was tested with Geiger-Müller counters about 5 cm in length and 1·4 cm diameter. The walls of the counter were of thin aluminium foil, 0·1 to 0·2 mm in thickness. The applied voltage ranged between 1000 and 1500 volts. The amplified impulses were counted on a mechanical meter worked by a thyratron.

The substances to be investigated were generally put into form of cylinders, which could be fitted round the counter in order to minimize the loss in intensity through geometrical factors. During irradiation the material was located as close as possible round the source. Substances which had to be treated chemically after irradiation were often irradiated as concentrated water solutions in a test tube.

The decay curves of the induced activity were for many elements simple exponentials. Sometimes they could be analysed into two or more exponentials ; it was then convenient to irradiate the substance for different lengths of time in order to activate the various components with different intensity. The existence of several mean lives is, sometimes, certainly due to different isotopic constituents of the element ; when a single isotope is present it may be attributed to alternative processes of disintegration, and sometimes (uranium) to a chain of disintegrations. The intensity of activation varies within a wide range among the different elements.   In some the effect is hardly measurable,

the number of impulses produced by the irradiated substance being of the order of magnitude of the number of spontaneous impulses in the counter. In others the activation is so strong that when the substance is placed too near the counter the number of impulses is of the order of some thousands per minute, so that they cannot be counted because of lack of resolving power.

No accurate measurement of the intensity of activation of the different elements was carried out, as it would require experimenting in well-defined geometrical condition, and a knowledge of the efficiency of our counters in counting electrons, and of the absorption in the substance and in the aluminium foil. However, a very rough evaluation of these factors was made, and for some elements a number expressing the intensity of activation (i) is given. This intensity is defined as the number of disintegrations per second which take place in 1 gm of the element, placed at the distance of 1 cm from a neutron source consisting of one millicurie of radon (in equilibrium with its decay products) and beryllium powder. The substance was always irradiated until saturation of the active product was reached. The efficiency of our counters (including absorption in the aluminium foil and geometrical factors) was about 1/20, as determined by the measurement of the impulses from known quantities of potassium and uranium.

From this number expressing the intensity it is easy to obtain the cross-section for the activating neutron impact, if the number of neutrons emitted per second by a one millicurie source is known. Assuming this number to be 1000, one finds immediately the cross-section

$$\sigma = 2 \cdot 10^{-26}\, i \, . \, A,$$

A being the atomic weight of the element.

In order to be able to discuss the nuclear reaction giving rise to the active element, it is essential to identify it chemically. It is reasonable to assume that the atomic number of the active element should be close to the atomic number Z of the bombarded element. As the amount of the active substance is exceedingly small (in the most favourable cases about 10^9 atoms), there is no hope of separating it by ordinary methods. The irradiated substance was therefore dissolved, and small amounts of the inactive elements, which are suspected of being isotopic with the active product, were added. These added elements and the irradiated element were then chemically separated from each other, and separately tested for activity. It is generally found that the activity follows definitely one element. The active product can then be considered as identified with this element.

A preliminary investigation of the penetrating power of the β-rays of the new radio-elements has been carried out. For this purpose counters of the standard type were used, and the substance, instead of being put quite close to the counter, was shaped in the form of a cylinder of inner diameter somewhat larger than the diameter of the counter in order to allow cylindrical aluminium screens of different thicknesses to be interposed. In this way absorption curves of a more or less exponential type were obtained. As the geometrical conditions of this absorption measure are different from the standard ones, and moreover, the number of impulses instead of the total ionization is computed, we checked the method by measuring the absorption coefficients for known radioactive substances; as expected, we found a difference (about 20%). The data are corrected for this factor.

In several cases the absorption by 2 mm of lead was not complete; this was assumed as a proof of the existence of a γ-radiation.

It was very important to determine whether the emitted particles were positive or negative electrons. Owing to the weakness of the radiation it seemed convenient to use for this purpose Thibaud's* method of the inhomogeneous magnetic field. Even by this intensive arrangement this investigation had to be limited to elements which could be strongly activated (Al, Si, P, S, Cr, As, Br, Rh, Ag, I, Ir, U). In every case only negative electrons were observed. This, however, does not exclude that a small percentage (up to about 15%) of the emitted particles might be positrons.

For a few very strongly activated elements the emitted electrons could be also photographed in a Wilson chamber.

3—*Experimental Results*

The elements investigated are here arranged in order of atomic number; a summary of the results is to be found in a table at the end of the paper.

1—*Hydrogen*—Shows no effect when water is irradiated 14 hours with a 670-millicuries source.

