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PROCESS FOR THE PRODUCTION OF RADIOACTIVE SUBSTANCES

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Fig. 1.

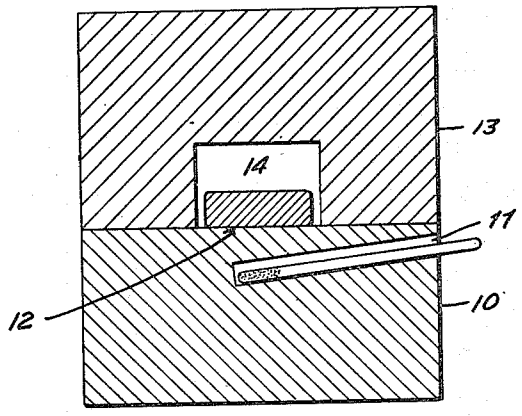
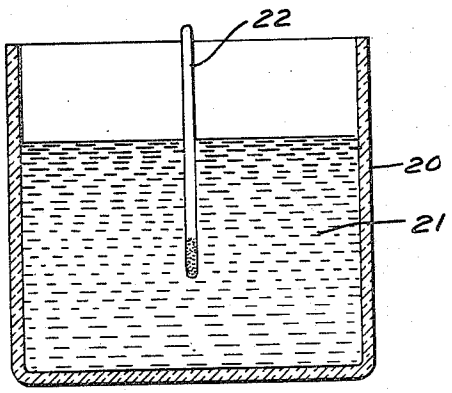


Fig. 2.



INVENTORS

ENRICO FERMI, EDOARDO AMALDI,
BRUNO PONTECORVO, FRANCO RASETTI, EMILIO SEGRE

BY

ATTORNEYS

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PROCESS FOR THE PRODUCTION OF RADIOACTIVE SUBSTANCES

Enrico Fermi, Edoardo Amaldi, Bruno Pontecorvo, Franco Rasetti, and Emilio Segre, Rome, Italy, assignors to G. M. Giannini & Co., Inc., New York, N. Y., a corporation of New York

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This invention relates to the production of isotopes of elements from other isotopes of the same or different elements by reaction with neutrons, and especially to the production of artificial radio activity by formation of unstable isotopes.

It has been known for many years that, although each chemical element has always the same atomic number or charge, it may exist in different forms having different atomic weights. These forms of the elements are referred to as isotopes.

It has also been known that the radio-active elements, by disintegration or break down occurring in their nuclei are spontaneously converted into various isotopes of other elements. Thus, for example, the radio-active element uranium may be converted into lead of atomic weight 206, while the element thorium may be converted into a different isotope of atomic weight 208.

It has long been known that such spontaneous disintegration of radio-active elements is accompanied by emission of alpha, beta, and gamma rays, that is to say, of the helium nucleus, electrons, and electromagnetic radiation of extremely short wave length.

In more recent years it has been demonstrated that isotopes of various elements could be converted into other isotopes of the same or different elements by bombardment with alpha particles, diplons, protons or gamma rays of very high energy, and that the isotopes thus produced may be unstable so as to decompose with features similar to those of the naturally radio-active bodies. That is to say, radio-active isotopes may, in this way, be artificially produced.

Accordingly, it is an object of the present invention to provide a method and apparatus by which nuclear reactions can be carried on with high efficiency and with the heavier as well as with the lighter elements. A more specific object of the invention is to provide a method and apparatus for artificially producing radio-active substances with efficiency such that their cost may be brought below that of natural radio-active materials.

Our invention is based upon the use of neutrons instead of charged particles for the bombardment and transformation of the isotopes.

All of the prior work on nuclear reactions has been done with high energy particles and every effort has been bent toward increasing the energy of the particles as the means of extending and making more efficient the nuclear reactions. We have now discovered that effort in this direction is sound only when charged particles are used which require tremendous energy to break through the potential barrier surrounding the nucleus; and that if, instead of charged particles, neutrons are used for the nuclear reactions, the greatest efficiencies are in some cases attained with low energy or "slow" neutrons, e. g., of the order of a few hundred electron volts, or even much less down to a small fraction of an electron volt.

Neutrons when produced in any ordinary manner, e. g., by the action of radon on beryllium or of polonium on beryllium or by bombardment of atomic nuclei with artificially accelerated particles, might have a very wide range of energies but high average energy. These energies range up to several million volts. It is necessary, therefore, if the greatest efficiency of reaction is to be attained, to reduce by artificial means the energy of these neutrons. We describe below a method for slowing down fast neutrons.