3—*Lithium*—The hydroxide irradiated 14 hours with 750 millicuries is inactive.

4—*Beryllium*—Shows an extremely weak activity which might well be due to impurities.

5—*Boron*—Same as beryllium.

* 'C.R. Acad. Sci. Paris,' vol. 197, p. 447 (1933).

6—*Carbon*—Paraffin irradiated 15 hours with 220 millicuries is inactive.

7—*Nitrogen*—Guanidine carbonate (about 35% N) irradiated 14 hours with 500 millicuries is inactive.

8—*Oxygen*—No activity : see hydrogen.

9—*Fluorine*—This element irradiated as calcium fluoride can be strongly activated ($i = 0 \cdot 7$). As calcium proves to be inactive, the effect is due to fluorine. The activity decreases with a very short half period, about 9 seconds. No chemical separation was possible in this case. However, as it is known that fluorine disintegrates under neutron bombardment with emission of an α-particle, the active nucleus is probably N^{16}. This unstable isotope goes over to stable O^{16} with emission of an electron. The remarkable stability of the latter nucleus agrees with the observed very high energy of the β-rays ; the intensity reduces to half value in $0 \cdot 24$ gm/cm^2 Al. This and all the following absorption data are given for aluminium.

11—*Sodium*—This element has been irradiated as carbonate. Sodium shows a fairly strong activation, decreasing with a period of about 40 seconds.

12—*Magnesium*—This element can be fairly strongly activated, and the decay curves show the existence of two periods, of about 40 seconds and 15 hours. Half-value thickness for the long period $0 \cdot 06$ gm/cm^2.

The active element decaying with the 15 hours' period could be chemically separated. The irradiated magnesium was dissolved, and a sodium salt was added. The magnesium was then precipitated as phosphate and found to be inactive, while the sodium which remains in the solution carries the activity. The active atom is thus proved not to be an isotope of magnesium, and as neon also can be excluded, we assume it to be an isotope of sodium, formed according to the reaction :

$$Mg_{12}{}^{24} + n_0{}^1 = Na_{11}{}^{24} + H_1{}^1.$$
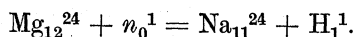
13—*Aluminium*—This element acquires a strong activity under neutron bombardment. The decay curves indicate two periods of about 12 minutes ($i = 0 \cdot 8$) and 15 hours ($i = 0 \cdot 5$). Half-value thickness respectively, $0 \cdot 07$ and $0 \cdot 06$ gm/cm^2.

The long-period activity could be chemically separated by dissolving the irradiated aluminium and adding to it small quantities of sodium and magnesium. Aluminium and magnesium are then precipitated as hydroxides and phosphates, and are found to be inactive. The solution containing sodium is then dried up, and shows an activity decaying with the 15 hours' period.

The active element is probably the same as in the former case of magnesium, as the identity of the periods and of the half-value thickness suggests. In the present case the nuclear reaction is

$$Al_{13}^{27} + n_0^1 = Na_{11}^{24} + He_2^4.$$
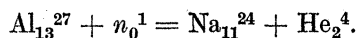
This active isotope Na^{24} then goes over to the stable isotope Mg^{24} with emission of an electron.

The active product with the 12-minute period has not been separated. However, we consider it likely to be Mg^{27}, as the other two possible cases, Al^{28} and Al^{26}, are probably to be excluded, the first because Al^{28}, as we shall next see, is a radioactive isotope with a period of 3 minutes, and the latter because Al^{26} should probably disintegrate with emission of positrons.

14—*Silicon*—Silicon is also strongly active ($i = 0.7$), and has a period probably somewhat shorter than 3 minutes. Half-value absorption thickness of the β-rays, 0.16 gm/cm².

The chemical separation of the active element has been performed by evaporating irradiated silica with hydrofluoric and sulphuric acids, after addition of aluminium and magnesium. Silicon is eliminated as fluoride, and aluminium precipitated from the residue is found to contain the activity. The active product is therefore probably an isotope of aluminium, and the nuclear reaction is

$$Si_{14}^{28} + n_0^1 = Al_{13}^{28} + H_1^1.$$
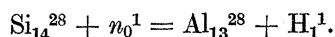
This is in accordance with the hypothesis of Curie, Joliot and Preiswerk* about the identity of this active isotope with the one which is formed by the impact of α-particles on Mg^{25}, and which has actually the same period.

15—*Phosphorus*—This element shows a strong activity ($i = 0.6$) decaying with a period of about 3 hours, and also an activity ten times less intense with a period of 3 minutes, first noticed by Curie, Joliot and Preiswerk. The half-value thickness for the β-rays of the 3 hours' product is 0.09.

The 3 hours' active product could be chemically separated. For this purpose phosphorus was irradiated as a concentrated solution of phosphoric acid. This solution was afterwards diluted with water, adding sulphuric acid and a small amount of sodium silicate. The substance is dried up to render silica insoluble, and then dissolved in water and filtered. The activity is found with the silica.

The nuclear reaction is then probably

$$P_{15}^{31} + n_0^1 = Si_{14}^{31} + H_1^1.$$
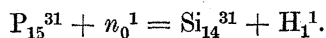
* 'C.R. Acad. Sci. Paris,' vol. 198, p. 2089 (1934).

The 3 minutes' active product has not been chemically separated. The identity of period suggests that it might be the same Al^{28} obtained from silicon.

16—*Sulphur*—Sulphur shows a fairly strong activity, decaying with a period of about 13 days (rather inaccurately measured). Half-value absorption thickness of the β-rays $0 \cdot 10$ gm/cm².

A chemical separation of the active product was carried out as follows : irradiated sulphuric acid was diluted, a trace of sodium phosphate added, and phosphorus precipitated as phosphomolibdate by addition of ammonium molibdate. The activity was found in the precipitate.

We think, in consequence, that the nuclear reaction is

$$S_{16}{}^{32} + n_0{}^1 = P_{15}{}^{32} + H_1{}^1.$$
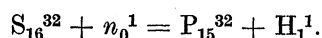
This active P^{32} is transformed by emission of an electron into the stable isotope S^{32}.

17—*Chlorine*—Half-period and penetration of the β-rays of this element are about the same as for sulphur. Intensity, $i = 0 \cdot 1$. The active substance was separated with a method quite similar to that used for sulphur. Irradiated ammonium chloride was dissolved in diluted nitric acid, and then phosphorus added and separated as in the former case. This element carried the activity as before.

The nuclear reaction which gives rise to the same active phosphorus as obtained from sulphur is

$$Cl_{17}{}^{35} + n_0{}^1 = P_{15}{}^{32} + He_2{}^4.$$
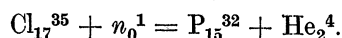
20—*Calcium*—No activity could be detected.

22—*Titanium*—A very weak effect, with a period of a few minutes, could be observed. However, it cannot be excluded that it might be due to impurities.

23—*Vanadium*—This element shows a medium activity. The half-period is about 4 minutes, and coincides within experimental error with those observed in chromium and manganese, which are due to isotopes of vanadium. This suggests the hypothesis that the active element might be V^{52}. The half-value thickness is also the same as for chromium, $0 \cdot 16$ gm/cm².

24—*Chromium*—Metallic chromium becomes fairly strongly active under neutron irradiation. The half-period is, as in the former case, about 4 minutes. Half-value absorption thickness, $0 \cdot 16$ gm/cm².

In order to identify the active element, we proceeded as follows : to irradiated ammonium chromate some sodium vanadate was added, and vanadium precipitated by addition of ammonium chloride. The activity being found

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in the precipitate, it is certainly not due to an isotope of chromium. To see whether it was an isotope of titanium, a titanium salt was added besides vanadium to the irradiated chromium compound, and titanium precipitated by hydrolysis. The precipitate showed no activity. In consequence, we consider the active substance to be probably the same isotope V^{52} as before, formed according to the nuclear reaction

$$Cr_{24}^{52} + n_0^1 = V_{23}^{52} + H_1^1.$$
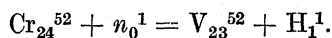
25—*Manganese*—We irradiated manganese dioxide and found a fairly intense activity decaying with two periods : about four minutes and two and a half hours. Half-value absorption thickness for the electrons of the 2·5 hours' product, 0·16 gm/cm².

The activity with the long period cannot be separated from manganese by adding chromium and vanadium and precipitating them respectively as chromate and vanadate of lead. It is therefore probably due to an isotope of manganese, Mn^{56}, the same which is extracted from irradiated iron and cobalt, as the identity of the half-life periods suggests.

In order to identify the 4-minute active element, we irradiated manganese nitrate, added a vanadium compound and precipitated vanadium as vanadate of lead. The activity was carried down in the precipitate. A similar reaction was carried out with chromium, and also in this case the precipitate was found to be active, but apparently with a definitely lower yield. The active principle is probably the isotope V^{52}, and the nuclear reaction is

$$Mn_{25}^{55} + n_0^1 = V_{23}^{52} + He_2^4.$$
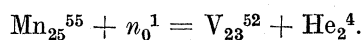
The same active vanadium is thus probably obtained from chromium, vanadium and manganese, fig. 1.