We have demonstrated that the absorption of slow neutrons is anomalously large as compared with that of the faster or higher energy neutrons. The simplest explanation for most cases 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 is unstable a strong induced radio-activity may be expected. This occurs, for example, with silver and iridium which go over into radio-active isotopes. In other cases it is found that no activation, or at least no strong activation, follows an anomalously large absorption. This is the case with many elements, e. g., yttrium and cadmium. In these cases the formation of a stable nucleus upon the capture of the neutrons is to be expected.

In some cases the absorption of the slow neutrons results in the emission of a relatively strong gamma-radiation with energy corresponding to the binding energy of the neutron. This gives a reliable source of very hard gamma rays, even harder than the naturally produced gamma rays, e. g., from radium.

In view of these considerations it is obviously desirable to convert as many as possible of the available neutrons into the slow or low energy condition in which they may be readily captured by the nuclei of the substance being reacted. We have found that it is possible to achieve the desired results by passing the neutron radiation against or through a screen of a suitable material.

The materials which have been found best suited to this purpose are those containing hydrogen (including all its isotopes, but the light

isotope which predominates in natural occurrence being most efficient) and especially water and the hydrocarbons, such as paraffin for example. Other materials, as for example beryllium, carbon, silicon, lead, show this effect to a lesser degree. Other materials, of which iron is an example, do not produce a similar effect to any practical extent, probably because of a relatively large absorption of the neutrons when their energy is reduced.

The increase in activity of the neutrons by such substances is apparently due to two effects both resulting from collisions of the neutrons. In the first place it is readily 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 ten impacts reduce the energy to about $\frac{1}{20,000}$ of its original value. Assuming the initial energy to be $4 \cdot 10^6$ electron volts, the energy after ten impacts would be about 200 electron volts and 20 impacts would reduce the energy of the neutron down to a value corresponding to thermal agitation. Thus the first important effect is probably the reduction of the energy of the faster neutrons by impact and the efficiency of hydrogen for this purpose is probably due to the low mass of the hydrogen nucleus. Although we refer to the nucleus, because almost the entire mass is represented by the nucleus, it will be understood that the impact for slowing down may be, and for reasons of economy ordinarily will be, with atoms, i. e., combined nuclei.

The second probable effect is the scattering and reflection of the neutrons.

Hydrogen is so much more effective than any of the other elements for reducing the energy of neutrons that it will ordinarily be used. It must not be overlooked, however, that the elements having a lesser effect offer possibilities for control of the neutron energy. Where neutrons of initially lower energy are used or where their use requires a higher energy than in the reactions with which we are here especially concerned, the less effective elements may be used singly or combined with elements of different energy reducing power.

The density of the energy reducing or scattering substance has also been found to be an important factor. This follows, likewise, from the theoretical explanation given above. If the energy reduction and scattering of the neutrons is due to impact with atomic nuclei, the probable frequency of such impacts will be directly dependent upon the number of atoms in a given space. For this reason such substances ordinarily should be used in liquid or solid (i. e., non-gaseous) form and, so far as is practicable, substances will be chosen having as high as possible a proportion of hydrogen in the molecule. Here again, the gaseous state under various pressures, and substances having less hydrogen offer the possibility for accurate control if less than the maximum slowing of neutrons is desirable.

It will be readily understood from what has been said above that the greatest effect is attained if the source of neutrons and the substance being irradiated are both surrounded by the energy reducing and scattering material. This could easily be accomplished in many cases by immersing the neutron source in a solution or emulsion of the substance being bombarded. We have illustrated diagrammatically in Figures 1 and 2 arrangements by which this may be accomplished.

In Figure 1 a cylindrical paraffin block **10** is provided with a hole **11** into which is inserted a source of neutrons, e. g., a tube containing radon and beryllium. The material being irradiated is placed above the source on the paraffin block as shown at **12** and is covered by a second paraffin block **13** having a central opening **14** to accommodate the material being irradiated. For the treatment of small amounts of materials the block **10** may be, for example, about 24 centimeters in diameter and about 14 centimeters in height with the neutron source about 2 centimeters under the upper surface. It will be observed that these dimensions give radial thickness of the material surrounding the neutron source approximately equal to the mean free path in the substance of the high energy neutrons.

Where the substances to be irradiated are soluble in or can be suspended in water or a hydrocarbon or other energy reducing or dispersing substance, etc., may be formed and the substances irradiated therein by immersing the neutron source directly into the solution etc. (See Figure 2.)