26—*Iron*—This element shows a fairly intense activity ($i = 0·05$), decaying with a period of 2·5 hours.* Half-value absorption thickness, 0·16 gm/cm². The active product can be separated as follows : the irradiated iron is dissolved in nitric acid, a small amount of a soluble manganese salt is added, and then the Mn is precipitated as MnO_2 by addition of sodium chlorate. The activity is found in the manganese precipitate. The active element is probably formed according to the reaction

$$Fe_{26}^{56} + n_0^1 = Mn_{25}^{56} + H_1^1.$$
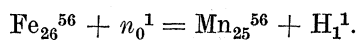
and is the same as obtained from manganese.

* See also Fleischmann, ' Naturw.,' vol. 22, p. 434 (1934).

27—*Cobalt*—Cobalt can be activated, and the decay curves show the same half-period as iron. The active product can be chemically separated with manganese by the same method as that described for iron. This suggests that it may be again the isotope Mn^{56}. In the present case the nuclear reaction would be
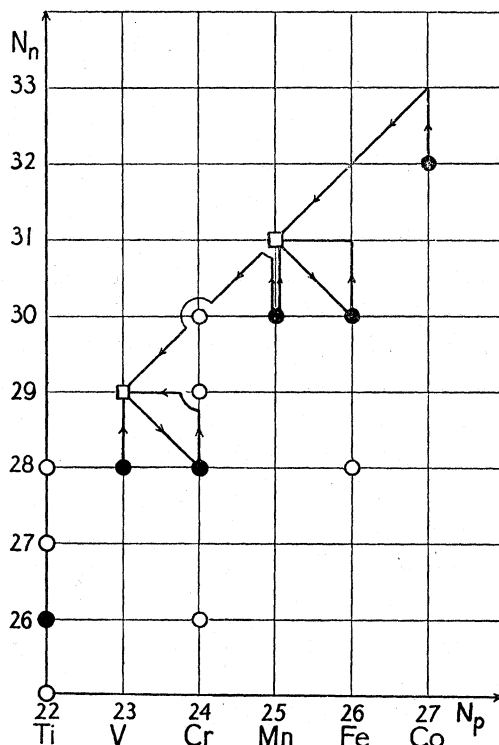
$$Co_{27}^{59} + n_0^1 = Mn_{25}^{56} + He_2^4.$$
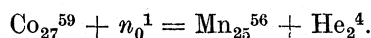
Fig. 1—The figure shows a possible scheme for the transformations which take place in the elements of atomic number 23 to 27, plotted in a proton-neutron diagram. Stable isotopes are indicated by dots (●) when representing more than 20% of the element, otherwise by circles (O). Active isotopes are indicated by small squares (□). The arrows indicate the transformations.

The active isotope Mn^{56} appears to originate from cobalt, iron and manganese (see fig. 1).

28—*Nickel*—The metal, irradiated 13 hours with 250 millicuries, was found to be inactive.

29—*Copper*—This element shows a medium activity, decaying with a period of about 6 minutes. We suggest as a possibility that the active nucleus might be the same as that which is produced from zinc, that is, an isotope of copper.

30—*Zinc*—Shows a weak activity, whose decay curve is composed of two periods, one of about 6 minutes and the other much longer and not yet measured.

The active element with the 6-minute period has been chemically separated, dissolving the irradiated metal, adding copper and nickel, and then precipitating copper as sulphide, or depositing it electrochemically on a zinc plate. In both cases copper carries the activity. The added nickel, precipitated with dimethylglyoxime, was found to be inactive. The active element is thus probably an isotope of copper.

31—*Gallium*—This element shows a moderately intense activity, decaying with a period of about 30 minutes.

33—*Arsenic*—Has a strong effect, the activity decaying with a period of about one day. Half-value absorption thickness, $0 \cdot 16$ gm/cm^2. We have tried to separate the active substance by adding gallium and germanium and precipitating the former as ferrocyanide, the latter as sulphide after elimination of the arsenic. While the precipitation of the gallium appeared to be complete, in the case of germanium we are not sure that this element had been completely separated. In both cases the substance separated was found to be inactive, suggesting as probable that the activity belongs to an isotope of arsenic. However, as the reaction with germanium gave a somewhat uncertain result, we cannot give this conclusion as proved beyond doubt.