The hydrogen which serves to reduce the energy of the neutrons may also be in chemical combination with the substance being irradiated.

Figure 2 is a diagrammatic illustration of a neutron irradiating device in which the substance being irradiated is dissolved or dispersed in the energy reducing or dispersing material. A suitable vessel **20** is provided for holding the solution or dispersion **21** and into this is immersed the neutron source **22**.

Instead of the radon beryllium source, any other source of neutrons may be used, as for example neutron tubes as developed by Oliphant and as more recently developed by laboratories of the General Electric Company and the Westinghouse Electric and Manufacturing Company or cyclatrons as developed by Lawrence. In such tubes deuteron oxide (heavy water), which may be separated by known methods from naturally occurring water, is bombarded with deuterons accelerated in an electric field produced by a grid tube. The deuteron nucleus is disintegrated with the emission of neutrons.

Obviously the apparatus can be endlessly modified, the essential being the combination of the energy reducing substance near the substance being irradiated, and a suitable source of neutron radiation.

In the following we have tabulated the results of various irradiations which we have carried out:

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.^{2} This absorption is not accompanied by a γ-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,

$$^6_3\mathrm{Li} + ^1_0\mathrm{n} = ^4_2\mathrm{He} + ^3_1\mathrm{H}$$

* "Nature", vol. 135, p. 65 (1935).

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.^{2}, corresponding to a cross-section of about 3.10^{-21}cm.^{2}. No γ-rays have been found to accompany this absorption: instead of a γ-radiation in this case as well as for lithium, α-particles are emitted, as was shown by Chadwick and Goldhaber (Nature, vol. 135, 1935, p. 65) 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 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 α-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,

$$^{10}_{5}B+^{1}_{0}n=^{7}_{3}Li+^{3}_{2}He$$

6. *Carbon.*—No activity; see hydrogen.

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 hydrogen containing substances.

11. *Sodium.*—This element has two activities: one of these (period 40 seconds) is not sensitive to hydrogen-containing substances. A very weak activity with a long period was reported by Bjerge and Westcott. As this activity is strongly enhanced by water, we were able to measure its periods with reasonable accuracy and found it to be 15 hours. Owing to the theoretical importance of this activity, 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. We dissolved the irradiated substance in hydrochloric acid and added aluminum 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}Na. The same isotope was produced by us last year by bombarding magnesium or aluminum with neutrons. ^{24}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, 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 aluminum, which is known to be due to ^{27}Mg (see aluminium), it is very likely that it is due to the same isotope formed by the capture of a neutron by ^{26}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}Al, we assume that this activity is also due to the same isotope formed by capture of a neutron from ^{27}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}Mg formed according to the reaction,

$$^{27}_{13}Al+^{1}_{0}n=^{27}_{12}2Mg+^{1}_{1}H$$

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}Si which is obtained by irradiated phosphorus and has a period of 2–4 hours. ^{31}Si could be formed by capture of a neutron from ^{30}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}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 β-rays and found it to be 0–15 gm./cm.^{2} 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 β-rays 0/10 gm./cm.^{2} Al.

17. *Chlorine.*—Chlorine irradiated under water showed a new period of 35 minutes measured electrometrically.

Chlorine absorbs fairly strongly the slow neutrons (half-value thickness $\delta=0.3$ gm./cm.^{2}).

The process of absorption is accompanied by emission of γ-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}K, that was obtained by him by neutron bombardment of scandium, and has the same decay period.

20. *Calcium.*—No activity was found in calcium fluoride irradiated 14 hours in water with a 600 millicuries source. A very weak activity sensitive to water has been demonstrated.

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 β-rays 0–17 gm./cm.² Al. The β-rays are accompanied by a γ-radiation. The activation of vanadium is strongly sensitive to hydrogenated substances.

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

25. *Manganese.*—The activity with short period (3–75 minutes) is insensitive to water. On the other hand, the activity with longer period (2–5 hours measured in the ionization chamber) is strongly enhanced by water. Half-value thickness of β-rays measured electrometrically is 0–14 gm./cm.² Al; the disintegration is accompanied by γ-rays. The 2–5-hour product is known to be an isotope of manganese. 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 maganese 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.².

27. *Cobalt.*—This element absorbs fairly strongly the slow neutrons; half-value thickness 0–7 gm./cm.². The absorption is accompanied by the emission of a γ-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. Copper absorbs the slow neutrons with a half-value thickness of about 3 gm./cm.²; this absorption is accompanied by a weak γ-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.