34—*Selenium*—This element could be weakly activated. The half-period is about 35 minutes.

35—*Bromine*—Ammonium bromide was found to be strongly activated by neutron irradiation. The decay curves show two periods, of about 30 minutes and 6 hours. In order to identify the active substance we added arsenic and selenium, and separated the former as sulphide, the latter electrochemically. Both were found to be inactive. The active elements seem thus to be isotopes of bromine. As a control we added arsenic and selenium and precipitated bromine as silver bromide, and found that the precipitate carried down the activity. We suggest that the two periods may be due to two active isotopes, Br^{80} and Br^{82}, formed from the two ordinary isotopes, Br^{79} and Br^{81}.

37—*Rubidium*—Rubidium nitrate showed a very weak activity, decaying with a period of about 20 minutes.

38—*Strontium*—This element irradiated 13 hours with 400 mC was found inactive.

39—*Yttrium*—This element irradiated 30 minutes with 750 mC was found inactive.

40—*Zirconium*—Zirconium oxide showed an extremely weak activity with a period of a few minutes. It is possibly due to impurities.

42—*Molybdenum*—This element shows a very weak activity. The decay curves indicate at least two periods, one of about 15 minutes and one longer than a day.

44—*Ruthenium*—This element irradiated 40 minutes with 750 mC was found inactive.

45—*Rhodium*—Metallic rhodium could be strongly activated, and the decay curves found are consistent with the existence of two periods, one of about 50 seconds and the other of 5 minutes. Most of the activity belongs to the shorter period. Half-value thickness for the electrons of the 5 minutes' active product, $0 \cdot 10$ gm/cm².

46—*Palladium*—This element becomes moderately active under neutron bombardment. The decay curves are not very accurate, but indicate a period of the order of six hours. Half-value thickness, $0 \cdot 03$ gm/cm².

47—*Silver*—A strongly active element. The decay curves show a period of about 20 seconds and one of about 2 minutes (intensity of the latter, $i = 0 \cdot 5$; half-value thickness, $0 \cdot 08$ gm/cm²).

48—*Cadmium*—Metallic cadmium could be only weakly activated. The decay curves indicate a period slightly longer than one hour.

50—*Tin*—This metal showed an extremely weak activity, perhaps due to impurities.

51—*Antimony*—The same as tin.

52—*Tellurium*—Shows a weak activity decaying with a period of about half an hour.

53—*Iodine*—We irradiated both the element and ammonium iodide. Both showed a strong activity, decaying with a period of 30 minutes. Intensity, $i = 0 \cdot 4$. Half-value thickness, $0 \cdot 09$ gm/cm². The active element follows the iodine when this substance is precipitated by adding nitric acid to the solution of the irradiated ammonium iodide, to which tellurium and antimonium had been added. It also follows the same element when precipitated as silver iodide in presence of tellurium and antimonium. It is very likely that we have here the formation of an active isotope of iodine.

55—*Cæsium*—Cæsium (tartrate) showed a very weak activity, whose period has not yet been measured with any accuracy.

56—*Barium*—We irradiated the hydroxide and found it weakly active. The curves indicate a period of about 3 minutes.

57—*Lanthanum*—This element irradiated 40 minutes with 400 mC was found inactive.

58—*Cerium*—This element irradiated 30 minutes with 400 mC was found inactive.

59—*Praseodymium*—This element, as well as the other rare earths—Nd, Sm, and Pr—were kindly given to us by Professor Rolla, of the University of Florence.   They are very pure, and in the form of oxides.

Praseodymium shows a rather weak activity.   The decay curves indicate a period of about 5 minutes, and perhaps a longer one.

60—*Neodymium*—Shows a rather weak activity.   Period about one hour.

62—*Samarium*—Also has a rather weak activity.   Period about 40 minutes.

73—*Tantalum*—This element irradiated 2·5 hours with 750 mC was found inactive.

74—*Tungsten*—This element shows an extremely weak activity of probably long period.   This might possibly be due to impurities.

75—*Rhenium*—This element irradiated 10 minutes with 750 mC was found inactive.

76—*Osmium*—This element irradiated 15 hours with 450 mC was found inactive.

77—*Iridium*—This element has a strong activity, decaying with a period of about 20 hours.   Half-value thickness, $0·13$ gm/cm^2.

In order to establish the chemical nature of the activity, we irradiated the tetrachloride, added osmium and rhenium, and separated the former by distillation of the tetroxide, and the latter as sulphide.   Both were found to be inactive.   Iridium still contained the activity, which appears to be due to an isotope of this element.