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.

31. *Gallium.*—The 20-minute period (measured electrometrically) is not very sensitive to water. Half-value thickness of the corresponding β-rays is 0.17 gm./cm.² 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 γ-radiation; it decays with a period of 23 hours (measured electrometrically).

33. *Arsenic.*—The activity of this element is strongly sensitive to water. We have measured electrometrically its period (26 hours) and its half-value thickness of the β-rays (0–16 gm./cm.² Al).

34. *Selenium.*—The activity of this element (period 35 minutes) is sensitive to water. 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 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 periods have been measured electrometrically; they are 18 minutes and 4–2 hours. The half-value thickness of the γ-rays is for both activities 0–12 gm./cm.² Al, and both are accompanied by γ-rays.

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 $\delta=0.015$ gm./cm.²). This absorption is accompanied by γ-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. Period and half-value thickness of the β-rays have been determined electro-

metrically (44 seconds; 0–15 gm./cm.² 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 γ-radiation. Rhodium absorbs fairly strongly the slow neutrons (half-value thickness 0–3 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.

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. To the strong activation of this element corresponds a considerable absorption for slow neutrons (half-value thickness δ=1–2 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 dimethylglioxime 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 gm./cm.².) The corresponding cross-section is the largest as yet found for slow neutrons (σ=10^{20}cm.²). The absorption is accompanied by an intensive γ-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) has an activity highly sensitive to water. 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 gm./cm.² 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 δ=0–3 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 β-rays is 0–09 gm./cm.² Al. The following chemical test indicates that the carrier of this activity is probably an isotope of antimony. We dissolved metallic 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.

53. *Iodine.*—Period and half-value thickness of the β-rays were determined electrometrically: period 25 minutes; half-value thickness 0–11 gm./cm.² Al. The activation is moderately sensitive to water.

56. *Barium.*—A new activity sensitive to water 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 β-rays 0–12 gm./cm.² Al (both measured electrometrically).

72. Hafnium v. Hevesy has found an activity having a long period of several months which is sensitive to water.

64. *Gadolinium.*—We irradiated under water a very pure sample of gadolinium oxide. 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 decaying with a period of about 1 day.

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 gm./cm.² 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 β-rays have been measured in the ionization chamber; period 19 hours, half-value thickness 0–12 gm./cm.² Al. To the strong activation of iridium corresponds a strong absorption of the slow neutrons; half-value thickness 0–3 gm./cm.²; the absorption is accompanied by the emission of γ-rays.

78. *Platinum.*—Very pure metallic platinum irradiated under water showed an activity decaying with a period of about 50 minutes.

79. *Gold.*—The activity of this element is sensitive to water; its period has been measured electrometrically and is 2–7 days. The β-rays were magnetically deflected and found to be negative. They have a very small penetrating power: half-value thickness 0–04 gm./cm.² Al. Strong γ-radiation is omitted during bombardment with slow neutrons.

80. *Mercury.*—No activity was found after strong irradiation. This element absorbs intensely the slow neutrons, half-value thickness 0–2 gm./cm.². γ-rays are emitted during the absorption.

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

82. *Lead.*—The same as thallium.

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.

Chemical evidence seems 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.

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 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 by the non-identity of the carrier of the activity with a protoactinium isotope. The 15-second, 13-minute and 100-minute activities are probably chain products, with atomic number 92, 93 and 94 respectively and atomic weight 239.

From the above tabulation it is apparent that the increase in activities by the hydrogen containing substances, etc., is particularly applicable to those nuclear reactions in which the neutron is captured with the formation of a heavier isotope of the same element; and the present invention makes possible numerous reactions of this type which could not be appreciably carried out without the use of our invention.

With these procedures the isotopes produced by the nuclear reactions are ordinarily mixed with other substances, although in much higher concentrations than was heretofore possible. We have utilized for the separation of these isotopes and especially of the radio-active isotopes the method of Szilard and Chalmers (Nature, vol. 134, page 462, 1934) and extended their procedure to cover other cases. This and further examples and theoretical discussion of our invention are set forth in our publications: Fermi, Amaldi, D'Agostino, Rasetti, Segre, 'Proc. Roy Soc.,' A, vol. 146, p. 483 (1934); A, vol. 149, p. 522; Fermi, Amaldi, Pontecorvo, Rasetti, 'Ric. Scient.,' vol. 2, p. 380 (1934); Fermi, Pontecorvo, Rasetti, 'Ric. Scient.,' vol. 2, p. 380 (1934); Amaldi, D'Agostino, Segre, 'Ric. Scient.,' vol. 2, p. 381 (1934); Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti, Segre, 'Ric. Scient.,' vol. 2, p. 467 (1934); vol. 1, p. 123 (1935).