78—*Platinum*—Pure platinum from Heræus showed an extremely weak activity of short period.

79—*Gold*—We irradiated the metal, which showed a fairly intense activity. The period is about two days.   Half-value thickness $0·33$ gm/cm.^{2}

We tried to separate the active substance by dissolving the irradiated gold foil in aqua regia, adding platinum and iridium, and precipitating these elements as respectively chloroplatinates and chloroiridiates.   Both were found to be inactive, while the activity was still found in gold, thus indicating an isotope of this element as the active nucleus.   The activity remains with gold also when mercury is added and evaporated.

80—*Mercury*—This element, strongly irradiated, showed an extremely weak activity which might be due to impurities.

81—*Thallium*—The same as mercury.

82—*Lead*—This element irradiated 10 hours with 500 mC was found inactive.

83—*Bismuth*—The same as lead.

90—*Thorium*—This element has not been investigated sufficiently at present. Thorium nitrate, freed of its ordinary β-active decay products and irradiated, becomes strongly active. The decay curves indicate at least two periods, of less than 1 minute and about 15 minutes.

92—*Uranium*—We give here only the main results on this element, as its behaviour has been discussed recently elsewhere.*

Besides the half-periods of 10 seconds, 40 seconds and 13 minutes, we have identified later one more of about 1·5 hours. The intensity of the activation is of the order of 0·5 for each of these lives. We have already discussed experiments which appear to prove that the 13 minutes' active product is not isotopic with any of the elements with atomic number from 86 to 92 (emanation, ekacæsium, radium, actinium, thorium, protactinium, uranium). These experiments have been repeated under different conditions, chiefly in order to obtain a negative proof of the identity of the 13-minute product with pro-tactinium, this proof being the most difficult to establish on account of the short period of the available Pa isotope, UX_2. The manganese reaction which has already been described gives a yield of about 15% for the 13-minute pro-duct. Its yield for UX_2 depends widely upon the conditions of the reaction, and may be varied between 2% and 10%, account being taken of the natural decay of this substance. A more effective reaction for obtaining the 13-minute active product is the following : irradiated uranium nitrate is dissolved in diluted hydrochloric acid ; some rhenium nitrate is added, and then rhenium is precipitated as sulphide by addition of sodium thiosulphate. This precipitate carries about 50% of the activity; and sometimes more. The percentage of UX_1 and of UX_2 found in the rhenium precipitate varies also with the condi-tions of the reaction (particularly with the acidity), but can be made very low, probably less than 1%. It was actually possible to separate the 13-minute active product and to measure its period using uranium which had not been purified at all from UX. The 90-minute active product has apparently chemical properties very analogous to those of the 13-minute active product, as in every type of reaction they are always obtained in about the same percent-age. These activities seem, therefore, both to be due to products with atomic number higher than 92, and possibly to isotopes of a same element.

* Fermi, ' Nature,' vol. 133, p. 898 (1934).

4—*Theoretical Discussion*

We want here to discuss, from the theoretical point of view, the processes that may take place under neutron bombardment. At the present state of the nuclear theory these considerations can have only a provisional character. We can resume the empirical results of the preceding sections in the following points.

(*a*) A large percentage of elements of any atomic weight can be activated from this point of view no special difference can be noticed between light and heavy elements.

(*b*) The cross-sections for neutron impact for the elements which can be most intensely activated are of the order of the geometrical cross-section of the nucleus. This means that a large percentage of the neutrons which hit the nucleus produce an active atom.

(*c*) The active product is sometimes an isotope of the original atom (atomic number Z) ; in other cases its atomic number is lower by one or two units. In this respect there appears to be a difference between light and heavy elements. For light elements the atomic number of the active product is usually lower than Z, while in the five cases investigated for heavy, non-spontaneously radioactive elements, the active product is always an isotope of the bombarded element.

(*d*) The emitted electrons always have a negative charge, or at least no positrons could ever be detected.

There seems to be no special difficulty in explaining the general mechanism of the activation for light elements. This seems to consist usually in the capture of the impinging neutron, followed immediately by the expulsion of an α-particle or a proton. If the energy of the emitted α-particle or proton is of some million volts, it results from Gamow's theory that the time which is necessary to emit the particle is extremely short, and there is therefore a fairly high probability for the process to happen before the neutron has left the nucleus. After this process, which may last a time of the order of 10^{-20} seconds, the nucleus has been transformed into a new one having, on the average, an atomic weight higher than would belong to its nuclear charge, as the processes of absorbing a neutron and emitting an α-particle or a proton increase the neutron/proton ratio in the nucleus. This is probably the reason why an emission of negative electrons is always observed. The process of the electron emission re-establishes the correct value for neutron/proton ratio, and corresponds to the formation of a stable isotope.

As the atomic weight of the bombarded element increases, the potential barrier around the nucleus becomes an increasingly strong obstacle to the emission of heavy, positively-charged particles ; it is therefore easy to understand why processes with emission of protons and α-particles become very improbable.

The reactions whose theoretical interpretation seems to meet with difficulties are those, normally occurring among heavy elements, in which the activated atom is isotopic with the original atom. The simplest hypothesis would be to assume a capture of the impinging neutron, giving rise to an unstable isotope of the bombarded element with an atomic weight higher by one unit than before the process. This hypothesis, which would be in agreement with the observed fact of the emission of negative electrons, gives rise, however, to serious theoretical difficulties when one tries to explain how a neutron can be captured by the nucleus in a stable or quasi-stable state. It is generally admitted that a neutron is attracted by a nucleus only when its distance from the centre of the nucleus is of the order of 10^{-12} cm. It follows that a neutron of a few million volts' energy can remain in the nucleus (*i.e.*, have a strong interaction with the constituent particles of the nucleus) only for a time of the order of 10^{-21} seconds; that is, of the classical time needed to cross the nucleus. The neutron is captured if, during this time, it is able to lose its excess energy (*e.g.*, by emission of a γ-quantum). If one evaluates the probability of this emission process by the ordinary methods one finds a value much too small to account for the observed cross-sections. In order to maintain the capture hypothesis, one must then either admit that the probability of emission of a γ-quantum (or of an equivalent process, as, for example, the formation of an electron-positron pair) should be much larger than is generally assumed ; or that, for reasons that cannot be understood in the present theory, a nucleus could remain for at least 10^{-16} seconds in an energy state high enough to permit the emission of a neutron.

An alternative hypothesis is to admit that the impinging neutron is not captured, but causes the expulsion of another neutron from the nucleus. This process could be described as follows : the primary neutron loses part of its energy, bringing the nucleus into an excited state by a sort of inelastic impact. One can easily understand theoretically that these processes may take place in a large percentage of the collisions between nuclei and neutrons. If the excitation energy is large enough, a neutron can be emitted before the nucleus loses its energy by emission of a γ-quantum. The atom formed by such a process is an isotope of the original one, with atomic weight lower by one unit.

An objection which may be raised against this hypothesis is that if the number of neutrons decreases instead of increasing, it is, *a priori*, more likely that the atom, in the following disintegration, should emit a positron than a negative electron as observed. However, in the few cases investigated of heavy elements which are activated by neutron bombardment and are transformed into their isotopes, when the isotopic constitution of the neighbouring elements is known, there always exists a stable isotope of the element $Z + 1$, having atomic weight one unit less than the original element, as a possible end-product of the transformation.

One has, moreover, to bear in mind that if an unstable nucleus has energetically the possibility of emitting both an electron and a positron, the theory of the β-rays* gives *ceteris paribus* the emission of an electron as the most probable.

In conclusion, the choice between these two alternatives seems at present rather uncertain, and further experiments must be performed to test this point.

* Fermi, ' Z. Physik,' vol. 88, p. 161 (1934).

5—*Tabular Summary*

The main results of this investigation are summarized in the table. Column 1 contains the atomic number and symbol 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-periods; a heavy —— means that the activity was sought for and not found. Column 4 gives a rough evaluation of the intensity s (strong), m (medium), w (weak). Column 5 gives the average energy of the electrons in million volts; this was obtained from the absorption measurement by a rather rough extrapolation based on the absorption coefficients of ordinary β-active substances without strong γ-rays (Ra E and UX_2). Column 6 shows whether γ-rays were observed or not; a line means that γ-rays have been sought for and not observed. Column 7 gives a probable active product; for simplicity we have always assumed that the neutron is captured; if, instead, a neutron was emitted, the corresponding atomic weights should be decreased by two units. When two or more periods are present, the data of columns 4, 5, 6 and 7 refer to the different periods in their order.