Claims directed to the broad method of producing radio-active substances by a neutron reaction and to beta-emissive substances so produced are being presented in the copending application Serial No. 57,325 to Enrico Fermi, filed January 2, 1936.

Although we have herein described our invention in detail and specified particular examples of apparatus and processes and various modifications thereof, and have proposed various theoretical explanations, it is to be understood that these are not binding nor exhaustive but are intended rather for the assistance of others skilled in the art to enable them more easily to apply our invention under widely varying conditions encountered in actual practice and to change and modify the particular embodiments and examples herein described as may be necessary or desirable under such varying conditions. The theoretical statements and explanations are, of course, not conclusive and our invention is in no way dependent upon their correctness. We have found them helpful and give them for the aid of others, but our invention will be equally useful if it should prove that our theoretical conclusions are not altogether correct.

We claim:

1. The process for the production of radio-active isotopes, which comprises generating neutrons having a high average electron voltage, slowing down and scattering said neutrons by projecting them through a screen of an element

of the class consisting of hydrogen, helium, beryllium, carbon, silicon and lead which screen is of such thickness that the neutrons are slowed down to an average energy of not more than a few hundred electron volts, then passing said neutrons into a mass of an element of the groups having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92, and thereby producing from the latter element a radio-active isotope capable of emitting beta rays.

2. The process of producing radio-active isotopes which comprises generating neutrons having a high average energy, slowing down said neutrons so that they have an average energy of not more than a few hundred electron volts by projecting them through an energy-reducing screen of an element of the class consisting of hydrogen, helium, beryllium, carbon, silicon and lead, and passing said neutrons of reduced energy into a mass of an element of the group having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92 to thereby produce from the latter element a radio-active isotope capable of emitting beta rays.

3. The process of producing radio-active isotopes which comprises, generating neutrons having a high average energy, slowing down said neutrons so that they have an average energy of not more than a few hundred electron volts by projecting them through a hydrocarbon, and passing said neutrons of reduced energy into a mass of an element of the group having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92 to thereby produce from said latter element a radio-active isotope capable of emitting beta rays.

4. The process of producing radio-active isotopes which comprises, generating neutrons having a high average energy, slowing down said neutrons so that they have an average energy of not more than a few hundred electron volts by projecting them through paraffin, and passing said neutrons of reduced energy into a mass of an element of the group having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92 to thereby produce from the latter element a radio-active isotope capable of emitting beta rays.

5. The process of producing radio-active isotopes which comprises, generating neutrons having a high average energy, slowing down said neutrons so that they have an average energy of not more than a few hundred electron volts by projecting them through water, and passing said neutrons of reduced energy into a mass of an element of the group having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92 to thereby produce from the latter element a radio-active isotope capable of emitting beta rays.

6. The process of producing a radio-active isotope from an element of the group having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92, comprising the steps distributing the element throughout an energy-reducing screen of an element of the class consisting of hydrogen, helium, beryllium, carbon, silicon and lead, generating neutrons having a high average energy, and projecting the high-energy neutrons through said energy-reducing screen to slow down the neutrons to an average energy of not more than a few hundred electron volts and thereby produce from said first element by the reaction of the neutrons of reduced energy a radio-active isotope capable of emitting beta rays.

7. The process of producing a radio-active isotope from an element of the group having atomic numbers 11, 12, 13, 14, 17, 19, 23, 25, 29, 31, 33, 34, 35, 43, 47, 48, 49, 51, 52, 53, 56, 64, 72, 74, 75, 77, 78, 79 and 92, comprising the steps distributing the element throughout an energy-reducing screen of an element of the class consisting of hydrogen, helium, beryllium, carbon, silicon and lead, generating at a point substantially surrounded by said screen neutrons having a high average energy, and projecting the high-energy neutrons through said energy-reducing screen to slow down the neutrons to an average energy of not more than a few hundred electron volts and thereby produce from said first element by the reaction of the neutrons of reduced energy a radio-active isotope capable of emitting beta rays.

EMILIO SEGRE.
ENRICO FERMI.
EDOARDO AMALDI.
BRUNO PONTECORVO.
FRANCO RASETTI.