Atomic number.	Isotopes.	Half-period.	Intensity.	Mean energy of β-rays in 10^6 volts.	γ-rays.	Active isotope.
1 H	1, 2	——	——			
3 Li	**6, 7**	——	——			
4 Be	**9**	?	?			
5 B	**10, 11**	?	?			
6 C	**12**, 13	——	——			
7 N	**14**, 15	——	——			
8 O	**16**, 17, 18	——	——			
9 F	**19**	9 s.	s	2	yes	N^{16} (?)
11 Na	**23**	40 s.	m			
12 Mg	**24**, 25, 26	40 s. ; 15 h.	m ; m	— ; ·5	? ; yes	— ; Na^{24}
13 Al	**27**	12 m. ; 15 h.	s ; s	·6 ; ·5	yes ; yes	— ; Na^{24}
14 Si	**28**, 29, 30	3 m.	s	1·3	yes	Al^{28}
15 P	**31**	3 m. ; 3 h.	m ; s	— ; ·7	?	Si^{31}
16 S	**32**, 33, 34	13 d.	m	·8	——	P^{32}
17 Cl	**35, 37**	13 d.	m	·8		P^{32}
20 Ca	**40**, 42, 43, 44	——	——			
22 Ti	46, 47, **48**, 49, 50	3 m.	w			
23 V	**51**	4 m.	m	1·3		V^{52} (?)
24 Cr	50, **52**, 53, 54	4 m.	m	1·3	yes	V^{52}
25 Mn	**55**	4 m. ; 150 m.	m ; m	— ; 1·3		V^{52} ; Mn^{56}
26 Fe	54, **56**	150 m.	m	1·3	yes	Mn^{56}
27 Co	**59**	150 m.	w			Mn^{56}
28 Ni	**58, 60**, 61, 62	——	——			
29 Cu	**63, 65**	6 m.	m			
30 Zn	**64, 66**, 67, 68, 70	6 m. ; ?	w ; w			Cu ; —
31 Ga	**69, 71**	30 m.	m			
33 As	**75**	1 d.	s	1·3	yes	As^{76}
34 Se	74, 76, 77, **78, 80**, 82	35 m.	w			
35 Br	**79, 81**	30 m. ; 6 h.	s ; s	— ; ·7 (?)	——	Br^{80} ; Br^{82}
37 Rb	**85**, 87	20 m.	w			
38 Sr	86, 87, **88**	——	——			
39 Y	**89**	——	——			
40 Zr	**90**, 91, **92**, 94, 96	?	w			
42 Mo	92, 94, 95, 96, 97, **98**, 100	15 m. ; (?)	w ; w			
44 Ru	96, 98, 99, 100, **101, 102**, 104	——				
45 Rh	——	50 s. ; 5 m.	s ; m	·8	——	
46 Pd	——	6 h. (?)	w	·3	——	
47 Ag	**107, 109**	20 s. ; 2 m.	s ; s	— ; ·7	——	
48 Cd	110, 111, **112**, 113, **114**, 116	70 m.	w			
50 Sn	112, 114, 115, 116, 117, **118**, 119, **120**, 121, 122, 124	——	——			

Tabular Summary—(continued).

Atomic number.	Isotopes.	Half-period.	Intensity.	Mean energy of β-rays in 10^6 volts.	γ-rays.	Active isotope.
51 Sb	**121, 123**	?	?			
52 Te	122, 123, 124, 125, **126,** (127), **128, 130**	30 m. (?)	*w*			
53 I	**127**	30 m.	*s*	·7	——	I^{128}
55 Cs	**133**	(?)	*w* (?)			
56 Ba	135, 136, 137, **138**	3 m.	*w*			
57 La	**139**	——	——			
58 Ce	**140**, 142	——	——			
59 Pr	**141**	5 m.	*w*			
60 Nd	**142**, 143, **144**, 145, 146....	1 h.	*w*			
62 Sm	144, 147, 148, 149, 150, **152**, 154	40 m.	*w*			
73 Ta	**181**	——	——			
74 W	**182**, 183, **184**, **186**...........	(?)	*w* (?)			
75 Re	**185, 187**	——	——			
76 Os	186, 187, 188, 189, **190**, **192**	——	——			
77 Ir	——	20 h.	*s*	1·1	yes	Ir
78 Pt	——	(?)	*w* (?)			
79 Au	——	2 d.	*s*	·3	——	Au
80 Hg	196, 197, 198, 199, **200**, 201, **202**, 203, 204	(?)	(?)			
81 Tl	**203, 205**	(?)	(?)			
82 Pb	203, 204, 205, **206**, **207**, **208**, 209	——	——			
83 Bi	**209**	——	——			
90 Th	**232**	1 m. (?); 15 m.	*s* ; *s*			
92 U	**238**	15 s. ; 40 s. ; 13 m. 100 m.	*s* ; *s* ; *s* ; *s*		yes	*

* See p. 495.